

Chemical Engineering Doctoral Defense

Synthesis and Stability of Ceramic-Carbonate Dual-Phase Membrane For Carbon Dioxide Separation

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abstract

Of the potential technologies for pre-combustion capture, membranes offer the advantages of being temperature resistant, able to handle large flow rates, and having a relatively small footprint. A significant amount of research has centered on the use of polymeric and microporous inorganic membranes to separate CO₂. These membranes, however, have limitations at high temperature resulting in poor permeation performance. To address these limitations, the use of a dense dual-phase membrane has been studied. These membranes are composed of a conductive solid and conductive liquid phase that have the ability to selectively permeate CO₂ by forming carbonate ions that diffuse through the membrane at high temperature. The driving force for transport through the membrane is a CO₂ partial pressure gradient. The membrane provides a theoretically infinite selectivity.

To address stability of the ceramic-carbonate dual-phase membrane for CO₂ capture at high temperature, the ceramic phase of the membrane was studied and replaced with materials previously shown to be stable in harsh conditions. The permeation properties and stability of La_{0.6}Sr_{0.4}Co_{0.8}Fe_{0.2}O_{3-δ} (LSCF)-carbonate, La_{0.85}Ce_{0.1}Ga_{0.3}Fe_{0.65}Al_{0.05}O_{3-δ} (LCGFA)-carbonate, and Ce_{0.8}Sm_{0.2}O_{1.9} (SDC)-carbonate membranes were examined under a wide range of experimental conditions at high temperature. LSCF-carbonate membranes were shown to be unstable without the presence of O₂ due to reaction of CO₂ with the ceramic phase. In the presence of O₂, however, the membrane shows stable permeation behavior for more than one month at 900°C. LCGFA-carbonate membranes show great chemical and permeation stability in the presence of various conditions including exposure to CH₄ and H₂, however, the permeation performance is quite low when compared to membranes in the literature. Finally, SDC-carbonate membranes show great chemical and permeation stability both in a CO₂:N₂ environment for more than two weeks at 900°C as well as more than one month of exposure to simulated syngas conditions at 700°C. Ceramic phase chemical stability increased in the order of LSCF < LCGFA < SDC while permeation performance increased in the order of LCGFA < LSCF < SDC.



October 9, 2013; 2:30 PM; ERC 490