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Oxygen integration between a gasification process and oxygen production using a mass exchange heuristic

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ABSTRACT

In this work we analyze different design alternatives for the integration of a gasification process with the oxygen production process, through ITM membranes. We analyze the conventional separation design compared with a novel configuration in a countercurrent arrangement with sweep gas (using the gas permeation module as a mass exchanger). To assess the oxygen transfer in the permeation modules, they are modeled with Aspen Custom Modeler V8.4 and the different design alternatives are simulated in Aspen Plus V8.6. The economic analysis carried out shows that the counter-current arrangement with a sweep stream has a Total Annualized Cost 13.5% lower than the conventional separation design.

Introduction

In previous papers we worked on hydrogen exchange between different streams of the same process [1–4] using membrane modules in countercurrent arrangements as mass exchangers. This was done at different stages of the process design procedure, achieving significant reductions in both the consumption of hydrogen and the compression energy to recycle the recovered hydrogen. These results suggested a heuristic rule that can be applied at different stages of the hierarchical process design methodology by Douglas [5]: Instead of using gas permeation modules to separate hydrogen from a gaseous stream and then recompress this hydrogen to recycle it to the process, it may be convenient to exchange hydrogen between process streams, without spending energy in the recycle compressor. In the gas separation by semi permeable membranes, a trans membrane pressure difference is applied as the driving force for hydrogen transport. Furthermore, in laboratory practice it is common to use a sweep gas in the low-pressure side of the membrane, to reduce the partial pressure of hydrogen (or any other permeating component), increasing the trans membrane partial pressure difference [6]. This practice has the disadvantage that the permeating component (e.g. hydrogen), needs to be afterwards separated from the sweep gas to be reused in the process. Therefore, the sweep gas is generally selected such that it can be readily separated from the

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permeate component, to avoid a costly new separation. Moreover, the product recovered is usually at low pressure, so to reuse it in the process must be recompressed up to the working pressure. These complications explain why the use of a sweep gas is not common within the industry, and it is mainly limited to laboratory practice. Another common technique to increase the trans membrane difference of partial pressure is to apply vacuum in the low-pressure side, but if the permeate component must be afterwards compressed to the process working pressure, extra energy is needed for this. Application of the design heuristic rule above mentioned, would exchange between process streams in a countercurrent arrangement, without the addition of a new sweep gas stream or using vacuum systems. That is using as a carrier gas, an input stream of the process (free of the component to be recovered) already compressed to the working pressure: this avoids any further separation, nor recompression. This concept of exchange generates new and interesting process design options, which need be carefully analyzed to determine the most suitable. In this paper, we will leverage the benefits of the concept of countercurrent exchange between process streams, to integrate different processes, focusing on new options for possible integration between oxygen production processes and a gasification process using a moving bed gasifier that consumes oxygen, using ITM membranes (Ion Transport Membranes) to transfer the oxygen.

Following, there is a brief overview of the gasification and the oxygen separation processes. Subsequently we describe the main features of the model used for the numerical evaluation. Afterwards we assess different separation systems: the conventional separation system design using ITM membrane and the separation system arrived at by using the ITM membranes in a countercurrent arrangement with a sweep stream. Next, we analyze the economic impact of adopting the proposed new design and compared it with conventional separation designs. Finally, we draw the conclusions of this work.

Overview

Overview of gasification

For different gasification processes it is possible to use either air or oxygen to oxidize the feedstock. The processes that use oxygen, have the advantage of requiring smaller equipments, spend less energy and therefore be less expensive; nevertheless they have the disadvantage of the costs associated with the oxygen separation from air. It is usual that the pressure at which the gasification is performed be between 25 and 35 bar [7,8], especially if the products are to be used for power generation, e.g. in any of the possible alternatives of IGCC (Integrated Gasifier Combined Cycle). Moreover, many chemical processes are performed at high pressures over 70 bar, and even exceeding 250 bar. These processes require huge amounts of energy for compression, so it deserves careful determination of in which stage of the process the gaseous streams must be compressed to such high pressure. Normally, it is desirable to compress the raw materials at low temperature and before gasifying, since in this way the gas volume is smaller, with considerable savings in the compression energy demand, in the order of about 77.6% [8], although this argument is not valid for very high pressures (70–100 bar), since the gasification becomes impractical for equipment reasons. For processes carried out at high pressures and temperatures, it is particularly important to separate the oxygen from the air before the compression, to lower the compression energy since the nitrogen is then not compressed (unless this were useful, as in the production of ammonia [9]).

Overview of oxygen separation

The most commercially suitable alternative for obtaining oxygen, has long been the cryogenic distillation of air [10]. The separation of oxygen amounts to a significant percentage of the final cost of the product (either a chemical or energy): between 10 and 21% [11]. And if this oxygen has to be used at high pressures, it is worth find out if it is advantageous pumping it in the liquid state, or compressing it at the gaseous state [8]. Here again, the alternatives must be studied carefully.

The tradeoff between air and oxygen is as follows. The simplest and least expensive source of oxygen for gasification is compressed air, however this introduces nitrogen and argon that increase the size of all downstream equipment in the gas loop and additional oxygen is required to raise these inerts to the reaction temperature. All current large-scale industrial applications of Fischer-Tropsch technology use pure oxygen for syngas production [9]. If an air separation unit ASU, typically cryogenic air distillation, is adopted, the oxygen purity is typically 95%. The main impurity is argon, with smaller quantities of nitrogen. A portion of the syngas is often burned to generate electric power to operate the ASU [12].

Presently, ITM (Ion Transport Membrane), or also named MCM (Mixed Conducting Membrane) have been developed, that efficiently perform the oxygen separation from air [13,14]. It is reported that these membranes get a cost reduction of about 31% if they are properly integrated with gasification in a IGCC (Integrated Gasificator Combined Cycle) [15]. If these membranes are integrated in a CCGT (Combined Cycle Gas Turbine) the cost reduction is about 6% respect to a CCGT with carbon capture and secuestration [16].

The ITM membranes were proposed by Researchers of Air Products and Chemicals Inc. [10,15,17], to be used as membrane separators (the most simple configuration), as membrane separators using a sweep gas, or as membrane reactor for the production of syngas (CO + H2) or combustor (or burner) (CO2 + H2O). As ITM membranes work at high temperature and a considerable pressure is necessary, it is usual that for adequate performance ITM membranes work integrated to a gas turbine in a combined cycle for the generation of power. Air Products and Chemicals Inc. is close to the industrialization of ITM membranes.

Other researchers such as Yantovski et al. [18,19], Möller et al. [16], Foy and Yantovski [20], developed zero emission cycles based on these membranes. Yantovsky et al. [18,19] propose the use of ITM membranes as separators using sweep gas for a so-called advanced zero emission power cycle (AZEP). They proposed to use part of the combustion residue (CO2 + H2O) as a sweep gas to enhance the partial pressure gradient through the membrane, to reduce the requirement of membrane area. These cycles that use the combustion products have the important drawback of continuously recycling inert elements, with considerable consumption of compression energy (and separation in some cases), along with the need for a separator of greater volume (even if the required area be the same) with its corresponding extra cost. Furthermore (this may be the worst drawback), the recycle of combustion residue precludes the direct use of this process alternative from any other application but the generation of heat for power cycles (e.g. precludes its use for the generation of hydrogen to produce chemicals).

