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Oxygen Separation Process using Ceramic-Based Membrane: A Review

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ABSTRACT

Membrane technology is known as a cheaper alternative approach that capable to yield high purity oxygen compared to the conventional separation technologies which are pressure swing adsorption (PSA) and cryogenic distillation. The status of advanced ceramic-based membranes in the application of separating oxygen at high temperature is discussed in this paper, including arising issues related to performance in selectivity and separation of membranes. This review paper covers various approaches to solve the problems and issues of the ceramic-based membrane, also, its expected tendency in separating oxygen from the air.

1. Introduction

Oxygen contributes about 21 volume percentage towards the air. Oxygen is one of the gases that extensively used globally in which it is in the top five rankings for chemicals production as it involves in various applications for most industrial sectors [1]. The oxygen separation is a very significant process in the industry in which the annual oxygen production is about hundred million tonnes [2]. For the reason that every large-scale and clean energy technologies need feed gas (oxygen), oxygen gas is expected to receive increasing demands soon [3,4]. The high demand for operations of the chemical process requires a great capacity of oxygen such as the process of oxygen-blown gasification and oxyfuel combustion. Oxygen is required in converting natural gas and coal to form intermediate synthesis gas which will be processed for electricity and chemical products including fuel

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transportation [5]. There are two conventional technologies in separating desired gas from air called cryogenic distillation and non-cryogenic distillation. By comparing both technologies, cryogenic distillation is more preferable for the production of oxygen in a large quantity at ultra-low temperature as illustrated in Figure 1 [6]. Whereas, non-cryogenic distillations are either pressure swing adsorption (PSA) involving separating air at ambient temperatures by passing through molecular sieve adsorptions, or membrane technology which involves gas separation via polymeric membranes [7]. Nowadays, membrane technology is a leading separation method as this technology is capable of fast process operation with energy-efficient and involves no chemical additives and changes of phase [42]. This technology has been implemented in various sectors, for example, chemical and food industries [43,44]. The implementation of membrane is not only because it is economically excellent about 30% compared to other technologies, but also due to its ability for the reduction of waste disposal by allowing material to be recycled and recovered as required by regulation of environment [41].

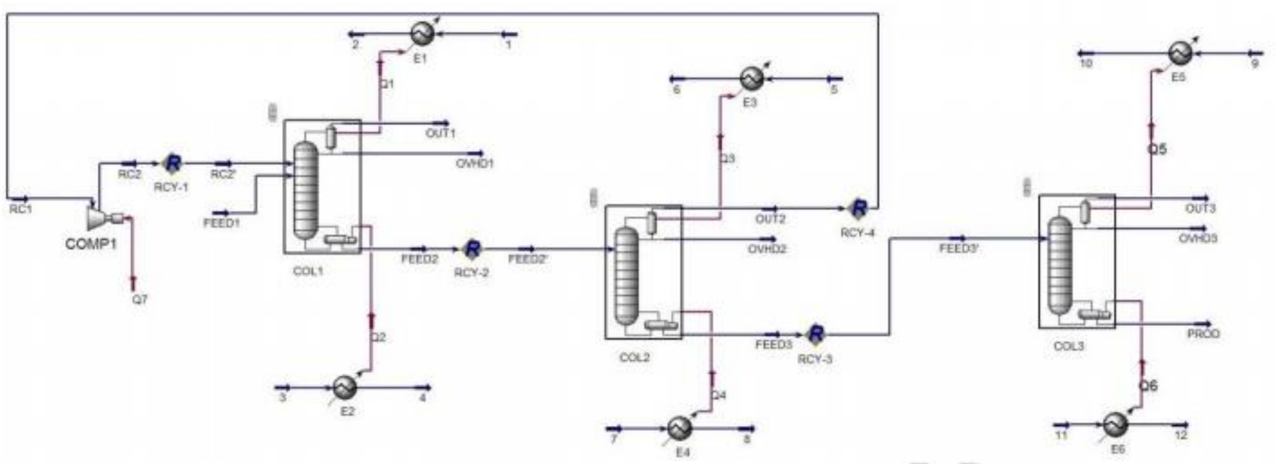

Fig. 1. Process flow diagram of oxygen-18 production using cryogenic oxygen distillation [6]

