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A study of methane oxy-combustion characteristics inside a modified design button-cell membrane reactor utilizing a modified oxygen permeation model for reacting flows

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ABSTRACT

This work studies numerically the use of a modified oxygen permeation equation in a modified button-cell ion transport membrane reactor (ITMR). The permeation equation is modified in order to account for Reynolds number variations and non-elementary surface reactions on both sides of the membrane. The near membrane zone in the ITMR is modified to create recirculation zones in the vicinity of the membrane to sustain stable flame. Air is fed to the ITMR feed side and a mixture of methane and CO_2 is fed to the sweep side. The ITMR is axisymmetric and the mesh was constructed using Gambit 2.2 before importing it to the CFD fluent 12.1. A series of user defined functions (UDFs) written in C++ were compiled and hooked to fluent to solve for oxygen permeation. Effects of reactivity on oxygen permeation characteristics are investigated over wide ranges of operating parameters. Considering reacting flow conditions, the effects of sweep flow rate and feed oxygen partial pressure are investigated. The results showed that two, inner and outer, recirculation zones were created in the vicinity of the membrane with a stable flame. Significant improvements in oxygen permeation and combustion temperature are encountered utilizing the present reactor design.

1. Introduction

Nowadays, the increased level of emissions of carbon dioxide as the main greenhouse gas represents one of the most critical problems facing our present and future lives. There are many running research works around the world trying to reduce the emissions of carbon dioxide targeting the control of the globe temperature. Some focus is made on the use of renewable and clean energies; however, for all of these natural sources of energy, the economic cost is the main barrier toward their intensive applications (Habib et al., 2013a). At present, the whole world is forced to use the hydrocarbons fossil fuels in order to cover the tremendously increasing demand of energy. As a result, carbon dioxide emissions will be continuously increasing. This necessitates the treatment of carbon dioxide through its capture or storage (Wall, 2007). Based on that, the research work is directed toward technologies for carbon capture and sequestration. These technologies include pre-combustion, post-combustion and oxy-combustion carbon capture technologies. In the pre-combustion carbon capture technology, carbon dioxide is captured before the combustion process through gasification and shift conversion reaction processes of the hydrocarbon fuel. In the post-combustion capture, carbon dioxide is captured after the combustion process from the exhaust gases through scrubbing process. Chemically active agents like monoethanolamine (MEA) and methyldiethanolamine (MDEA) are primarily considered in the scrubbing process (Ben-Mansour et al., 2012). In the above mentioned two technologies for carbon capture, additional devices and modifications in the existing systems are required which will result in additional big cost. As one of the most promising techniques for CO_2 capture, oxy-combustion technology can be applied into the existing systems with slight modifications. In oxy-combustion technology, the hydrocarbon fuel is burned in a medium of pure oxygen and some recycled exhaust gases instead of air (nitrogen is not introduced into the combustion chamber). In this process, the combustion products consist of a mixture of only carbon dioxide and water vapor (Buhre et al., 2005). Water vapor can be easily

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condensed and accordingly carbon dioxide can be captured for industrial use or storage.

Oxy-combustion technology can be applied in the conventional combustion systems; however, this requires the separation of oxygen in another unit called air separation unit (ASU). This unit requires a part of the output power of the combustion system, and as a result, the overall system efficiency is reduced (Haslbeck et al., 2007). Recently, another approach for the application of oxy-combustion technology was proposed through the use of what's called oxygen transport membrane reactors (OTMR). In such kind of reactors, oxygen is being separated inside the combustion system through the use of ion transport membranes (ITM). At temperatures ranging from 650 °C to 950 °C, these membranes are activated for oxygen separation from the feed side to the permeate side of the membrane (Ahmed et al., 2014). In the permeate side of the membrane, fuel is being burned with the separated oxygen in a medium of recycled carbon dioxide. There are many membrane materials which can be used in such applications including lanthanum cobaltite perovskite ceramics, modified proviskite ceramics (Balachandran et al., 1997), brownmillerite structured ceramics (Schwartz et al., 2000), ceramic metal dual phase membranes (Chen et al., 1999), in addition to, thin duel phase membranes which consists of chemically stable yttria-stabilized zirconia (YSZ) (Kim and Lin, 2000).