Möller et al. [16] also propose the use of ITM membranes as separators using a sweep gas. They define a set of equipment (ITM membrane, combustion chamber, several heat exchangers, mixers and splitters) as a MCM reactor. Some of the products of combustion (at high temperature) are recycled and used as sweep gas in the ITM membrane. The set of equipment does not consider the recycle compressor, even if it should certainly be considered given the importance of the costs of capital investment and energy consumed to operate it. The MCM reactor is used in different configurations achieving 85–100% AZEP cycles that renders high efficiencies and reduced cost of 6% compared to a CO2 recovery system afterburners [16]. To avoid rapid deterioration of the ITM membrane the working temperature is limited to 1200 °C. Yantovsky et al. [18] refer to these drawbacks.

While in the referenced work the ITM membrane is used with sweep gas at the low pressure side, the sweep gas is a product of combustion which is recirculated continuously between the inlet and the outlet, with considerable costs associated with the recycle compressor. The arrangements on which we have worked on, do not have the drawback that we are discussing here. The sweep gas that we propose to use in the ITM membrane is a raw material input to the process, not a product (or co-product) output of the process, thus, it does not need to be recycled. This difference is the main point of our proposal, since in this way we avoid costs associated with recycling, while still having the benefit of a greater driving force through the ITM membrane.

Model

We used Aspen Custom Modeler 8.4 (ACM) to model both the conventional and the counter current arrangements for using the ITM membrane permeate module with sweep gas. Using ACM for the aforementioned modules has some advantages, among them that the modules can be exported to be used in Aspen Plus, allowing us to simulate the whole process in the same simulation package, without resorting to external software, which would complicate the simulation and convergence. In addition, our custom modules utilize the physical properties taken from the database of Aspen Properties, using predefined models for estimating the physical properties (physical property model in Aspen Plus V8.7). Thereby all the properties determined from the equation of state (such as the enthalpy, entropy, volume, pressure and temperature, mole fraction, etc.) are much more accurate.

Oxygen transport across ITM membranes is a complex phenomenon and has been studied for many years. In order for oxygen transfer to take place, different phenomena occur, such as polarization layer, surface reaction, bulk diffusion, etc. [21,22]. As an approximation, ussually valid for low permeate fluxes, only the bulk diffusion phenomenon is considered, represented by Wagner equation [23]. But actually, the performance of the permeation is very dependent on the conditions of temperature, pressure and gas composition at each side of the membrane, and in practice it is normally difficult to determine all parameters for an accurate modeling of the oxygen transfer, since the number of phenomena occurring simultaneously is large [22]. For our ITM membrane modules, we use the permeances reported by Baumann et al. [22] for ITM BSFO supported membranes and with an activation layer. Baumann et al. [22] propose that oxygen transfer through the membrane is limited by several resistances in series, which become important or not, depending on operating conditions. However, at the operating conditions considered in our flow sheets the oxygen transfer is dominated by the bulk diffusion phenomenon. There are different models in use for this oxygen transfer [15], in this paper we use the one described in Stadler et al. [24] shown in equation (1). In this equation j_{O_2} is the oxygen permeation flux; d is the membrane thickness; T is the membrane temperature; C_{Wagner} is the Wagner pre-exponential factor; K_{Warner} is the Wagner constant (related to the activation energy); $p_{O_2,feed}$ and $p_{O_2,permeate}$ are the oxygen partial pressures (in bar) up and downstream of the membrane respectively.

$$j_{O_2} = \frac{C_{Wagner} \cdot T}{d} \cdot e^{\left(\frac{-K_{Warner}}{T}\right)} \cdot \ln\left(\frac{p_{O_2,feed}}{p_{O_2,permeate}}\right) \tag{1}$$

For our study, we take both temperature and thickness constant, which simplifies the equation and eases using it to correlate experimental data. We correlated the permeances reported by Baumann et al. [22] usign this simplified equation, obtaining $\frac{C_{Wagner}}{d}$ 0.01881 (kmol/hr.m^2.°K), and K_{Warner} 5780 °K.

We modeled the permeate module for conventional cross flow and for the countercurrent arrangement with sweep stream. Figs. 1 and 2 show a schematization of both configurations.

The total area of the permeation module is subdivided into a number N of cells. For each of these cells the oxygen transfer is computed by considering the average driving force between the input and the output of the cell, achieving a good representation of the phenomenon, with a small number of cells (achieving quick convergence simulations). Fig. 3 shows a schematization of the cells for conventional cross flow and for countercurrent arrangement with sweep stream.

Case study: integration of the oxygen separation process with the gasification process

In this paper we use the model of a moving bed coal gasifier by AspenTech [25], which represents a case taken from the literature [26]. A Scheme of this gasifier is show in Fig. 4. We integrate this gasifier with the oxygen production process using ITM membrane in both the traditional separation

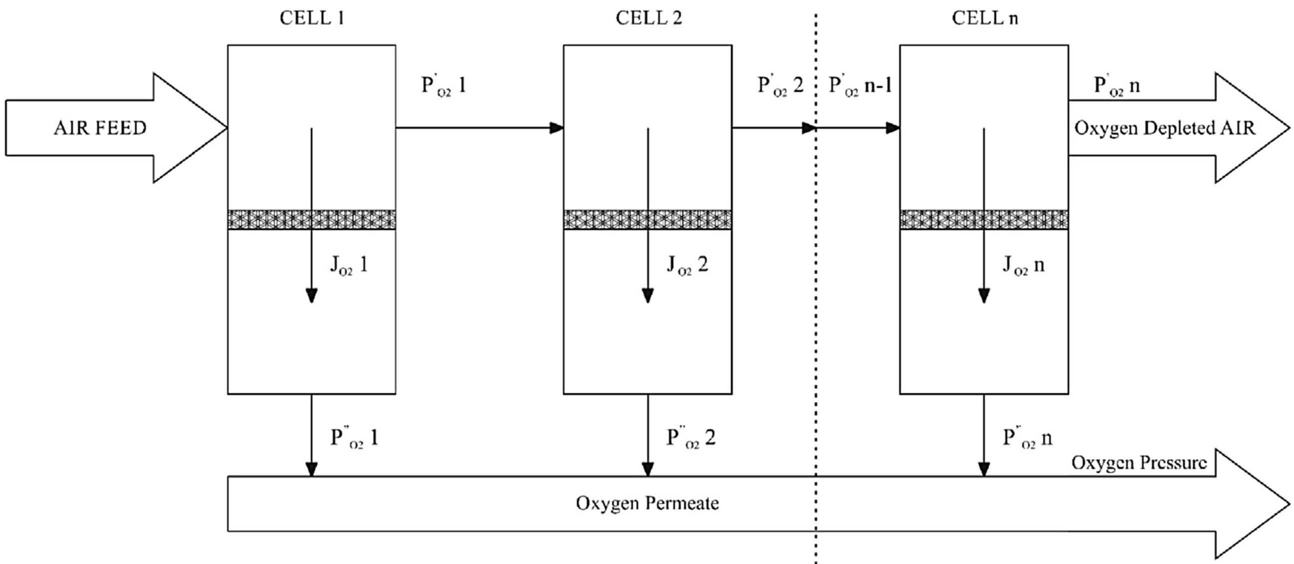

Fig. 1 – Scheme of permeate module for conventional cross flow.

