ICCDU XVI
27-30 August, 2018
Rio de Janeiro, Brazil

16th INTERNATIONAL CONFERENCE ON CARBON DIOXIDE UTILIZATION

Fluorite-perovskite-molten carbonates dense membrane for CO_2 separation at high temperatures, process enhanced by surface and composition modifications

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Keywords: dense membrane, CO_2 separation, fluorite-perovskite

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Membrane technologies, for gas separation, have been used in the industry for CO_2 capture since the last 2-3 decades, although most of these technologies are implemented at low temperatures. During the last 10 years, it has been proposed the use of dense ceramic-carbonate membranes to separate CO_2 selectively from combustion gas at high temperatures ($T \geq 700$ ºC). These membranes show a new CO_2 separation mechanism with an ideal infinite selectivity. These membranes could reduce costs in the industrial processes because membranes reactors can be developed to separate and utilize CO_2 as raw material without the need of extra energy. In this work, our research group has implemented a composite, mixture of fluorite and perovskite phases, as the ceramic section of membranes. Mixed conduction, through the perovskite phase, increased CO_2 permeation when the feed gas was a mixture of CO_2 and a low concentration of O_2, but some O_2 was permeated as well. However, when the perovskite concentration was modified a high CO_2 permeation remains, while O_2 permeation decreases. Also, some membranes were superficially modified by physical vapor deposition of metallic particles. Results showed higher CO_2 permeation due to the reaction between CO_2, O_2 and electrons coming from the metallic particles.

Specifically, this work investigates the stability of the $Ce_{0.85}Sm_{0.15}O_2$-$Sm_{0.6}Sr_{0.4}Al_{0.3}Fe_{0.7}O_3$ (CSO-SSAF) composite for the fabrication of dense ceramic-carbonate membranes. Membranes were fabricated by direct infiltration of molten carbonates ($K/Na/LiCO_3$) in porous CSO-SSAF disks. CO_2 separation tests were performed using a home-made set-up at high temperatures. A mixture of CO_2, O_2, N_2 and Ar, as carrier gas, was used as feed gas as well as Ar at the sweep gas, in some cases O_2 was also injected in the permeate side as sweep gas. The composite was synthesized by the EDTA-citrate (one-step) method. Results show that this synthesis method makes a stable composite. These membrane presented high CO_2 permeance above 700°C. The membrane presents two CO_2 separation mechanism (Figure 1), the first one due to the reaction between the fluorite and perovskite O^{2-} ions and the CO_2 from feed side. The second mechanism consists in the surface reaction between the electrons from the perovskite and metallic particles and O_2 from the feed side. In both cases, carbonate ions are produced and transported through the molten carbonate phase to the downstream side, where CO_2 is released, while O^{2-} ions and electrons return to the ceramic phases.

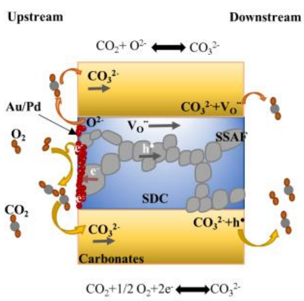

Figure 1. Schematic representation of CSO-SSAF-molten carbonates membrane for CO_2 separation.