Chemical Engineering Doctoral Defense

Synthesis and Gas Transport Properties of Graphene Oxide Membranes

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abstract

Graphene oxide (GO) membranes have shown promising gas separation characteristics that make them of interest for large-scale industrial applications. However, the gas transport mechanism for GO membranes is unclear due to inconsistent permeation and separation results reported in literature. GO membranes examined for gas separation were prepared on substrates and/or by synthesis methods that are difficult to scale up that makes their impact on industrial applications insignificant. Moreover, the production of GO membranes with fine-tuned interlayer galleries for improved molecular separation is still a challenge. In this study, high quality GO membranes are synthesized on polyester track etch substrates (PETE) by different deposition methods. The produced membranes were characterized by conventional characterization techniques such as XRD, FT-IR and SEM in addition to single gas permeation and binary H2/CO2 mixture separation experiments. GO membranes are made from large GO sheets of different sizes (33 and 17 □m) using vacuum filtration to understand the gas permeation and separation characteristics of these membranes and shed more light on their transport mechanism. On the scalable PETE polymer substrate, GO membranes are synthesized using spray coating methods as a simple, scalable and cost-effective approach for membrane synthesis for large scale industrial applications. Finally, Brodie's derived GO sheets were used to prepare GO membranes with narrow interlayer spacing to improve H2/CO2 separation performance. An inter-sheet and inner-sheet two-pathway model is proposed to explain the permeation and separation results of GO membranes obtained in this study. At room temperature, the large molecules (CH4, CO2, and N2) permeate through inter-sheet pathway of the GO membranes, exhibiting Knudsen diffusion characteristics, with the permeance for the small sheet GO membrane about twice that for the large sheet GO membrane. The smaller gases (H2 and He) exhibit much higher permeance, showing significant flow through the inner-sheet pathway in addition to the flow through the inter-sheet pathway. GO sheets' edge to edge interactions and hence the formation of GO extrinsic wrinkles in the finally assembled membrane was minimized using dilute GO suspensions and spray deposition technique, which leads to membranes with a smooth surface and enhanced separation quality. Brodie's derived GO membranes show a sheet spacing height of~ 3 Å which led to significant reduction in the permeability of large gas molecules compared to Hummers' derived GO membranes and thus a notable improvement in H2/CO2 selectivity could be achieved.

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