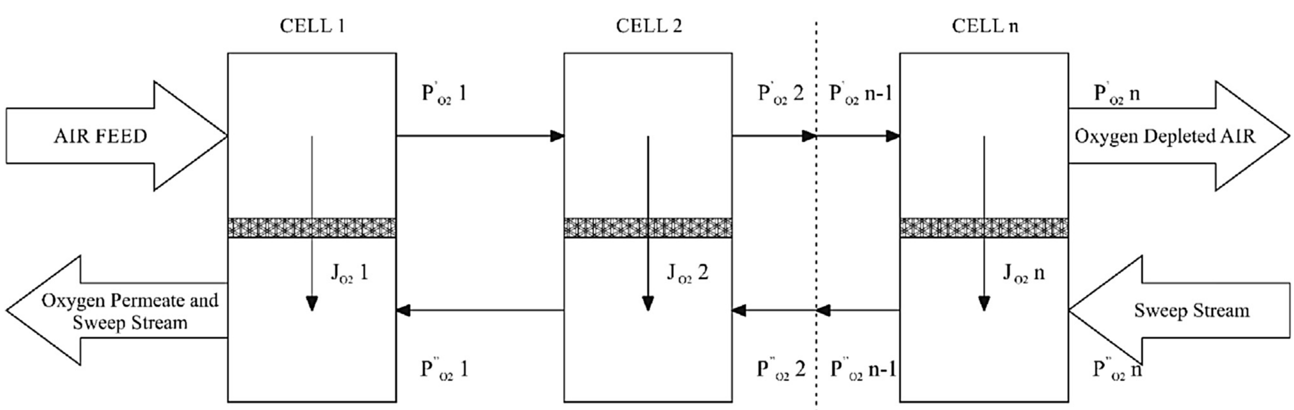

Fig. 2 – Scheme of permeate module for countercurrent arrangement with sweep stream.

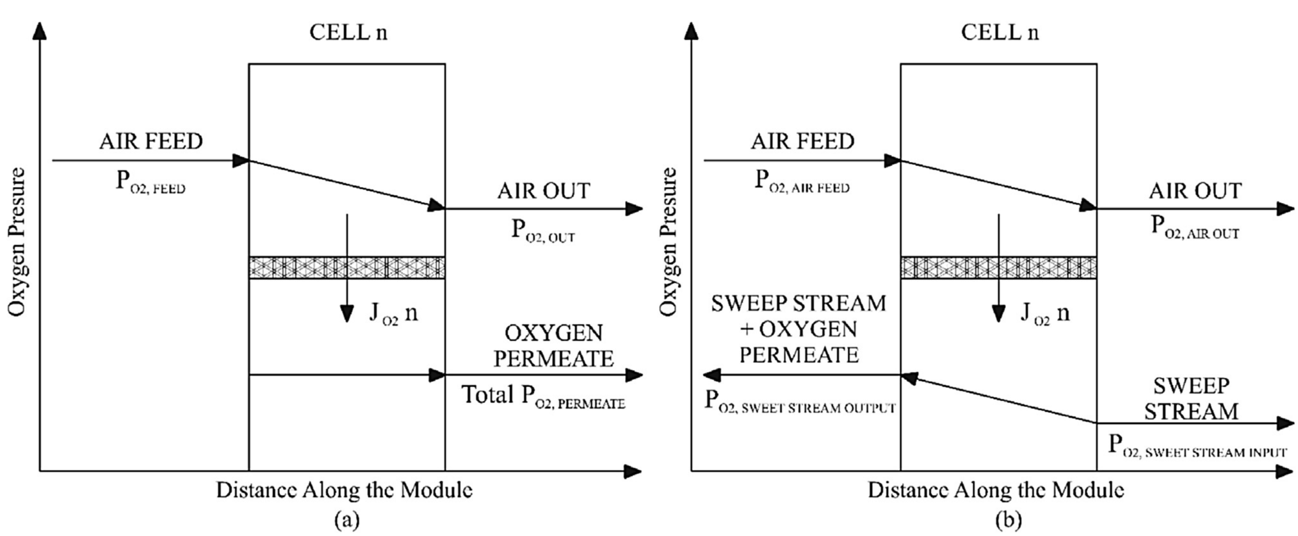

Fig. 3 – (a) Scheme for each Cell in Conventional Cross Flow. (b) Scheme for each Cell in Countercurrent Arrangement with Sweep Stream.

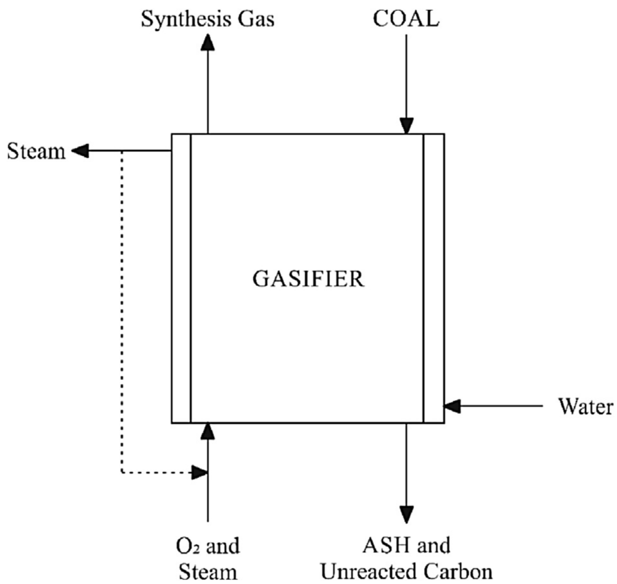

Fig. 4 – Scheme of moving bed coal gasifier.

scheme (one inlet and two outlets), and using the separation scheme in counter-current arrangement with the sweep gas (mass exchanger: two inlets and two outlets). The gasifier is fed with 11,052.97 kg/hr (345.42 kmol/h) of oxygen, 19,597.00 kg/hr of bituminous coal, and 55,851.73 kg/hr (3100.24 kmol/h) of steam. It produces 85,114.33 kg/hr (4412.542 kmol/h) of synthesis gas with a molar fraction of 0.1203 and 0.1588 for carbon monoxide and hydrogen respectively, while the mole fraction of water is 0.56. Table 1 presents the main features of this gasifier, while Tables 2 and 3 present the input and output conditions. The steam is produced in the jacket of the gasifier at a temperature of 371.11 °C and a pressure of 35.024 atm. This steam is mixed with oxygen (also at a pressure of 35.024 atm) and sent to the bottom of the gasifier while the coal is supplied at the top. Moreover, the synthesis gases produced are removed from the top of the gasifier.

We took the operating conditions reported in the literature, without any modification, as we are only interested in characterizing the oxygen integration. As we did not modify any of the input nor operation conditions, the synthesis gas produced after carrying out the integration is the same as before of it.

Given the large number of feasible operations and applications, there are many possible design alternatives to achieve integration. In this paper, we choose the application of generating hydrogen for later use in the production of

Table 1 – Characteristics of the gasifier.

Parameter	Value	Unit
Height	2.32	m
Diameter	3.66	m
Pressure	35.024	Atm
Wall temperature	371.11	°C

Table 2 – Conditions of feedstocks.