Besides, the study of membrane technology is increasing which produces various findings related to precursors that are suitable for the gas separation process [8-10]. One of the material is specialized ceramic membranes that capable of oxygen separation process at an arising temperatures which are different compared to cryogenic distillation where the super-cooled temperature is required. These innovative ceramic-based membranes separate oxygen from the air at an arising temperature in the range between 800 up to 900 °C. The ceramic membranes which have high solvents and chemical resistance, high mechanical strength and stability, also long term durability causes them to be preferable [41]. The acronyms for high-temperature ceramic membranes that typically used are Oxygen Transport Membranes (OTM), Ion Transport Membranes (ITM) and Mixed Ionic Electronic Conducting (MIEC) which will be used in this paper. Various scientific publications regarding this study have been made in the early 2000s up to now showing there is high attention has been given in this research field [11]. A review has been done by Sunarso *et al.*, for the last three decades, on the development of dense ceramic-based membranes focusing on separating oxygen using mixed ionic-conducting ceramic membranes [12]. The development of ion transport membranes (ITM) in gas separation, specifically for separation of oxygen from the air has been discussed in a chapter of a book published by Bose *et al.*, [13]. The changes of oxygen permeation flux by modifying mixed ionic-electron materials has been emphasized in several publications such as Yang *et al.*, have reviewed the developments and issues of perovskite-type ceramic membrane including their applications in SOP, POH, POM, OCM, and SOE [14]. Since ceramic-based membranes have shown their potential to separate oxygen from the air, the objective of this study is to review the advancement of ceramic-based membranes in separating oxygen from air under high temperature including the challenges in the context of separation and selectivity in membrane performance also, various methods to counter the challenges. This paper covers the potential of ceramic-based membranes for oxygen separation applications and to be commercially marketed in the industry shortly.

2. Implementation of Ceramic-Based Membranes in Separating Oxygen

Researches focusing on the development and enhancement of ceramic-based membranes at high temperatures has been done for oxygen applications in the past years. There are two categories of oxygen separation systems using ceramic-based membranes which are pure oxygen conducting membranes and impure oxygen conducting membranes. Pure oxygen conducting membranes use solid electrolytes that create an electron pathway in the presence of electrodes [15]. Its major advantage compared to impure oxygen conducting membranes is the ability to control the generation of oxygen through the electric current application. Impure oxygen conducting membranes or also commonly known as mixed ionic-electronic conducting membranes which can operate without the presence of electrodes and an external circuit. Even though the partial pressure gradient of oxygen involved in this system where oxygen permeates from higher partial pressure side oxygen to lower partial pressure side oxygen, its conductivity of electrons acts as an internal short circuit [16]. The neutrality of charge is controlled by neutralizing the electrons' flux. The advantage of using this system is its capability to produce high purity oxygen. The combination of ceramic materials and mixed ionic-electronic conducting characteristics commonly produce well-defined phase structures fabricated from precursors such as perovskite, brownmillerite, fluorite and other corresponding materials [17]. Perovskite and fluorite are the best materials among other combinations of ceramic membranes and mixed ionic-electronic conducting characteristics in terms of permeation properties for oxygen. This paper is focusing on the combination of perovskite and ceramic membranes with the highest permeability that shows good potential to be enhanced in the future [3,18].

