Energy 84 (2015) 600–611

Contents lists available at ScienceDirect

Energy

journal homepage: www.elsevier.com/locate/energy

Experimental and numerical study of oxygen separation and oxy-combustion characteristics inside a button-cell LNO-ITM reactor

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ARTICLE INFO

Article history:
Received 24 October 2014
Received in revised form
5 March 2015
Accepted 7 March 2015
Available online 29 March 2015

Keywords:
Button-cell reactor
LNO-ITM reactor
Oxygen permeation
Oxy-combustion characteristics

ABSTRACT

A combined experimental and numerical study is performed on a button-cell LNO-ITM reactor. A semi-empirical model for oxygen permeation is considered, *ABn* model, and the values of the empirical constants are calculated based on the fitting of the available experimental data in the literature. A validation study for the present model is performed using the present experimental data. A detailed numerical study is presented on an LNO-ITM button-cell reactor under oxy-fuel combustion conditions. CH_4 is used as the working fuel forming a mixture with CO_2 at the permeate side inlet. The model results showed reasonable agreements under different operating conditions. The effect of reactivity in the permeate side of the membrane on oxygen permeation flux is considered. It is found that the oxygen permeation flux is increased by about 50% for the case of reacting flow as compared to the case of non-reacting flow. Distinct behavior of oxygen permeation flux values through the present button-cell ITM reactor is encountered while varying the operating conditions as compared to other reactors in the literature. This may be attributed to the complicated design of the flow path close to the membrane surface which maximizes the effects of flow momentum on the oxygen flux.

1. Introduction

During last decades, especial care has been taken for the different applications of ion transport membranes (ITMs). These applications include pure oxygen separation from air (non-reacting medium), production of syngas (partial fuel conversion), and recently, oxy-fuel combustion technology [1]. All of these applications of ITMs are targeting lowered concentrations of the greenhouse gases in the ambient atmosphere as an action to reduce the globe temperature [2]. For the purpose of pure oxygen separation, cryogenic air separation units (CASU) are used and then the produced oxygen can be used in combustion in conventional combustion systems. However, the CASU is highly energy intensive, about 3–4% of the produced energy in oxy-combustion process is consumed in the CASU [3]. As an alternative to CASU, chemical looping air separation (CLAS) process can be used for pure oxygen separation for oxy-fuel thermal power plants. Due to its simplicity in terms of hardware, low cost, and the possibility of operation under atmospheric pressure; the CLAS process results in significantly low capital cost [4,5]. Recently, a new idea has been appeared in order to combine both the oxygen separation and oxy-combustion processes in one unit; this unit is called ITM reactors. In such kind of reactors, membrane is used to selectively separate oxygen from the flowing air in the feed side to the permeate side where the oxy-combustion process occurs. The membrane is heated using portion of the heat released from the combustion process in the permeate side. Different ITM reactor designs using different ceramic membrane materials are considered in the literature. In all cases, the main goal is to maximize the oxygen permeation rate while keeping the membrane temperature within the operating limits of the considered membrane material. Different membrane materials were considered in the literature including the lanthanum cobaltite perovskite ceramics [6–9]. Due to the specific needs raised by different membrane applications, especially in ITM reactor technology, successive research is being done for the development of new ceramic membrane materials that can withstand higher operating temperatures. These new materials include ceramic–metal dual phase membranes, modified perovskite ceramics, thin dual phase membranes including the Pd phases and yttria-stabilized zirconia (YSZ), and structured ceramic [10–13]. Recently, Nemitallah et al. [14,15] reported similar results at elevated membrane temperature. Using ultra-thin $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ ceramic membranes, Zydorczak et al.

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http://dx.doi.org/10.1016/j.energy.2015.03.022
0360-5442/

Nomenclature

ABn	oxygen permeation model
CASU	cryogenic air separation units
D_i	inner tube diameter
D_o	outer tube diameter
G.C.	gas chromatography
J_{O2}	oxygen permeation flux
LSCF	Lanthanum strontium cobaltite ferrite
P	gap height
SIMPLE	semi-implicit method for pressure-linked equations
TCD	thermal conductivity detector
T_∞	surrounding temperature
YSZ	Yttria-stabilized zirconia
CFD	computational fluid dynamics
CLAS	chemical looping air separation
DO	discrete ordinates
2-D	two dimensional
ITM	ion transport membrane
LNO	La_2NiO_4
MIEC	mixed ionic and electronic conducting
RTE	radiative transfer equation
S_i	source/sink term
T_M	membrane temperature
UDF	user defined function
ZEPP	zero emission power plant

[16] studied experimentally the influences of both the membrane surface temperature and the sweep gas flux on the oxygen permeation flux. Higher oxygen permeation fluxes were observed at higher membrane surface temperatures and higher sweep gas flow rates.