Feedstock	Flow rate (kg/hr)	Temperature (°C)	Pressure (atm)
Coal	19,597.00	25	35.024
Pure oxygen	11,052.97	371.11	35.024
Steam	55,851.73	371.11	35.024

Table 3 – Product gas composition.

Product gas composition (dry basis, mol.%)						
CO	H_2	CO_2	CH_4	H_2S	N_2	C_6H_6
28.57	37.70	21.98	9.00	0.28	1.72	0.75

chemicals, so we used design alternatives related to this purpose. Even though the separation process is performed at high temperature, it is necessary to consider the residual heat recovery from the effluents. Power generation is the most studied option, because usually the ITM membranes are intended to be used in power generation plants, although the thermal energy of the effluent could be used in any process that needs it. In this paper we consider heat recovery to generate steam at different pressures that could be used in steam turbines, but do not consider any particular process of power generation (e.g. Rankine cycle) because it is not the aim of this study. To facilitate comparison between alternatives, we seek that they be as similar as possible, and essentially differ in the use of the sweep stream in either the conventional or the countercurrent arrangement. Figs. 5 and 6 present the flow sheets for the conventional configuration (the membrane is used as a separator) and for the configuration in countercurrent with the sweep stream (the membrane is used as a mass exchanger).

For the conventional configuration, it can be seen that the air is fed to the compressor at room temperature and pressure (25 °C and 1 atm). The compressor increases the pressure of air which causes a significant improvement in the driving force across the membrane, so the membrane area required is reduced. After adiabatic compression the air is heated, but not enough to reach the operating temperature of the ITM membrane (typically 750–1000 °C), so it is necessary to further increase the temperature. In the traditional configuration, we chose to use the heat of the oxygen permeate to preheat the air, and afterwards heat with a direct fired heater (the most simplest alternative) up to the selected operating temperature. We selected an operating temperature of 900 °C because at this temperature the membrane has a good permeability without substantial degradation, that normally occurs at temperatures higher than 1200 °C [18]. The operating conditions are also lower than those usually used in gas turbines (1500 °C) and gasifiers (up to 2000 °C) [12], and they should not present operational problems. Moreover, as the temperature variations produced by the oxygen transfer are minimal, we use the simpler isothermal custom models which consider that the temperature is constant along the ITM membrane (to facilitate comparison between the proposed alternatives).

In the configuration with sweep stream in countercurrent only a direct fired heater is used, since the energy of the sweep stream plus the permeate oxygen is used for reheating the

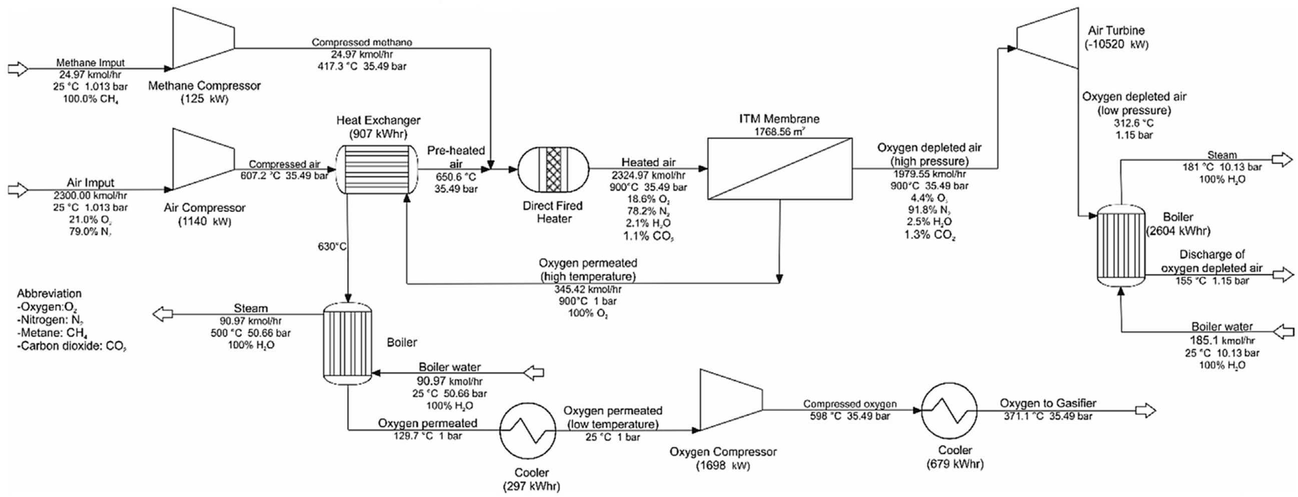

Fig. 5 – Flow sheet for conventional configuration (separator).

steam from the gasifier as they have similar MCp (mass multiplied by heat capacity at constant pressure). In both configurations, given the large volume of the air and the large compression powers, we adopted axial compressors, as they are more efficient than centrifugal compressors for large flow rates and powers. Furthermore, the methane (used in the direct fired heater) is fed at atmospheric pressure and compressed to the same pressure of the air with a centrifugal compressor driven by an electric motor, since the power required is much smaller. The oxygen depleted air exiting from the permeate module is at high pressure and temperature, and to take advantage of this remaining enthalpy, it is used in axial turbines for both configurations. As both the powers of compression and expansion are similar within the operating range (with a variation according to the flow, the pressure, the temperature and the mole fraction of residual oxygen), both units may share the same main shaft and are associated with a moto-generator group. This configuration (compressor-turbine generator) is similar to a gas turbine and is studied by several authors [10,18,24]. For low air flow rates, the turbine generates less power than is consumed by the compressor, whereas for higher air flow rates this is reversed. The air after passing through the turbine is still at high temperature. This energy is used to generate medium pressure steam (10.13 bar) which can then be used in a low pressure steam turbine. In both alternatives, the variables: air flow rate, air pressure, mole fraction of residual oxygen in the air, and the pressure of the permeate stream (only in the conventional configuration), were considered design variables and are determined in each case by optimization of the economic performance of the whole process. These variables are closely related to each other and with the performance of the ITM

