



## Design of an ion transport membrane reactor for application in fire tube boilers



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### ABSTRACT

A design of an ITM (ion transport membranes) reactor is introduced in a two-pass fire tube boiler furnace to produce steam for power generation toward the ZEPP (zero emission power plant) applications. Oxygen separation, combustion and heat exchange occur in the first pass containing the multiple-units ITM reactor. In the second pass, heat exchange between the combustion gases and the surrounding water at 485 K ( $P_{\text{sat}} = 20$  bar) occurs mainly by convection. The emphasis is to extract sufficient oxygen for combustion while maintaining the reactor size as compact as possible. Based on a required power in the range of 5–8 MWe, the fuel and gases flow rates were calculated. Accordingly, the channel width was determined to maximize oxygen permeation flux and keep the viscous pressure drop within a safe range for fixed reactor length of 1.8 m. Three-dimensional simulations were conducted for both counter and co-current flow configurations. Counter-current flow configuration proved its suitability in fire tube boilers for steam generation over the co-current flow configuration. The resultant reactor consists of 12,500 ITM units with a height of 5 m, membrane surface area of 2700 m<sup>2</sup> and a total volume of 45.45 m<sup>3</sup>.

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### 1. Introduction

Many studies have been conducted using different types of ion transport membranes and different experimental and numerical approaches. These studies aimed at understanding the oxygen permeation characteristics through the ion transport membranes. Mixed ionic and electronic conducting ITM (ion transport membranes) are very promising materials for oxygen separation from air at elevated temperatures of the membrane, typically higher than 700 °C [1]. The mathematical formulation for the oxygen permeation mechanism through the ITMs is fairly complex [2]. This mechanism includes the gas phase mass transfer and surface on both surfaces of the membrane and oxygen vacancy and electron diffusion through the membrane bulk [3]. The mechanism of oxygen permeation is dependent on both the membrane surface temperature and the partial pressure difference of oxygen in both

sides of the membrane [4]. The most extensively used membrane materials are the lanthanum cobaltite perovskite ceramics [5]. However, research is continued in order to develop new ceramic membrane materials for many applications including the ITM reactor technology. Those new materials include modified perovskite ceramics, ceramic–metal dual phase membranes, structured ceramic, and thin dual phase membranes including the Pd phases and YSZ (yttria-stabilized zirconia) [6].

The dependence of oxygen permeation flux on the temperature of a disk ceramic membrane was examined by Sunarso et al. [7]. They observed an insignificant oxygen permeation flux at low operating temperatures, lower than 600 °C. On the other hand, they reported considerable oxygen flux at temperatures higher than 650 °C and a sharp increase in the oxygen flux was observed at temperatures higher than 800 °C. Zhu et al. [8] studied the oxygen permeation characteristics of BaCe<sub>0.15</sub>Fe<sub>0.85</sub>O<sub>3-δ</sub> (BCF1585) ceramic membranes which were synthesized by different methods. They observed a strong dependence of the permeated oxygen flux on the operating temperature. Investigations of the influences of the operating surface temperature of the membrane and also the influences of the operating sweep gas flux on the amount of oxygen permeation were experimentally conducted by Zydorczak et al. [9] using ultra-thin La<sub>0.6</sub>Sr<sub>0.4</sub>Co<sub>0.2</sub>Fe<sub>0.8</sub>O<sub>3-δ</sub> ceramic membranes. They

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### Nomenclature

CFD	computational fluid dynamics
CPO	catalytic partial oxidation
EWBM	exponential wide band model
ITM	ion transport membranes
MIEC	mixed ionic and electronic conducting
$P_{sat}$	saturation pressure
$S_i$	source/sink term
$T_{sat}$	saturation temperature
ZEPP	zero emission power plant
3-D	three dimensional
DO	discrete ordinates
F	feed side
$J_{O_2}$	oxygen permeation flux
P	permeate side
RTE	radiative transfer equation
$T_m$	membrane temperature
YSZ	yttria-stabilized zirconia

observed higher oxygen permeation flux at elevated temperatures of the membrane and higher values of sweep gas flow rate. Tan and Li [10] studied the effect of the sweep gas flow rate on the oxygen permeation flux using hollow fiber membranes. They reported a reduction in the oxygen partial pressure in the permeate side of the membrane when the sweep flux is increased under all operating temperatures. This was their justification for the increased oxygen permeation flux when the sweep flux was increased.

