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Investigation of oxygen transport membrane reactors for oxy-fuel combustion and carbon capture purposes

Ryan Falkenstein-Smith*, Pingying Zeng, Jeongmin Ahn

Department of Mechanical and Aerospace Engineering, Syracuse University, Syracuse, NY 13244 USA

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Abstract

There is an increasing pressure for industry to reduce carbon dioxide (CO_2) emissions from combustion processes, resulting in an increased interest in the development of methods to sequester and recycle CO_2 from flue gases. Current methods available to separate nitrogen and CO_2 for environmental benefits, such as chemical looping combustion (CLC), are expensive at the high flow rates encountered in industry. One potential alternative is a ceramic membrane catalytic reactor, which produces pure oxygen and simultaneously conducts oxy-fuel combustion, thus, CO_2 in the product stream can successfully be separated from the nitrogen in air. This work investigates the performance of a perovskite-type $SrSc_{0.1}Co_{0.9}O_{3-\delta}$ (SSC) and $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF) membrane reactors for the combustion of methane in various configurations. The ceramic membranes exploited here are oxygen semi-permeable, dense ceramic membranes based on the composite oxides with a high mixed oxygen ionic and electronic conductivity at high temperatures. The prepared SSC and LSCF hollow fibre membranes with catalysts were used to perform reactions with a methane fuel. The oxygen permeability feasibility of the membrane reactors were studied and confirmed. The CO_2 selectivity at various test conditions were also reported with the maximum selectivity achieved for SSC membrane to be 85.4% while LSCF membrane achieved 87.1% CO_2 selectivity.

Keywords: Ceramic membrane reactor; Methane combustion; Oxy-fuel combustion; CO_2 recovery

1. Introduction

Currently, fossil fuels are the primary source of energy that satisfies the increasing demand of almost every major country throughout the world [1]. However, it is projected that by combusting fossil fuels across the entire world, an estimated 30 Gt of carbon dioxide (CO_2) is produced per year, making fossil fuels the primary source of CO_2 emissions [2]. In order to mitigate the amount of carbon entering our atmosphere, that is shown to directly influence climate disruption, there is an increasing pressure on industry to reduce and/or contain emissions from combustion processes, while still meeting current and future energy needs. Therefore, different techniques and practices are being developed to capture and sequester carbon from energy production processes.

* Corresponding author. Fax: +1 3154439099.
E-mail addresses: rlfalken@syr.edu, rlfalken@gmail.com (R. Falkenstein-Smith).

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Recently, oxy-fuel combustion has been receiving more attention due to its "off the shelf appeal" and ability to be integrated into current energy production facilities. Oxy-fuel combustion is the process of using pure oxygen for fuel combustion, creating a CO_2 enriched flue gas that can easily be separated and sequestered [3]. Its components include an air separation unit (ASU), a furnace, and a CO_2 capture and compression unit [4]. Recent research mainly focuses on supplying high amounts of pure oxygen using cryogenic ASUs. This complicated process mainly involves cooling and compressing air to the point of filtrating nitrogen out, in order to deliver pure oxygen to the combustor. Although feasible in practice, cryogenic ASUs still require a tremendous amount of power to operate, reducing plant output by 6–8% [5], [6], [7] and [8].

Similarly, chemical looping combustion (CLC) is another method used to separate oxygen from air and deliver it to a combustion reactor [9]. CLC involves the use of an oxygen carrier such as nickel oxide, which is transported between the two reactors. In the first reactor, the oxide is reduced to form the metal in the presence of the fuel. In the second reactor, the metal is deoxidized to form the oxide in the presence of the combustion air. Thus direct contact between the combustion air and fuel is avoided [10]. However, this process is difficult to ensure sufficient energy transfer from metal reoxidation reactor to the endothermic oxide reduction reactor and requires the carrier to be mechanically and thermally stable [11].

Over the past few years, there has been a major focus on oxygen transport membranes (OTM) and their potential to transform the way oxygen is supplied and used in combustion processes. Currently, OTM technology is used for the production of hydrogen from natural gas, but has the potential to transcend the existing carbon capture technologies due to its low energy consumption and capture novelty [12]. OTM's oxygen semi-permeability is generally attributed to the partial substitution of both A and B cations in the perovskite lattice structure (ABO_3), facilitating the formation of oxygen vacancies via defect reactions.

As seen in Fig. 1, when the membrane is placed under elevated temperatures (> 700 °C), fuel can be introduced to one side of the membrane while exposed to air on the other side. This forces the positively charged oxygen vacancy to become disordered, creating a chemical potential gradient across the membrane and allowing oxygen to permeate through the material and react with the fuel on the other side. The motion of oxygen vacancies, which gives rise to the nature of oxygen permeability of perovskite membranes, is charge-compensated by the transport of electron-holes in the reverse direction [13]. Since there is a lack of nitrogen, the combustion process is simplified to elementary reactions, eliminating nitrogen oxide completely and producing a flue gas primarily composed of carbon

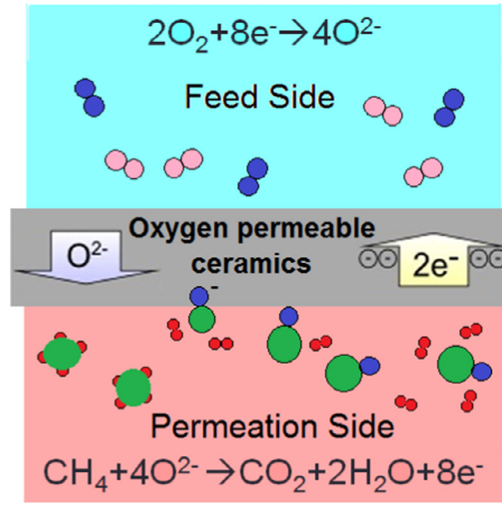

Fig. 1. Working concept of oxygen transport membrane used as a methane combustor reactor.

dioxide and water vapor. The CO_2 from the exhaust can then be captured and sequestered for further use.

Some studies have demonstrated dense perovskite ceramic membranes' oxygen permeability performance under industrial conditions. One example is Praxair, who has integrated OTM technology for the purpose of coal gasification and syngas production using their proposed OTM-Enhanced IG-NGCC concept. Additionally, Praxair has made great strides in demonstrating OTM stability and robustness of their designs under direct coal ash contamination and other harsh environments [14], [15] and [16]. However, limited investigation has been done concerning integrating OTM technology with methane combustion for carbon capture purposes [17] and [18]. In this study, the performance of oxy-fuel combustion reactors was investigated through modifying the reactor's composition, configuration, temperature, and sweeping gas flow rate. Oxy-fuel combustion performance was evaluated from the OTM reactor's oxygen permeability and CO_2 selectivity from the flue gas.

2. Experimental design

2.1. Material selection

In order to document the OTM's operation and performance, selecting a material that exhibits high oxygen permeation performance is crucial. Since Teraoka et al. first reported the remarkable high oxygen permeation flux through the ceramic disks based on the $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF) perovskite oxides in the 1980s, cobalt-contained perovskite membranes have been widely investigated