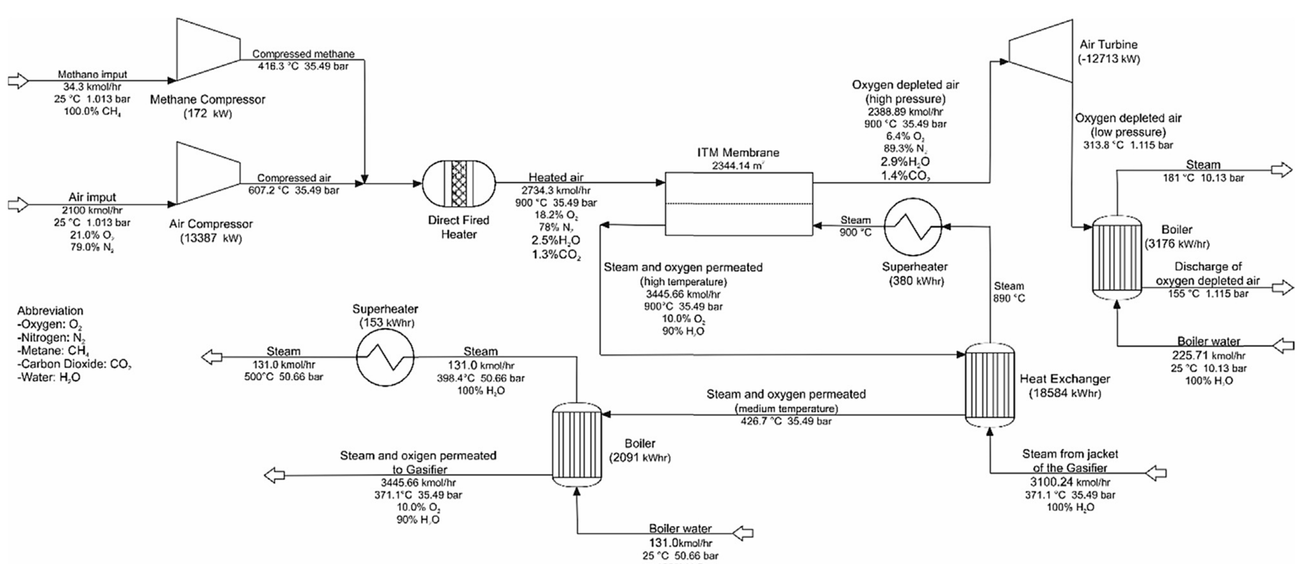

Fig. 6 – Flow sheet for counter-current configuration with sweep stream (mass exchanger).

membrane, and are studied in detail later. For the countercurrent configuration with a sweep stream, the pressure of the sweep stream (steam) is fixed by the gasifier that generates it (in the jacket) [26]. As shown in the flowsheets, the main difference between both alternatives is that for the conventional configuration it is necessary to use a compressor to raise the pressure of the permeated oxygen up to the inlet pressure of the gasifier, while for the countercurrent configuration with sweep stream the compressor is not necessary, since the sweep stream (plus oxygen permeated) is already at the input pressure of the gasifier. Another difference (although with less economic impact) is that the heat in the permeated oxygen stream are recovered differently, due to the reasons previously founded (the MCp of sweep stream and the sweep plus oxygen are similar). In the conventional alternative, given the conditions of temperature and pressure needed to feed the oxygen to the gasifier, and that it is desirable to compress a gas at low temperature (to decrease compression power), it is necessary (an extra task) to cool the permeate stream both before and after compressing it. The recovered energy is used to generate some extra steam.

In both alternatives, the amount of methane combusted in the direct fired heater is controlled to achieve the desired temperature (900 °C) for the permeation module. The Methane compressor operates at a pressure equal to the oxygen compressor. In turn, to take advantage of the residual energy of the oxygen depleted air, the amount of water fed to the boiler is controlled to achieve a steam at a pressure of 10.13 bar and a temperature of 181 °C.

The heat recovery scheme for the heat of the permeate oxygen (in the traditional configuration), and heat of oxygen permeated plus steam (in the countercurrent configuration), differs slightly given the conditions of the streams available. For the conventional configuration, the amount of water that feeds the boiler is controlled, achieving a superheated steam at a pressure of 50.66 bar and a temperature of 500 °C. For the configuration in the countercurrent arrangement with sweep stream, the amount of water fed into the boiler is controlled, achieving a superheated steam at a pressure of 50.66 bar and a temperature of about 398 °C. The remaining increase in temperature up to 500 °C is achieved taking advantage of the waste gases of the furnace to reheat the steam from the gasifier.

Conventional operation of the ITM membranes

For this case study we have as a parameter (a fixed figure) the amount of oxygen needed by the gasifier, and as design variables (to be optimized): the pressure above and below the ITM membrane, the air flow rate and the membrane area required. However, we cannot freely vary these variables because they are closely linked with the performance of the ITM membrane, and the amount of oxygen that we need to permeate. For example, the minimum air flow rate must contain more oxygen than the amount we want to permeate, and in turn, at the end of the permeate module, we must still have a positive partial pressure gradient (driving force). Therefore, the minimum air flow rate depends on the amount of oxygen that we want to permeate and the pressures both at the high and low-pressure side. While the minimum flow rate is constrained by the above considerations, the maximum flow rate (and maximum pressure at the high-pressure side) are determined by economic considerations.

The gasifier needs 345.42 kmol/h of pure oxygen. Figs. 7–9 show the variation of the oxygen partial pressure (in the high-pressure side at 35.49 bar of total pressure) in each cell along

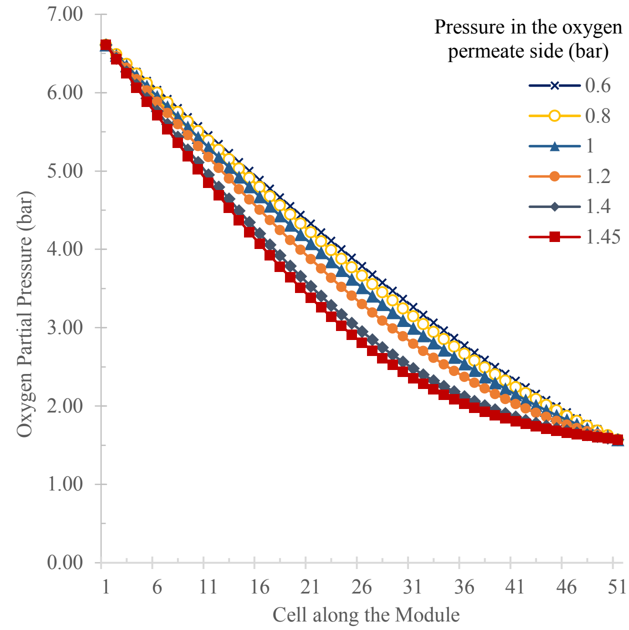

Fig. 7 – Variation of the oxygen partial pressure in the high-pressure side in each cell along the permeate module when air flow rate is 2300 kmol/h for different pressures in the oxygen permeate side.

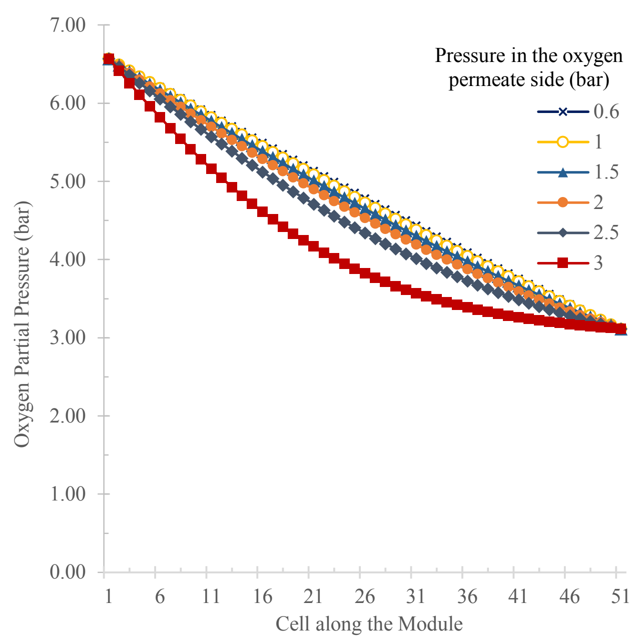

Fig. 8 – Variation of the oxygen partial pressure in the high-pressure side in each cell along the permeate module when air flow rate is 3200 kmol/h for different pressures in the oxygen permeate side.

