Chemical Engineering Doctoral Defense

Synthesis and Characterization of Thin Ceramic-Carbonate Dual-Phase Membranes for Carbon Dioxide Separation

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abstract

High temperature CO_2 perm-selective membranes offer potential for uses in various processes for CO_2 separation. Recently, efforts are reported on fabrication of dense ceramic-carbonate dual-phase membranes. The membranes provide selective permeation to CO_2 and exhibit high permeation flux at high temperature. Research on transport mechanism demonstrates that gas transport for ceramic-carbonate dual-phase membrane is rate limited by ion transport in ceramic support. Reducing membrane thickness proves effective to improve permeation flux. This dissertation reports strategy to prepare thin ceramic-carbonate dual-phase membranes to increase CO_2 permeance. The work also presents characteristics and gas permeation properties of the membranes.

Thin ceramic-carbonate dual-phase membrane was constructed with an asymmetric porous support consisting of a thin small-pore ionic conducting ceramic top-layer and a large pore base support. The base support must be carbonate non-wettable to ensure formation of supported dense, thin membrane. Macroporous yttria-stabilized zirconia (YSZ) layer was prepared on large pore $Bi_{1.5}Y_{0.3}Sm_{0.2}O_{3-\delta}$ (BYS) base support using suspension coating method. Thin YSZ-carbonate dual-phase membrane (d-YSZ/BYS) was prepared via direct infiltrating Li/Na/K carbonate mixtures into top YSZ layers. The thin membrane of 10 μm thick offered a CO_2 flux 5-10 times higher than the thick dual-phase membranes.

$Ce_{0.8}Sm_{0.2}O_{1.9}$ (SDC) exhibited highest CO_2 flux and long-term stability and was chosen as ceramic support for membrane performance improvement. Porous SDC layers were co-pressed on base supports using SDC and BYS powder mixtures which provided better sintering comparability and carbonate non-wettability. Thin SDC-carbonate dual-phase membrane (d-SDC/SDC60BYS40) of 150 μm thick was synthesized on SDC60BYS40. CO_2 permeation flux for d-SDC/SDC60BYS40 exhibited increasing dependence on temperature and partial pressure gradient. The flux was higher than other SDC-based dual-phase membranes. Reducing membrane thickness proves effective to increase CO_2 permeation flux for the dual-phase membrane.

April 09, 2014; 9:30 AM; ENGRC 490