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Numerical investigation of syngas oxy-combustion inside a LSCF-6428 oxygen transport membrane reactor

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ABSTRACT

The present work provides an investigation of the oxy-fuel combustion of syngas (mixture of CO and H_2) inside an OTR (oxygen transport reactor) of tubular shape and surrounded by air in an annulus. The syngas is generated from solar thermal reforming of methane. CFD (Computational fluid dynamics) calculations were performed using FLUENT 14.0 commercial code, where a series of UDFs (user defined functions) that enable the transfer of oxygen across the membrane were written in VC++, then compiled and hooked to FLUENT software. The models of oxygen permeation and reaction kinetics are validated against the available experimental data under similar oxy-combustion conditions. Simulations were performed considering non-reactive and reactive flow conditions. The results showed that the reactive flow results in increase in oxygen permeation flux of about four times the case of non-reactive flow. Oxy-combustion characteristics of synthetic gas in a medium of recirculated CO_2 are investigated. Considering reactive flow conditions, the effects of inlet temperature, CO_2 circulation, fuel composition and sweep gas flux on oxygen permeation and combustion temperature are studied. It was found that increase in inlet temperature, inlet fuel concentration, inlet hydrogen concentration and sweep flow rate result in high combustion temperature and improved oxygen permeation flux.

1. Introduction

Fossil fuels are used to satisfy a very significant fraction of the world energy demands. The use of fossil fuels involves some adverse effects, prominent among which is the emission of CO_2. Carbon dioxide significantly contributes to global warming and, consequently, causes climate change. This has called for international policies like the Kyoto protocol and Copenhagen summit that set limits on GHG (greenhouse gases) emission [1]. Renewable energy sources are carbon neutral and can present a favourable condition by reducing CO_2 emissions to the atmosphere. Unfortunately, they are not yet in abundant supply to meet man's demand. Much research and work are still needed to make them available in ample quantity to cater for our energy demands. Fossil fuels are, therefore, likely to remain as mankind major energy sources at least for the near future [2]. Carbon capture technologies are needed to enable the continuing usage of fossil fuels, yet decreasing the CO_2 emitted to the atmosphere, and thereby mitigating climate changes. Many methods have been proposed for carbon capture both numerically and experimentally [3]. Oxy-fuel combustion, which involves combustion with only oxygen resulting in exhaust gas that comprises CO_2 of high concentration and H_2O is a very promising technique compare to other carbon capture means. Oxy-fuel combustion is a very important carbon capture technology because almost all clean energy technologies require O_2 as their feed gas [4] and they are reported to have capture efficiency of around 90% [5].

Separation of the required oxygen from air needed for combustion can be achieved by various methods. ITM (Ion transport membrane) technology is one of these methods that utilizes excellent characteristics of high ionic and electronic conductivity [6]. ITMs, which have dense and non-porous characteristics, have very high flux and selectivity of oxygen. Their selectivity for oxygen is 100%, as they allow only oxygen ions to pass through them

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Nomenclature

C_i	Density of oxygen ions (mol/m^3)
CFD	Computational fluid dynamics (–)
$D_{i,j}$	diffusion coefficient of the component *i* in the component *j* (m^2/s)
$D_{i,m}$	diffusion coefficient in the mixture (m^2/s)
D_v	Diffusion coefficient of oxygen vacancy (cm^2/s)
DO	Discrete ordinates (–)
E	Activation energy (KJ/mol)
$\dot{h}$	electron hole (–)
ITM	Ion transport membrane (–)
J_{O_2}	oxygen permeation flux (mol/m^2-s)
k_f	forward reaction rate (cm $atm^{-0.5}s^{-1}$)
k_r	Reverse reaction rate (mol $cm^{-2}s^{-1}$)
LSCF	Lanthanum strontium cobaltite ferrite (–)
LSCF-6428	$La_{0.6}Sr_{0.4}$ $Co_{0.2}$ $Fe_{0.8}$ $O_{3-\delta}$ (–)
OTR	Oxygen transport reactor (–)
P'_{O_2}	Partial pressure of oxygen at the shell side or feed side (atm)
P''_{O_2}	Partial pressure of oxygen in the tube or permeate side (atm)
RTE	Radiative transfer equation (–)
$\vec{r}$	position vector (–)
$\vec{s}$	direction vector (–)
$\vec{s}'$	scattering direction vector (–)
S_i	Source/Sink term (Kg/m^3-s)
T	Temperature (K)
U	velocity vector (m/s)
UDF	User defined function (–)
$V_{\ddot{O}}$	Oxygen vacancy (–)
X_i	Mole fraction(–)
Y_i	Mass fraction (–)
σ	Stefan–Boltzmann constant (W/m^2-K^4)
σ_i	Ionic conductivity (S.m^{-1})
σ_s	Scattering coefficient (m^{-1})
μ	Dynamic viscosity (kg/m-s)
V_{cell}	Volume of cell (m^3)
ρ	Fluid density (Kg/m^3)