The ceramic-based membrane is considered as a new approach compared to the cryogenic distillation approach to separate oxygen from the air to supply for fuel production and power generation usage such as coal gasification systems, production of syngas and oxyfuel combustion process. The most significant criteria for membrane-based separation involving high temperatures under high oxygen flux are comprehensively strong, stable, and high creep-resistant [19]. Therefore, the membrane performance needs to be improved under reducing and oxidizing conditions by doing research and development (R&D) widely. Since a large amount of oxygen is needed in the industry, efforts in developing appropriate demonstration-scale projects need to be performed at practical operating conditions. A project called MEM-BRAIN is carried out with the aim to combine the skills and knowledge of scientists and engineers to improve the performance of membranes, to specify targets in developing the system of the membrane and to determine the technical specifications for optimum operation. Research and development work should be done extensively to obtain clear identification for optimum membrane enhancement allowing applications in a long-term and reduced operation time [20,21]. Membrane surface area is the most important factor to be controlled as reduces in differential oxygen partial pressure will increase the surface area that causes increases in oxygen recovery rate. Monolithic tubes, honeycomb structures, planar arrangements, and hollow fiber membranes are the convenience conceptual alternatives [5]. The previous study by Tablet *et al.*, recommended a hollow fiber membrane has high flux properties and high packing density. The engineering parameters for scaling up need to be considered in hollow fiber membrane usage since they are fragile and complex to be sealed [22]. Vente *et al*., have performed research on designing scale-up modules to separate oxygen resulting that tubes are preferable compared to flat plate multi-channel monoliths or hollow fibers [23]. Companies such as Air Products and Chemicals has used flat membranes with wafer configuration module in developing ion transport membrane with operating temperature 800 °C to 900 °C [24]. The combination of the composite multi-channel heat exchanger and membrane modules in extruded ceramics has been developed by Hashim and co-workers under operating temperatures which is around 1300 °C whereas tubes are used by Praxair at an operating temperature of 800 °C up to 900 °C [25]. Sealing multi-channel monoliths is a complex process especially involving small channels with the purpose of membrane surface area maximization compared to sealing single tube configurations. Glass ceramic seals which composed of boron silicate glass has been used. It requires a design that is suitable for each application and it can be extremely brittle [17,25]. Previous researcher has reviewed the material options, design of module and membrane, seal enhancement and issues related to the fabrication process [26]. Silica ceramic membrane have been synthesized by Nor *et al.,* using the method of sol-dip coating. In the research, variety number of silica sol dipping are used in which resulting conversion of rough surface into a smoother surface and ceramic support pore size is reduced [41].

3. Current Challenges and Future Prospect of Ceramic Based Membrane

Ceramics are well-known with the high compressive strength but have lower impact strength characteristics. Those characteristics make ceramics become highly brittle and easily fracture when low force is applied [27]. The sensitivity of ceramics makes it hard to fabricate. Based on the previous study, the lower force applied on ceramics can the driving force (pressure ratio) was lower, the selectivity results were higher and the separation process said to be more appropriate; in fact, the operating costs for the separation system also lower [8]. When the permeability was higher, the cost of the system was lowered and the membrane needed to be was also small. To overcome the challenges, the mixed ceramic membrane was studied and develop. On the other hand, the ceramic membrane also faces the high weight and high cost of ceramic components.

The ceramic membranes have a bright future ahead, the use of ceramic membrane not only applicable for produced and filter pure oxygen but the ceramic membrane was a function in medical applications. Due to the corrosion problem in medical mechanical appliances such as dental implants, hip sockets, joint implants, and heart valves, ceramic biomaterials have received much attention. The development of synthetic material such as ceramic membrane with specific characteristics can be induced by advances in material sciences [27]. The ceramic membrane also is known with bacteria resistance properties, high durability, high fluxes, high abrasion resistance, mechanical, chemical and thermal stability. The application of ceramic membrane was believed can be utilized not only for medical and oxygen separation but also for various industry such as food and beverages industry, textile industry, metal industry and environmental [24,27,28]. In the food and beverage industry, the ceramic membrane can be used to sterilize the milk and whey, to purify the drinking water, to dewatering of products and the clarification of juice produced. For the textile industry, the ceramic membrane can use for desalination, separation of yeast and disposal of fat emulsions. Moreover, the ceramic membrane has a bright future in the metal industry as a platform for water/oil treatment, recovery of heavy metals and treatment of wastewater from the glass fiber production. In terms of environment, the ceramic membrane can be used for purification of the drain sewage plants. The future of ceramic membrane was very promising and applicable in the various industry [22,24].

