

OXIDATIVELY STABLE MEMBRANES FOR CO₂ SEPARATION AND H₂ PURIFICATION

W.S. Winston Ho, William G. Lowrie Department of Chemical and Biomolecular Engineering ; Department of Materials Science and Engineering, The Ohio State University, USA
ho.192@osu.edu

Varun Vakharia, William G. Lowrie Department of Chemical and Biomolecular Engineering, The Ohio State University, USA

Witopo Salim, William G. Lowrie Department of Chemical and Biomolecular Engineering, The Ohio State University, USA

Michael Gasda, Bloom Energy Corporation, 1252 Orleans Drive, Sunnyvale, USA

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CO₂-selective facilitated transport membranes are well-known for providing remarkably high CO₂/H₂ selectivity along with high permeance at high temperatures (100 – 120°C). In some cases, it is desirable to use air as the sweep gas to enhance the driving force and membrane performance, and the membrane should be stable in the presence of oxygen. This work demonstrates the development of a new class of facilitated transport membranes containing quaternaryammonium hydroxide small molecules and quaternaryammonium hydroxide- and fluoride-containing polymers as mobile carriers and fixed-site carriers, respectively, for CO₂ separation and H₂ purification. The active nature of tetramethylquaternaryammonium hydroxide (TMAOH) as a mobile carrier was successfully demonstrated with the high CO₂ permeance obtained by the TMAOH-containing membranes. However, the membrane performance was improved significantly by the incorporation of quaternaryammonium hydroxide- and/or fluoride-containing polymers in the membrane. The resulting hydroxide- and fluoride-containing membranes exhibited CO₂ permeance > 100 GPU and CO₂/H₂ selectivity > 100 at 120°C using humid air as the sweep gas. The membrane composition was optimized, and the transport stability of the membrane was investigated. The membrane showed oxidatively stable during the 145-hour transport measurement at 120°C using air as the sweep gas. Furthermore, the effects of sweep steam content and membrane thickness were investigated. As the sweep steam content was increased (especially for steam content > 50%), both CO₂ permeance and CO₂/H₂ selectivity increased. As the membrane thickness was reduced from 15 μm to 2 μm, a sharp drop in the CO₂/H₂ selectivity was observed whereas the CO₂ permeance did not seem to increase as prominently as the H₂ permeance. In addition, the membrane was successfully scaled up using a roll-to-roll continuous membrane fabrication machine, and the scale-up membrane showed similar performance as the lab-scale membrane.