From all of the previous works, one can conclude that the oxygen permeation flux is highly affected by both the oxygen partial pressure in the permeate side of the membrane and the operating temperature of the membrane. Based on this fact, two new applications for the ion transport membrane have been developed. The first application is dependent on the idea of reducing the oxygen partial pressure in the permeate side by partial conversion of the fuel using the permeated oxygen in order to produce syngas. In some cases of fuel-rich mixtures in the permeate side and medium operating temperatures, the syngas can be produced. At those conditions, the chances for the production of CO and H<sub>2</sub> are higher as compared to the production of CO<sub>2</sub> and H<sub>2</sub>O. This conversion of the permeated oxygen should result in a reduction in the oxygen partial pressure in the permeate side and an increase in the oxygen permeation flux and the produced syngas is expected [11]. The second application of the ion transport membrane is dependent on the idea of increasing the operating temperature of the membrane by burning the fuel with the permeated oxygen (oxy-combustion technology) just beside the membrane surface in the permeate side of the membrane. This application is called the ion transport membrane reactor technology which is one of the most promising carbon capture technologies toward controlling the greenhouse gases emissions. The combination of both the oxygen separation and the fuel combustion into a single unit called ITM reactor should result in a reduction in cost and size of this promising carbon capture technology [12]. Recently, many research works have been conducted in order to reform methane into higher hydrogen content by Fischer Tropsch synthesis [13]. One of the most promising methods is the use of dense MIEC (mixed ionic and electronic conducting) ceramic membranes. The permeated oxygen is used in a CPO (catalytic partial oxidation) reaction in order to reform methane into CO plus H<sub>2</sub> (syngas) [14]. This process results in a targeted product ratio of hydrogen to carbon monoxide of 2, which is suitable for Fischer Tropsch synthesis [15]. Many recent research

works are concentrating the effort in order to develop and improve the performance and stability of the MIEC membrane materials, which are suitable for use under reaction conditions in the permeate side of the membrane [15]. In reacting medium conditions, high values of oxygen permeation flux were reported by many researchers. They used different ceramic membrane materials like Ba<sub>0.5</sub>Sr<sub>0.5</sub>Co<sub>0.8</sub>Fe<sub>0.2</sub>O<sub>3- $\delta$</sub>  (BSCF) [16], SrCo<sub>0.8</sub>Fe<sub>0.2</sub>O<sub>3- $\delta$</sub>  (SCF) [17], and La<sub>0.6</sub>Sr<sub>0.4</sub>Co<sub>0.2</sub>Fe<sub>0.8</sub>O<sub>3- $\delta$</sub>  (LSCF) [18].

Nowadays, the global climate changes due to the emissions of greenhouse gases, mainly CO<sub>2</sub>, forced the researchers toward more improvement in the carbon capture technologies. Oxy-fuel combustion technology is one of those carbon capture technologies through the modification in the combustion process. This modification is through the combustion using oxygen instead of air which should result in an increase in CO<sub>2</sub> concentrations in the exhaust stream, so that it can be easily captured. In order to avoid the high temperature operation, oxygen is mixed with the recycled flue gases [19]. Nowadays, Most of the research works are focused on the application of the oxy-fuel combustion in the permeate side of the membrane. Those studies aimed at the understanding the oxygen permeation and the oxy-fuel combustion characteristics inside ITM reactors. Nemitallah et al. [20] Performed numerical investigations on an LSCF ITM reactor trying to characterize the membrane performance under oxy-fuel combustion conditions using a modified two-step oxy-combustion reaction kinetics model for CH<sub>4</sub>. They reported sharp increase in the oxygen permeation flux when the reactions were activated in the permeate side of the membrane. Similar results were reported for the same membrane material by Ben-Mansour et al. [21] and Ben-Mansour et al. [22] using BSCF membrane material. An experimental study coupled with mathematical formulations has been conducted by Akin and Jerry [23] in order to examine the influences of sweep gas flux and reactivity in the permeate side on the oxygen flux. They showed a reduction in the oxygen permeation flux when the sweep gas flux was increased due to the lowered reaction rates of the fuel at these conditions. More details about the progress of the ITM reactor technology can be found in our recent review work, Ref. [24].

Recently, very few number of research works have been conducted in order to apply the ITM reactor technology into large scale power plants aiming at real application of the ZEPP (zero emission power plant) concept. The ITM reactor technology is supposed to replace the present cryogenics and decrease the cost of oxygen production by about 35% [25]. This cost reduction can result in a 50% reduction in the energy required for CO<sub>2</sub> capture, when the ITM reactor is integrated into power plants. For ZEPP applications, high oxygen mass flow rates are required which forced the research in this direction in order to maximize the ratio between the membrane surface area and the total volume of the reactor. Nemitallah et al. [26] presented a design for an ion transport membrane reactor for the substitution of a conventional gas turbine combustor. The ITM reactor in their work is monolith structure design using LSCF1991 membrane type. Optimizations for mass flow rates, channel geometry and percentage of the fuel in the sweep gas have been performed. They come up with an ITM reactor with a height of 3.35 m, membrane surface area of overall 2700 m<sup>2</sup>, total reactor volume of 10 m<sup>3</sup> and output power of 5–8 MWe. Another design of an ITM reactor was introduced by Mancini and Mitsos [27] in order to produce the required oxygen for combustion inside the reactor for power plant applications. The ITM reactor in their study is able to deliver a power of 300:500 MWe based on the cycle first law efficiency.

In those two studies for ITM reactor application for large scale power generation [26–28], the oxygen permeation equation was scaled up by one order of magnitude. This was attributed to the recent increase in the oxygen permeation flux due to the

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