4. Significant Issues of Mixed Conducting Ceramic Membranes and Directions in Future

Issues related to mixed conducting ceramic membranes remain even though various advancements have been applied for the past decades. The issues highlighted are

i. membranes should have extensively high oxygen permeation flux at high operational temperature
ii. membranes are required to be stable at elevated temperature with reduced atmosphere for a long operational period
iii. membranes are necessary to have acceptable mechanical strength
iv. materials of the membranes must be cost-effective and
v. involves high temperature sealing of the membranes.

Other than that, membrane stability is also an issue regarding mixed conducting ceramic membranes. High ionic conductivity membrane materials have a property of low chemical or/and mechanical stability. Perovskite-type ceramic membranes are usually composed of alkaline-earth compounds which have a very high oxygen permeation flux. However, these materials can react with gas, for example, SO_2, CO_2 or water vapor which will cause problems in terms of its long-term stability [3]. The efforts to improve its stability have been done by several researchers such as Tong *et al*., who blends B-site cation from perovskite compounds with zirconium B-site in which showing moderate prospects for certain composition [29]. The results from the previous study conducted by Xu *et al*., revealed that the new mixed ionic-electronic conducting compounds without cobalt causes a reduction in oxygen permeation flux [30]. Research on improvement of higher valence cations, for example, La into A also B-site cation of perovskite compounds has been carried out [31]. The result shows that there is an enhancement in membrane structure stability and reduction in electronic conductivity which reduces the oxygen permeation flux [32]. Figure 2 shows summary of synthesis methods for nanoporous metal oxide perovskites [33].

Presently, manufacturers are fabricating membranes with the thickness of the order of tens of micrometers as the relationship between oxygen permeation flux and thickness of the membrane is inversely proportional. The thin dense membrane that has structural support on the porous substrate in which functions to provide mechanical support is called an asymmetric structure. A very thin dense layer asymmetric membranes with porous support and porous substrate which usually fabricated from similar materials as the membrane may resolve these issues [34-36]. Research on changes of membrane surface having an oxide porous layer has been performed with properties of exchange kinetics in high oxygen surface [37]. Even though enhancement has been done using thick membrane configuration, the low absolute oxygen permeation flux is not suitable for practical applications due to the presence of low oxygen bulk diffusion [22,29]. The study of thin-film oxygen separation membranes with supported porous substrate shows that they are supposed to have a smaller size of pore than the top layer thickness used to allow them to have relatively high mechanical strength. Its support should have high-temperature resistance under the long-period operating temperature which will prevent them to undergo structural and chemical changes. Other than that, the top layer applied should be similar to both chemical and thermal expansion. Besides, the application of membrane gas absorption (MGA) can be used to enhance the performance of ceramic membrane by eliminating channeling and foaming issues during the process of separation takes place [44].