Xu and Thomson (Xu and Thomson, 1999a) developed an explicit oxygen permeation model for ion conducting membranes while emphasizing the surface exchange kinetics at both sides of a $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF) membrane. They performed a series of experimental measurements over a broad range of oxygen partial pressure and temperature. They reported increase in the oxygen permeation flux while increasing the temperature and the oxygen flux is controlled by bulk diffusion at high temperatures. Rui et al. (Ruia et al., 2009) studied the permeation of oxygen across ceramic oxygen ionic conducting membranes under finite rate reactions conditions. They reported drop in the partial pressure of oxygen in the sweep side due to combustion, and accordingly, an increase in the oxygen permeation flux was obtained. In addition, both the oxygen partial pressure in the permeate side and the reaction rate were lowered as a result of increasing the inlet inert gas in the permeate side. The permeation of oxygen across ceramic oxygen ionic conducting membranes was investigated by Akin and Lin (Akin and Lin, 2004) using a simple mathematical model for different oxidation reaction kinetics mechanisms including both extremely fast reaction and no reaction conditions. The results were compared with the experimental oxygen permeation data for $Bi_{1.5}Y_{0.3}Sm_{0.2}O_{3-\delta}$ (BYS) for different conditions of reaction using ethane and methane. They showed that the flux of oxygen is strongly affected by the reducing gas flux and oxidation reaction. Their results showed high oxygen permeation flux by one order of magnitude in case of ethane than with methane due to the faster reaction rate of ethane as compared to methane. Habib et al. (Habib et al., 2013b) examined the influence of reactivity of methane in the permeate side of a stagnation flow ITM reactor on the permeation flux of oxygen across a $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF-6428) ionic conducting ceramic membrane. They attributed the sharp rise in the flux of oxygen while the reactions are activated in the permeate side mainly to the rise in the combustion temperature which enhances the diffusion of oxygen and reduces the bulk diffusion resistance of the membrane.

Hong et al. (Hong et al., 2012) studied numerically the operation of a LSCF ITM reactor under reactions conditions while incorporating a detailed gas-phase model of methane. Their results showed the dependence of oxygen permeation flux on reactor geometry, membrane temperature and feed and sweep flux rates. Ben-Mansour et al. (Ben-Mansour et al., 2013) and Nemitallah et al. (Nemitallah et al., 2013) studied the performance of a $La_{0.1}Sr_{0.9}Co_{0.9}Fe_{0.1}O_{3-\delta}$ (LSCF-1991) ITM reactor under non-reacting and reacting conditions considering different operating conditions. They reported significant effects of a set of parameters including inlet temperature, inlet methane concentration, and reactor geometry on the operation of the IT reactor. Hong et al. (Hong et al., 2013) studied the interactions between oxygen permeation and homogeneous-phase methane combustion in the permeate side of a LSCF membrane. They found enhancement of the oxygen permeation flux while the inlet temperature and fuel concentration are increased. Hunt et al. (Hunt et al., 2014) measured the oxygen profiles normal to the membrane surface and oxygen permeation flux across a $La_{0.9}Ca_{0.1}FeO_{3-\delta}$ (LCF) membrane and they developed a multi-step surface exchange model. Kirchen et al. (Kirchen et al., 2013) presented a novel ITM reactor for fundamental investigations of oxygen permeation and oxy-fuel combustion under reactive flow conditions. They studied experimentally the influence of inlet methane fraction on the oxygen permeation and fuel conversion in the sweep side of the membrane. They reported that in order to use an ITM reactor for complete fuel conversion (i.e. oxy-combustion application) the inlet fuel concentration should be kept below a certain limit to satisfy the stoichiometric conditions for combustion.

Recently, Mancini and Mitsos (Mancini and Mitsos, 2011) presented a study on the application of ITM reactors for large scale power generation. They proposed a design of a monolith structure ITM reactor to be used for power generation based on detailed numerical analysis. Their reactor has a total volume of 1000 m^3, 100,000 total feed and permeate channels, 266,700 m^2 surface area, a height of 4.75 m, length of 44.44 m and the reactor is able to deliver power in the range of 300–500 MWe based on the cycle first law efficiency. Nemitallah et al. (Nemitallah et al., 2014) developed a multi-parallel short channels compact ITM reactor design for gas turbine combustion application based on detailed 3-D numerical analysis. Their ITM reactor has a length of only 0.9 m, height of 3.35 m, volume of 10 m^3, surface area of 2700 m^2, 25,000 channels per stream, and the reactor is able to produce power ranging from 5 to 8 MWe based on the cycle first law efficiency. However of the extensive work done on the analysis of ITM reactors, still more research work is required on membrane materials to increase their oxygen permeability and to make them able to withstand high temperature under reacting conditions. Also, there are some problems regarding to the sustainability of the flame inside the ITM reactor in such difficult conditions for combustion inside the ITM reactor. In the present research work, methane-oxygen combustion is investigated numerically inside a modified button-cell ITM reactor. The modification of the button-cell reactor is done in such a way in order to create recirculation zones close to the membrane surface in order to sustain the flame. A modified oxygen permeation equation for oxygen permeation under reacting conditions is used. The permeation equation is modified in order to account for the effects of Reynolds number variations in both feed and permeate sides of the membrane due to the heat release from the combustion process. The permeation model is validated using the available experimental data of the same kind of the membrane material employed in the present work, BSCF membrane material. Wide ranges of operating conditions including flow rates, temperature, feed side pressure, and fuel concentration are considered in the present work.

2. Oxygen permeation model

Based on the oxygen chemical potential differences between the feed and the permeate sides of the membrane, the surface temperature of the membrane and the membrane ambipolar