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# $CO_2/N_2$ separation via multilayer nanoslit graphene oxide membranes: Molecular dynamics simulation study

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ABSTRACT

The stacked graphene oxide (GO) membranes have received considerable attention due to their excellent advantage for gas separation. Recent experiments show that different structures of the membrane endow it with different separation performance. However, the underlying origin of the structure–property relationship, which is critically important in materials chemistry and engineering, is still unclear. In this work, adopting molecular dynamics simulations, the separation of $CO_2$ and $N_2$ through bilayer GO membranes was studied. The effects of nanoslit width (W), nanoslit offset (O) and interlayer space (D) on gas separation performance were investigated. Our research suggests that the separation performance could be optimized through regulating the microscopic structure of the GO membranes. This work also provides guidelines for rational design of GO membranes for gas separation.



## 1. Introduction

Global $CO_2$ emissions, largely coming from the combustion of fossil fuels, have increased steadily over the last century and become one of the most critical environmental issues [1,2]. Carbon capture and storage provides an option to mitigate greenhouse gas emissions and retard global warming [3,4]. Among various carbon capture techniques, membrane-based gas separation has evolved as a promising technological solution for the capture of $CO_2$ from large emitters due to its high energy efficiency and relatively low capital cost [5–8]. Traditional membranes such as metal, polymer, and zeolite membranes have been widely used for $CO_2$ separation [9–11]. However, for all types of membranes, there is a trade-off between selectivity and permeance. Since the permeance of a membrane is inversely proportional to its thickness [12], single-atom thick membranes are particularly interesting for gas separation.

Graphene, its only one-atom-layer thickness and perfect $sp^2$ hybridized carbon lattice facilitate the fabrication of ultrathin selective barriers with maximum flux and desired separation abilities if nanopores can be inserted artificially in the matrix [13–17]. The separation performance of such nanoporous membranes is largely dependent on the pore size/density [14]. Although the use of elaborate oxidative etching or electron/ion bombardment could drill holes in graphene, a precise, large area, and high-density perforation remains a technical challenge [18]. Compared with graphene, graphene oxide (GO) layers could be easily fabricated, and the two dimensional (2D) nanochannels among the GO layers could also be reconstructed via artificially design of the stacking structure [5,6,19,20]. Up to date, the experimental results indicate that the GO membrane has excellent gas separation performance. Kim et al. [21] identified that the well stacked graphene and GO membrane could enhance $CO_2/N_2$ separation. Li et al. [22] found ultrathin GO membrane (thickness < 1.8 nm) exhibited $H_2/CO_2$ separation. Shen et al. [6,19] found that 2D GO channels provided membrane with excellent $H_2/CO_2$ and $CO_2/N_2$ separation. In the GO-based stacked membranes, the percolated network features a rich microstructure with inclusion of the interlayer capillary gallery and the slits between edges of neighboring sheets or the pores within GO planes (Fig. 1a) [21,23–25]. Molecular permeation through the interconnected network of GO membrane occurs through the interlayer galleries, interedge voids and GO pores. Molecular transport channels can be created across or along the nanosheets. And the separation performance depends on the microscopic structure of the GO membranes which can be regulated in the process of making film, for example, by controlling humidity, inserting nanoparticles between adjacent layers, or modifying chemical groups with different chain lengths [26–28]. However, the relationship between sophisticated microstructure and separation performance of GO has not been fully revealed.

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