Other than pure oxygen separation, ITMs can be used in partial oxidation of methane to syngas. The separated oxygen is consumed in the sweep side of the membrane in the fuel conversion process [17]. Nowadays, most of the research works are focused on the application of the oxy-fuel combustion technology into ITM reactors. In ITM reactor technology, the separated oxygen is directly consumed in the combustion process with fuel in the sweep side of the membrane. In this case, the combustion products consist only of H_2O and CO_2. H_2O can be condensed easily through a simple condensation process, and then, CO_2 can be compressed for sequestration or industrial uses [14]. In case of ITM reactor applications, oxygen permeation rate is highly improved as compared to the non-reacting cases (pure oxygen separation). This is due to the reduced oxygen partial pressure in the sweep side of the membrane as a result of oxygen consumption in the combustion process. Also, due to elevated temperature as a result of exothermic reactions, the membrane total resistance for oxygen permeation is reduced, and then, the oxygen permeation flux is increased [15]. The characteristics of both oxygen permeation and oxy-combustion processes inside a stagnation flow ITM reactor were examined by Ben-Mansour et al. [18]. They reported significant increase in the oxygen permeation flux when the reactions are activated in the permeate side of the membrane as compared to the non-reactive case. Akin and Lin [19] conducted some experiments in order to investigate the influences of sweep gas flux and on the oxygen permeation flux. A reduction in the oxygen permeation flux was reported when the sweep gas flux was increased. Mancini and Mitsos [20] introduced a new design of an ITM reactor to be used for production of the required oxygen for the combustion process inside a power plant. Nemitallah et al. [15] presented an optimized monolith structure design of an ITM reactor in order to replace the combustor of a conventional gas turbine. The specifications of this reactor are 3.35 m height, 2700 m^2 total membrane surface area, 10 m^3 total reactor volume, and the reactor can deliver power ranging from 5 to 8 MWe, based on the cycle first law efficiency. Ahmed et al. [21] performed a CFD study of a novel ITM reactor design under oxy-combustion conditions. The novel reactor design in their work resulted in gradual burning of the fuel, and as a result, slow temperature rise and uniform temperature distribution are encountered along the membrane surface. Gunasekaran et al. [22] performed a detailed optimization study on the design and operating parameters of a membrane-based oxy-combustion power plant. The optimization resulted in considerable improvements in the efficiency and emissions of the power plant. Kotowicz and Michalski [23] conducted an efficiency analysis of a supercritical power plant fueled by hard coal utilizing high temperature membranes for air separation. The results showed significant improvement in the net efficiency of the analyzed supercritical power plant. Very recently, Habib and Nemitalah [24] designed an ITM reactor for application in fire tube boilers based on three-dimensional simulations. They came up with a reactor design that can deliver power up to 8 MWe.

Based on the above discussion, many ITM reactor designs were tried for oxygen separation and oxy-fuel combustion in membrane permeate side. However, the button cell ITM reactor design has never been tried for oxygen separation under oxy-fuel combustion conditions. In the present work, the effects of different geometries and flow conditions inside a button cell ITM reactor are investigated experimentally and numerically under oxy-fuel combustion conditions in the sweep side of the membrane.

2. Experimental setup

Fig. 1 shows the test ring facility of the present button cell ITM reactor. The furnace shown in the figure is opened in order to show the membrane reactor assembly. This is a split furnace which is used in order to heat up the membrane and the feed gases to the required operating temperature. The furnace forms an enclosure around the ITM assembly in order to maintain isothermal operating conditions while the measurements. During the heating up and cooling down processes, the heat rate is controlled by a rate of 2 °C/min. Two aligned sets of alumina tubes separated by the membrane are used. Each set has a length of 280 mm and it consists of two coaxial concentric tubes, as shown in the figure. The inner tube has a diameter of D_i = 4 mm, and the gap height between the sweep inner tube exit section and the membrane surface, *P*, is varied between 1 mm and 3 mm.

In this work, some experimental data points are recorded for non-reactive inert cases using helium as the sweep gas. The sweep side gas flows through the inner tube passing in contact with the membrane and then leaves from the annulus between the two tubes. Similarly, feed gas (mixture of oxygen plus nitrogen) enters the ITM unit from the inner top tube inlet section and moves towards the ITM. Portion of oxygen is permeated across the membrane and the oxygen depleted air leaves the system through the annulus between the two tubes. All feed and sweep gases are supplied from compressed gas cylinders, and the flow rates were controlled through mass flow controllers and pressure regulators. The mass flow controllers were connected to a data logger and computer for post processing of the collected data. Different mass flow controllers for different gases provided by Bronkhorest HIGH-TECH were used. All of these controllers are of D-type and model numbers of F202AI-M20-AAD-55-V for oxygen, F-201CI-10K-AGD-