Gas Permeation and Separation Properties of Large-Sheet Stacked Graphene Oxide Membranes

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Abstract

Graphene oxide (GO) membranes offer attractive gas separation properties. However, the gas separation mechanism for GO membranes is unclear due to inconsistent permeation and separation results reported in the literature. In this work, pure gas permeation and separation of equimolar (H_2/CO_2) mixture experiments were conducted on GO membranes made from large GO sheets of different sizes (33 and 17 μm) to understand the gas permeation and separation characteristics of these membranes. At room temperature the permeation of large molecules (CH_4, N_2 and CO_2,) through GO membranes exhibits Knudsen–like diffusion characteristics, with the premeance for the small sheet GO membrane about twice that for the large sheet GO membrane. The smaller gases (H_2 and He) exhibit much higher permeance, showing additional transport in additional pathway. The GO membranes show good H_2/CO_2 selectivity for both pure and binary gas feeds, without CO_2 pore blockage effect for mixture separation found for crystalline microporous membranes. An inter-sheet and inner-sheet two-pathway model is proposed which can explain the results obtained in this work. Gas permeation in GO membranes, more complex than in crystalline microporous membrane, is determined by solubility (surface properties), diffusivity (relative molecular size to pore size), porosity and tortuosity of both the inter-sheet pores and inner-sheet defect pores. These properties are strongly influenced by synthesis method and conditions for GO sheets and membranes.

Graphical abstract