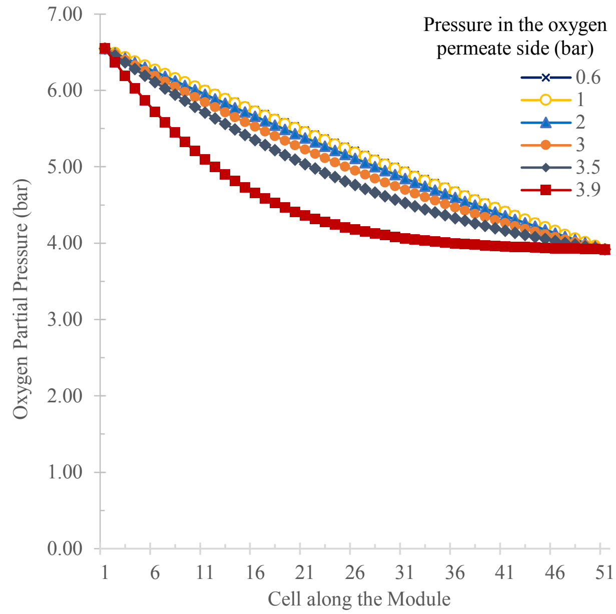

Fig. 9 – Variation of the oxygen partial pressure in the high-pressure side in each cell along the permeate module when air flow rate is 4100 kmol/h for different pressures in the oxygen permeate side.

the permeate module when air flow rates are 2300, 3200 and 4100 kmol/h, for different pressures in the oxygen permeate side.

In turn, in Fig. 10 it can be seen how the area required varies when air flow rates are 2300, 3200 and 4100 kmol/h, by varying the pressure in the oxygen permeate side.

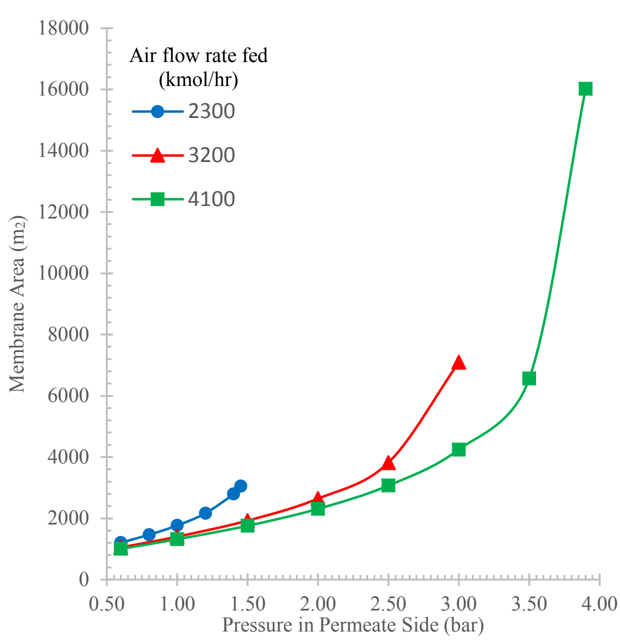

Fig. 10 – Membrane area required when air flow rates are 2300, 3200 and 4100 kmol/h, by varying the pressure in the oxygen permeate side.

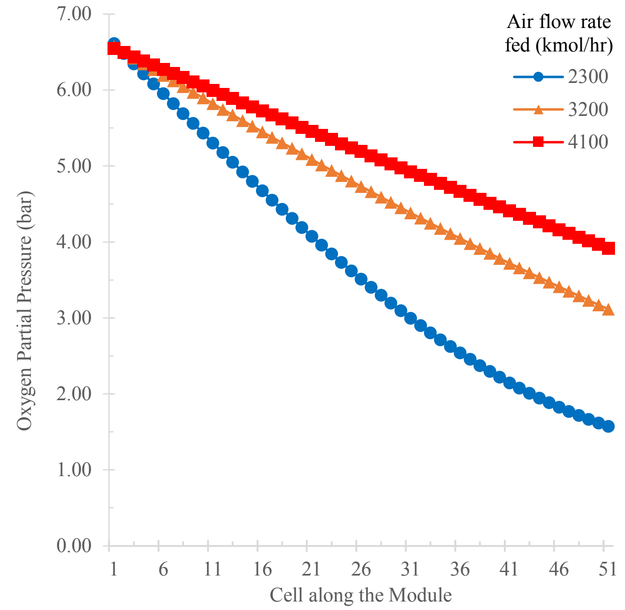

Fig. 11 – Variation of the partial pressure of oxygen in the high-pressure side in each cell along the permeate module when the pressure at the permeate side is of 1 bar, for the different air flow rates fed.

By inspection of Figs. 7–9, it may be noted that increasing the air flow rate fed to the permeate module, the oxygen depleted air output from the permeate module has a larger oxygen partial pressure. For the sake of clarity, Figs. 11–13 show how the partial pressure of oxygen in the high-

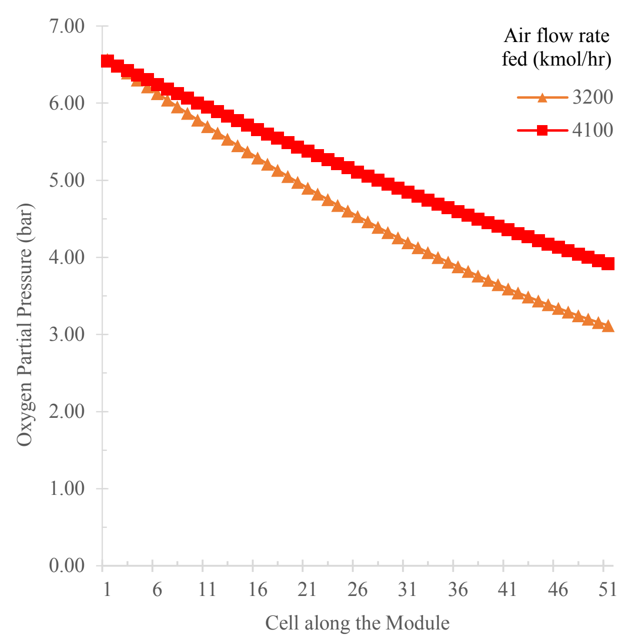

Fig. 12 – Variation of the partial pressure of oxygen in the high-pressure side in each cell along the permeate module when the pressure at the permeate side is of 2 bar, for the different air flow rates fed.

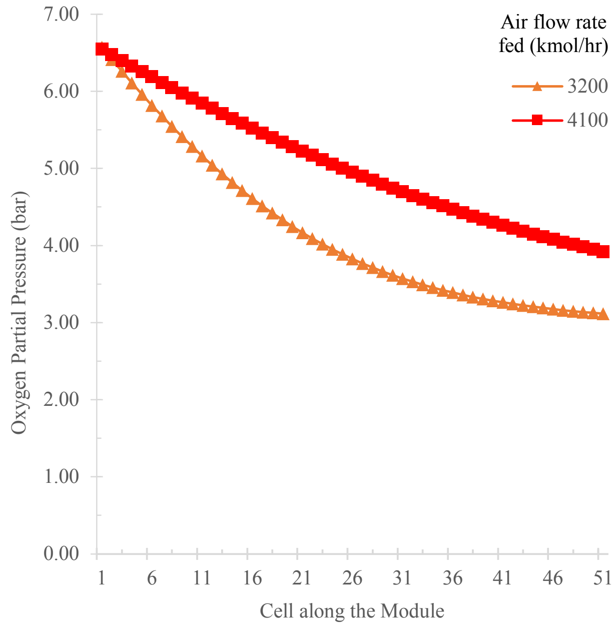

Fig. 13 – Variation of the partial pressure of oxygen in the high-pressure side in each cell along the permeate module when the pressure at the permeate side is of 3 bar, for the different air flow rates fed.

