

DENSE CERAMIC MEMBRANES FOR METHANE CONVERSION*

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Several perovskite-type oxides (ABO_3) that contain cation dopants show mixed (electronic/ionic) conductivity. These mixed-conductivity oxides are promising materials for oxygen-permeating membranes that can operate without electrodes in producing synthesis gas (syngas, i.e., $CO + H_2$) by partial oxidation of methane, the main component of natural gas. The syngas could be used to produce high-value-added products ranging from environmentally friendly liquid fuels to feedstocks for a wide range of chemical processes. This electrochemical conversion technology, based on dense ceramic membranes, is of great importance to petrochemical industries.

Ceramic powders in the Sr-Co-Fe-O system with various stoichiometries are fabricated into dense membrane tubes by a plastic extrusion technique. To prepare the ceramic powder for extrusion, it is mixed with several organic additives to make a homogenized mass with enough plasticity to be easily formed into various shapes while retaining satisfactory strength in the green state. The plastic mass is then forced through a die at high pressure to extrude hollow tubes, which are sintered at $\sim 1200^\circ\text{C}$ for 5-10 h and then characterized by optical and scanning electron microscopy, X-ray diffraction, and density measurements. Subsequently, they are evaluated as part of a reactor operated at $\sim 850^\circ\text{C}$ for conversion of methane into syngas in the presence of a reforming catalyst. Methane conversion efficiencies of $>98\%$ and CO selectivity of $\sim 90\%$ have been observed. As expected, the measured H_2 yield was about twice that of CO. Oxygen permeations >20 std $\text{cm}^3/\text{min}/\text{cm}^2$ have been observed under certain reactor operating conditions. In this presentation, we describe fabrication of the membrane tubes, their characterization, and their performance in a conversion reactor. Other geometric forms of the reactor, such as honeycomb or corrugated arrangements, have also been fabricated and details will be presented.

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