bringing about very pure oxygen. They have significant advantages over the traditional cryogenic air separation which produces oxygen with lesser purity [7] and has some energy penalties with low second law efficiency [8]. ITM can, therefore, be used in different reactors due to the very pure oxygen produced which is selectively used at the sweep side of the reactors through transport of oxygen ions [6]. They can also be incorporated into power generation cycles. ITMs are used for separating gas using transportation of ionized gas through a non-porous, dense ceramic membrane at elevated temperature (usually above 700 °C). With this, oxygen, which is necessary for oxy-combustion of fuels, can be separated from air. The water produced from oxy-combustion process can be removed by condensation and, therefore, enhances carbon capture. Permeation of oxygen through these membranes depends on the differences in partial pressures of oxygen across the membrane, operating temperature, gas flow rates and the membrane thickness [9–11].

The processes involved in the permeation of oxygen from the feed side to the sweep side of the membrane involve mass transfer of gaseous oxygen from the gas stream to the membrane surface at the higher pressure side (feed side). This is followed by adsorption of oxygen molecules then dissociation into ions (surface reaction at feed). Then, transport of oxygen ions across the membrane followed by surface reaction at the lower pressure side (sweep side) and finally transfer of oxygen from the membrane surface at the sweep side to the gas stream [4,6,12]. Permeation of oxygen can be affected by either surface exchange or bulk diffusion. At temperatures around 1033 K, the permeation is determined by the surface reaction at the sweep side but it is mostly controlled by bulk diffusion at more elevated degrees temperatures [10]. Many researchers have worked on various membrane materials used for oxygen permeation. Some of these investigations focused on experimental aspects while others were related to numerical work. The research by Qiu et al. [13] was on permeation characteristics of $SrCo_{0.8}Fe_{0.2}O_{3-\delta}$ membrane. Their results showed that the membrane with 1 mm thickness has a high flux of 3.1 ml/cm^2-min when operated at 850 °C, but the membrane was found to lack stability chemically and structurally. However, the stability was better at higher oxygen pressure. Kim et al. [14] formulated a model for predicting oxygen permeation flux for tubular membranes $Sm_{0.5}Sr_{0.5}CoO_{3-\delta}$ and $SrCo_{0.8}Fe_{0.2}O_{3-\delta}$. Wang et al. [15] studied the permeation characteristics of BSCF-5582 at elevated temperature and at different feed and sweep gas flow rates. Their results showed that increasing the sweep gas (helium) flow rate resulted in reduction in oxygen partial pressure at the sweep side; this consequently increases the permeation. Feed gas flow rate also have effect on oxygen permeation rate if the flow rate is less than 150 ml/min but it will have no effect at higher flow rates. Foy and McGovern [16] showed in their research that BSCF membranes have high oxygen permeation flux when compared to many other membrane types but, the BSCF membranes lack stability at high temperatures. LSCF membranes though have lower oxygen permeation flux when compared to BSCF but, the flux is sufficient in addition to its stability characteristics at elevated temperatures needed for combustion [17].

It is worth noting that conventional work in literature related to ion transport membrane whether experimental or numerical utilizes non-reacting/inert gases as their sweep gas. However, in recent years, few researchers have been able to predict oxygen permeation numerically by utilizing reactive gas [17–24]. Methane was utilized in the work by Hong et al. [6] and Habib et al. [17] both with LSCF membrane, as well as Ben-Mansour et al. [24] with BSCF membrane. In all cases, it was realized that the use of reactive gas as sweep gas raises the permeation flux. This is a result of the reaction of the gas with the permeated oxygen thereby increasing the chemical potential gradient. The higher permeation flux for the reaction mode was attributed to the increased oxygen partial pressure driving force and increased temperature of the membrane due to combustion. The membrane absorbs heat from the sweep side and transfers it to the feed side, leading to more permeation. All of the reactive mode studies proved the strong dependence of oxygen permeation on membrane temperature than on the partial pressure differences.

Other than pure oxygen separation, ion transport membranes can be used in partial oxidation of methane to produce syngas. The separated oxygen is consumed in the sweep side of the membrane in fuel conversion process. In order to enhance the fuel conversion process for syngas production in the membrane sweep side, certain catalyst coatings should be used in both sides of the membrane. In the feed side of the membrane, catalyst are required in order to promote oxygen separation form the feed gas, air cathodes similar to those used in solid oxide fuel cells can be used. Noble metals or