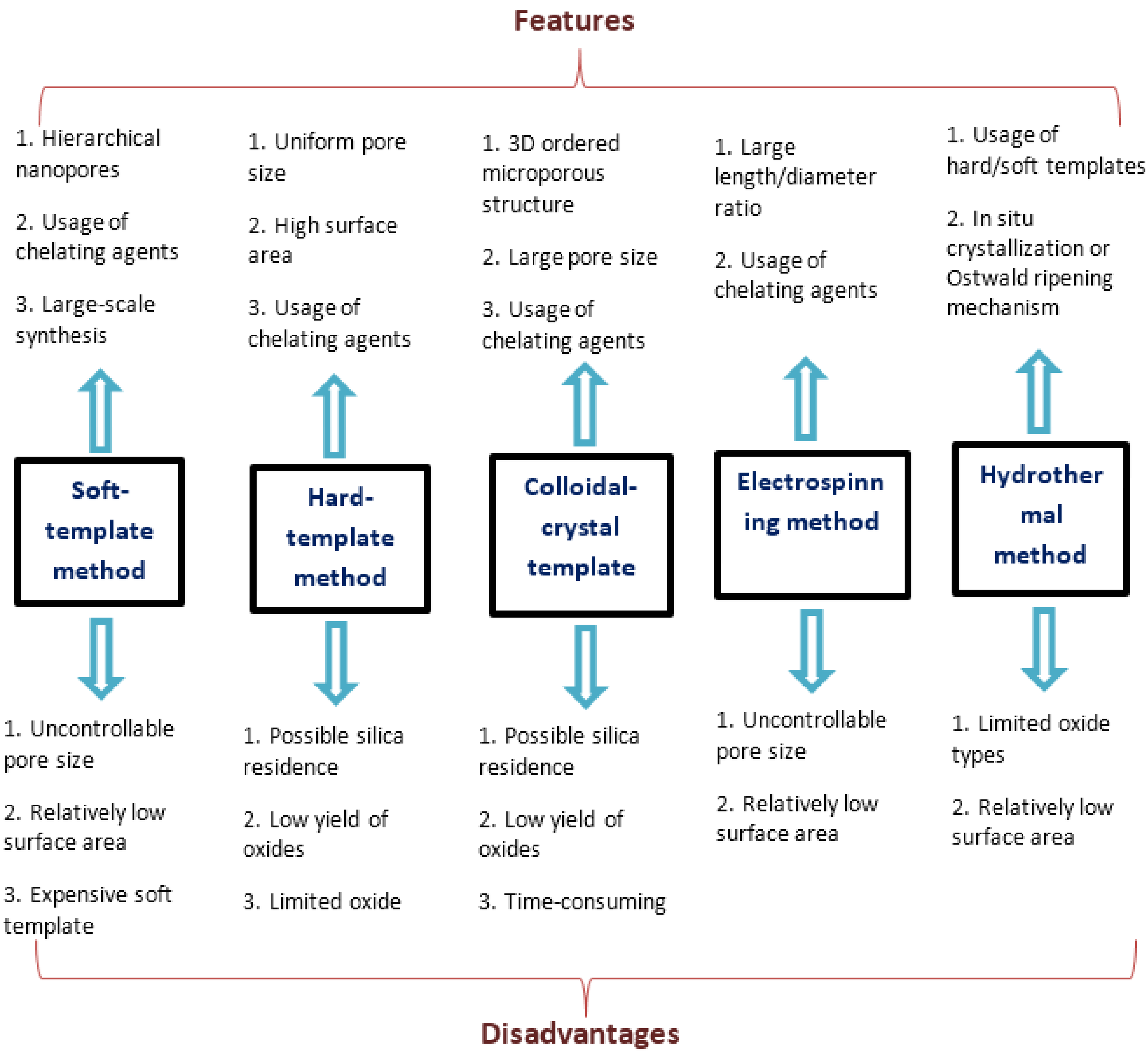

Fig. 2. Summary of synthesis methods for nanoporous metal oxide perovskites [33]

5. Conclusions

The ceramic-based membranes which have been enhanced for oxygen separation purposes and its potential to commercialized are already well-known globally. Several important issues such as module design, sealing, and geometry of membrane need to be considered to improve the performance of the membranes [38,39,40]. One of the interesting alternatives which shows a better quality and performance than planar, disk also tubular membranes is mixed-conducting ceramic-based hollow fiber membranes. These membranes have a large surface to volume ratio and high-temperature sealing. More research regarding perovskite-type mixed conducting membrane needs to be done on these lab scale membranes to widen its potential to be a large scale membrane for industrial applications. Even though a lot of improvements have been done on this membrane technology for oxygen separation, there are still challenges and problems that need to be considered. Therefore, studies focusing on properties of membrane especially thermal and chemical stability, geometries of membranes also sealing technology are required to be carried out. Scientific and engineering information is crucial to determine the effect of high-temperature membrane operation on the efficiency and energy required to implement this technology to produce and separate tonnage oxygen from the air. It is expected ceramic-based membrane technology with high potential in the context of production of fuel, sustainable energy also a cleaner environment can replace the energy-intensive traditional air separation method and can be implemented for the oxygen separation process in the near future.

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