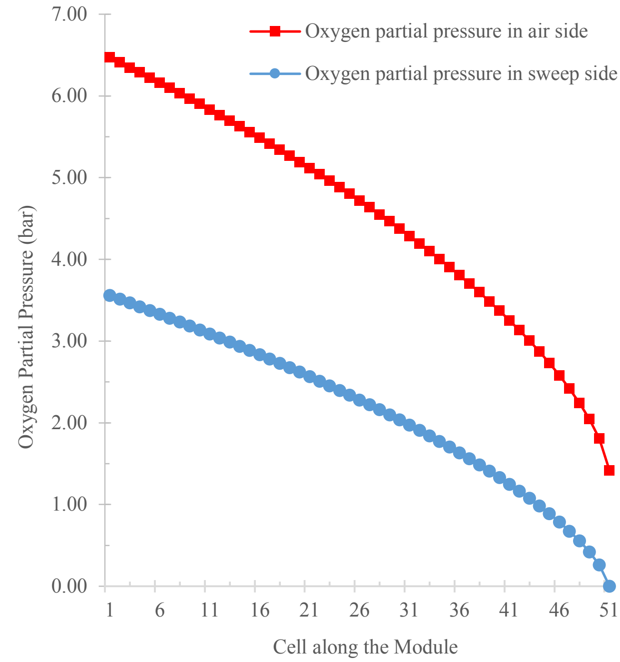

Fig. 14 – Distribution of the oxygen partial pressure for both the air and sweep streams, in each cell along the permeate module when air flow rate is 2300 kmol/h.

pressure side varies in each cell along the permeate module when pressures at the permeate side are of 1, 2 and 3 bar, and when the amount of air fed is 2300, 3200 and 4100 kmol/h.

ITM membranes in countercurrent arrangement with sweep stream

For the permeate module in counter-current arrangement the considerations for the inlet air stream are similar, but differ in that the partial pressure of oxygen (in the air) at the outlet of the permeate module can be smaller. This configuration can recover more oxygen from the air because exchanging in counter current with a stream free of oxygen, the partial pressure at the inlet of the permeate module is zero. The differences are accentuated when considering that the steam stream must be able to receive all the oxygen (345.42 kmol/h) without the partial pressure at the outlet of the permeate module exceeding the partial pressure of oxygen in the inlet air stream. This means than for the counter current arrangement, there is also a minimum flow rate for the sweep stream (steam). Steam flow rate becomes a design variable: increasing partial pressure difference across the membrane (increasing the steam flow rate) results in a smaller area of membrane required. Furthermore, for the case study that we propose, the gasifier requires a fixed amount of steam preset by the operating conditions, so that we also have a maximum steam flow rate. The gasifier needs a steam stream with a flow rate of 3100.24 kmol/h at 35.024 atm of total pressure. Figs. 14–16 show the distribution of the oxygen partial pressure for both the air stream and sweep stream, in each cell along the permeate module when air flow rates are 2300, 3200 and 4100 kmol/h, for a total pressure of 35.49 bar at both sides of the ITM membrane. Additionally, Fig. 17 shows the amount of the oxygen permeate in each cell along the permeate module when air flow rates are 2300, 3200 and 4100 kmol/h.

By inspection of Figs. 15–17, it may be noted that increasing the air flow rate fed to the permeate module, the oxygen depleted air has a larger oxygen partial pressure and

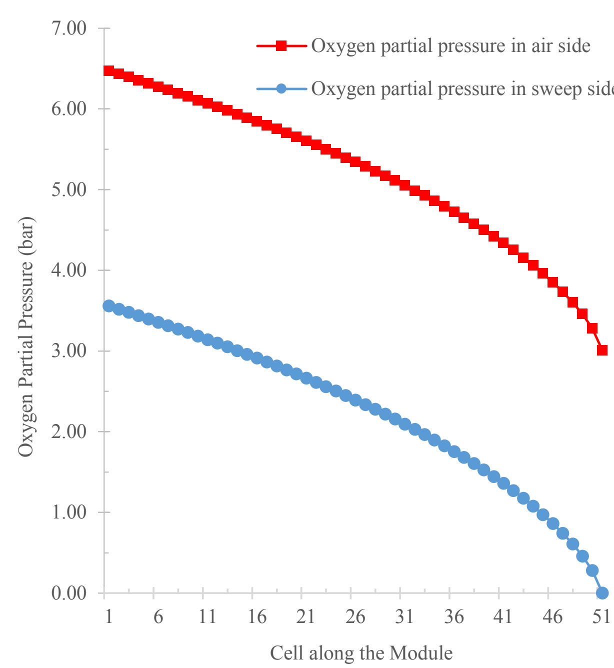

Fig. 15 – Distribution of the oxygen partial pressure for both the air and sweep streams, in each cell along the permeate module when air flow rate is 3200 kmol/h.

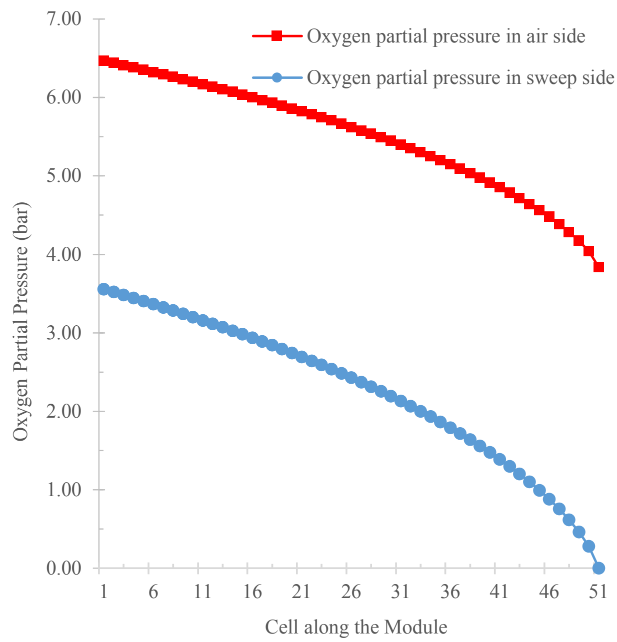

Fig. 16 – Distribution of the oxygen partial pressure for both the air and sweep streams, in each cell along the permeate module when air flow rate is 4100 kmol/h.

the oxygen partial pressure difference is larger along the permeate module.

