



## Review

## Reverse-selective polymeric membranes for gas separations

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## ABSTRACT

This paper reviews the material design and fabrication of a class of membranes where some larger molecules can permeate faster than smaller ones, or so-called reverse-selective membranes, for conventional gas or vapor separation applications. The main requirements for a good reverse-selective gas or vapor separation membrane are high permeability, good separation capability, stable performance and sufficient mechanical properties to withstand the harsh environments in commercial separation processes. High permeability in reverse-selective membranes is achieved by tuning the fractional free volume (FFV) content, i.e., free spaces for penetrant diffusion, while facilitating preferential interaction between functional moieties of the membrane material and penetrant molecules to enhance penetrant sorption, and suppressing crystallinity where applicable. The separation capabilities of these membranes are generally controlled by the preferential sorption of larger condensable penetrants over smaller penetrants rather than size-selective diffusion common for conventional gas separation polymers. Reverse-selective gas separation membranes have attracted much attention owing to their advantages for certain commercial gas separation applications. This review summarizes the different aspects of polymeric reverse-selective gas separation membranes reported in the literature and comments briefly on their commercialization potential.

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## Contents

|   |     |
|---|-----|
| 1. Introduction .....                                       | 741 |
| 1.1. The need for gas and vapor separation membranes .....  | 741 |
| 1.2. Brief history of membrane technology .....             | 741 |
| 1.3. The solution-diffusion model .....                     | 742 |
| 2. Fundamentals of gas/vapor separation membranes .....     | 742 |
| 3. Reverse-selective membranes from glassy materials .....  | 745 |
| 3.1. Polymers with intrinsic microporosity .....            | 745 |
| 3.2. Nanocomposites .....                                   | 749 |
| 3.3. Polar additives .....                                  | 749 |
| 4. Reverse-selective membranes from rubbery materials ..... | 749 |
| 4.1. Poly(ethylene oxide) (PEO) .....                       | 749 |

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|      |   |     |
|------|---|-----|
| 4.2. | Copolymers containing poly(ethylene oxide) .....              | 752 |
| 4.3. | Polymer blends based on poly(ethylene oxide) .....            | 754 |
| 4.4. | Poly(ethylene oxide) nanocomposites .....                     | 754 |
| 4.5. | Ionic liquid membranes .....                                  | 757 |
| 4.6. | Polymerized ionic liquid membranes and their composites ..... | 759 |
| 5.   | Future directions of reverse-selective membranes .....        | 760 |
| 6.   | Conclusions .....   | 761 |
|      | Acknowledgements .....  | 762 |
|      | References .....  | 762 |

## 1. Introduction

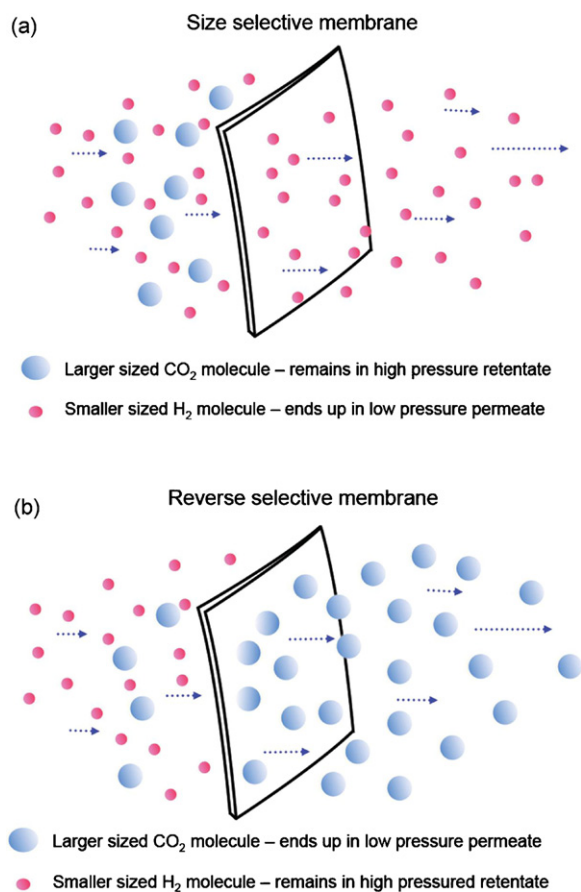
### 1.1. The need for gas and vapor separation membranes

It appears that one of the most important technological challenges of the 21st century will be overcoming the anthropogenic contributions to weather changes that could lead to serious socioeconomic and ecological consequences. A central contributor is carbon dioxide (and other acid gases) emissions from hydrocarbon combustion in power plants and industrial processes [1]. Many governments have implemented major initiatives to mitigate global warming for the next 20 years [2,3]. A futuristic approach to reduce, or even eliminate such emissions is to replace hydrocarbons with “green” resources like hydrogen [4] and nuclear energy [5], or renewable resources [6] like solar, wind, and geothermal energy to produce fuel and power. Prior to replacing hydrocarbons with the aforementioned green energies, safety concerns about nuclear energy, low reliability and high production cost of hydrogen supplies, and geographical constraints of renewable resources must be overcome. In the near term, energy or fuel production from green resources is more costly, and less effective, compared to hydrocarbon combustion. The short-term solution is to improve acid gas capture technologies and the efficiencies of processes that use fossil fuels. Traditional gas capture or vapor separation techniques like pressure swing adsorption and chemical absorption are efficient but at the expense of large carbon footprints [7–9]. Therefore, it is important that affordable separation techniques are identified and improved. Membrane technology will play a significant role in meeting these needs. Typically, one expects membranes to transmit small molecules more rapidly than larger ones; however, there are numerous applications where it would be advantageous to have membranes that preferentially transmit certain larger molecules faster than smaller ones, i.e., the reverse of the expected size selectivity. The purpose here is to review the growing literature on “reverse-selective” membranes that transmit some larger molecules faster than smaller ones and the opportunities for their application.

### 1.2. Brief history of membrane technology

Membranes are potentially energy-efficient, or “green”, candidates for acid gas capture and vapor/gas separations [10–13]. Fig. 1 shows a graphical representation of a membrane, which can be defined as a thin film that is selectively

permeable towards a penetrant molecule. A French cleric, Abbe Nollet, discovered the first membrane process, the phenomenon of osmosis, in 1748, see, for example, [14]. In 1855, German physiologist Adolf Fick mathematically described solution dialysis via artificial collodion membranes [15,16]. In 1866, Thomas Graham reported on gas separations using rubber membranes and postulated the solution-diffusion theory [17]. In the 1940s and 1950s, Barrer [18,19], van Amerongen [20], Stern [21,22], and Meares [23] contributed to the foundations for modern day gas permeation and separation theories. The first instance of



**Fig. 1.** Graphical representations of (a) size-selective and (b) reverse-selective polymeric gas separation membranes. Depending on applications and membrane choice, the target gas can be kept in the high-pressure retentate; thus, minimizing recompression costs. For example, if H<sub>2</sub> is the target gas, a reverse selective membrane is preferred as the H<sub>2</sub> remains in the high pressure retentate.

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