Economic assessment

In order to compare the economic performance of the mass exchanger in a countercurrent arrangement vs in a conventional separation, we computed the Total Annualized Cost

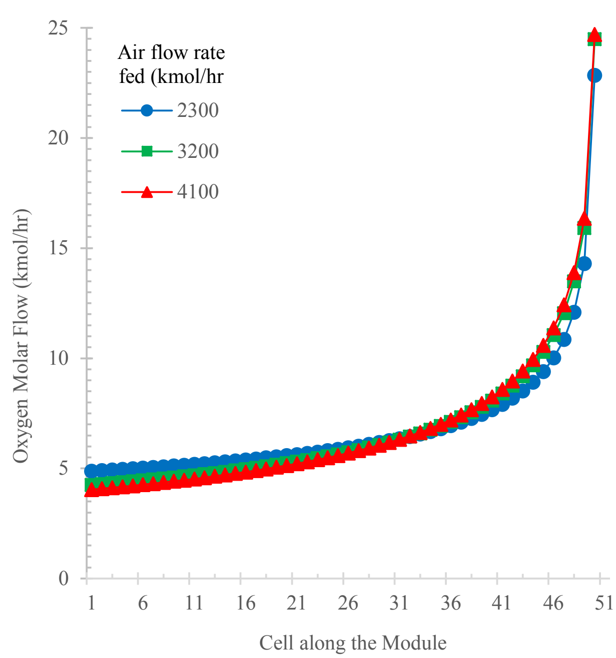

Fig. 17 – Amount of oxygen permeated in each cell along the permeate module when air flow rates are 2300, 3200 and 4100 kmol/h.

(TAC) of the main process units involved in the integration of the production of oxygen with the gasifier.

The TAC includes all the equipment installation costs annualized using a Capital Charge Factor of 0.351. For estimating the installation cost of the principal equipment we use the correlations in Turton et at [27]. and update the cost using the Chemical Engineering Plant Cost Index (CEPCI) of 593.8 for May 2012. We considered axial compressor and axial turbines (with 0.85 isentropic efficiency [28]) for compressing and expanding the air respectively. We also consider a centrifugal compressor (with 0.72 isentropic efficiency) driven by an electric motor to compress the methane. For heat exchangers and the reboiler we considered shell and tube heat exchanges with fixed tubes. For the cost of the ITM membrane modules we referred to Bose [13] who reported a cost of € 3000 per m^2, from which € 1500 correspond to ITM membrane tubes and € 1500 to the membrane module shells. With a conversion factor of 1.05 U.S $ per € it represents a cost of U.S.$ 3150 per m^2 of membrane area. For these membranes, we considered a life-time of five years, and the Capital Charge Factor of 0.351 of their installed cost covers both amortization and its annual maintenance.

The TAC includes purchases of methane, electricity and income from selling the steam produced. To compute the energy consumed by the compressor, we considered an electrical to mechanical conversion efficiency of 0.9 and an electric energy cost of US$0.07 per kW-hr. For methane, we take the price of 0.2327 U.S $/kg [29].

In Fig. 18 we plot the TAC of the process for the conventional configuration when air flow rates fed are 2100, 2300, 2900, 3200 and 3500 kmol/h, in the range of permeate pressures analyzed. The pressure on the permeate side is varied from a minimum of 0.25 bar to a maximum where the partial pressure difference for oxygen is near to zero. The minimum pressure of 0.25 bar is for the hypothetical case in which the permeate compressor can perform vacuum, which implies to use a vacuum pump, which would cause a jump in the total annualized cost (to operate below the atmospheric pressure 1.013 bar) and require a different flow sheet. Thus, the curves are strictly valid for permeate pressures above atmospheric. Thereby, the curve for 2100 kmol/h of air flow rate is dismissed because its maximum pressure in the permeate side is only 0.9 bar. For the configuration of conventional separation a minimum TAC of 13,535,121.00 is reached when the inlet air flow rate is of 2300 kmol/h and the pressure of the permeate side is 1 bar (cuasi atmospheric pressure).

In Fig. 19 we plot the TAC of the Process for the configuration in countercurrent arrangement, in the range of permeate pressures analyzed. For this alternative, it is observed that the air flow rates have a higher economic impact than in the conventional configuration. A minimal TAC of 11,732,858.00 is located at the minimal air flow rate (2100 kmol/h).

The alternative with the countercurrent arrangement with sweep stream has a TAC 13.5% lower than the alternative with the conventional separation. This reduction is mainly due to not needing the compressor to raise the pressure of permeated oxygen to the working pressure, avoiding the cost associated with it. The TAC does not include carbon taxes. However, both alternatives have a similar discharge of carbon

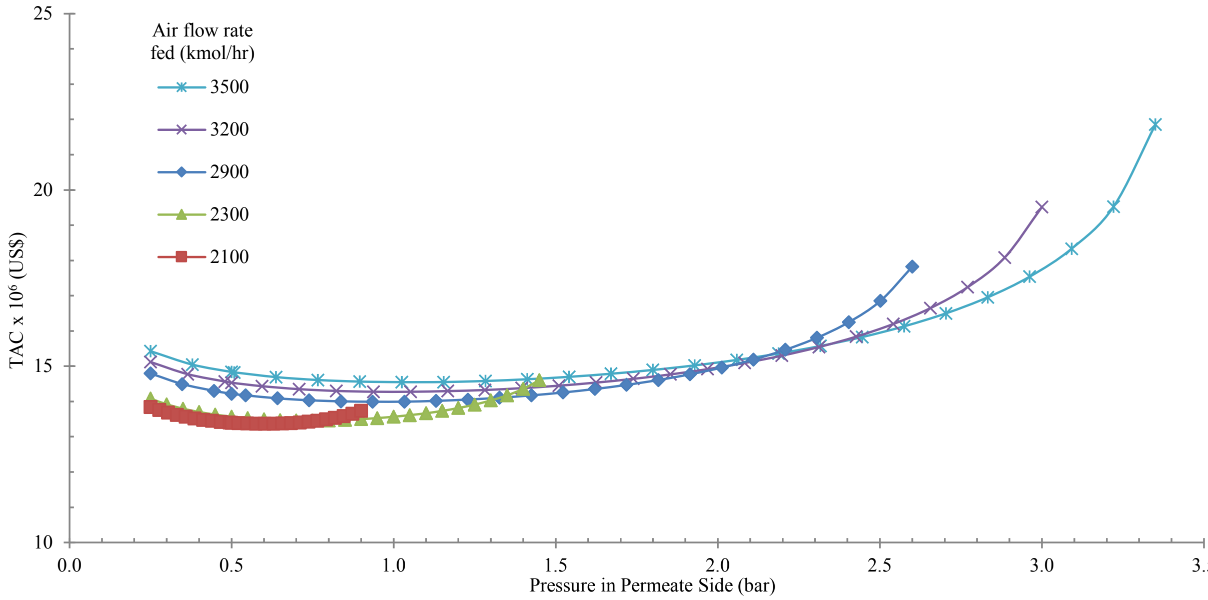

Fig. 18 – The Total Annual Cost of the Process for the conventional configuration when air flow rates fed are 2100, 2300, 2900, 3200 and 3500 kmol/h, varying the pressure in the permeate side.

dioxide, so even if they were considered this would not significantly alter the results.

Conclusions

In this work, we analyzed different design alternatives to achieve the integration of the production of synthesis gas by gasification of coal with the production of oxygen through ITM membranes. We compared the use of ITM membranes in the traditional separation configuration, with using them in a countercurrent arrangement with sweep stream (as a mass exchanger). We developed custom models in Aspen Custom Modeler V8.4 for the permeate modules in both configurations and compare their performance in different flow sheet alternatives using Aspen Plus V 8.7. At present, the ITM membranes are the focus of intense research, but its use in a countercurrent arrangement configuration as proposed in this paper has not been studied previously. The here proposed configuration reduced the Total Annualized Cost in about

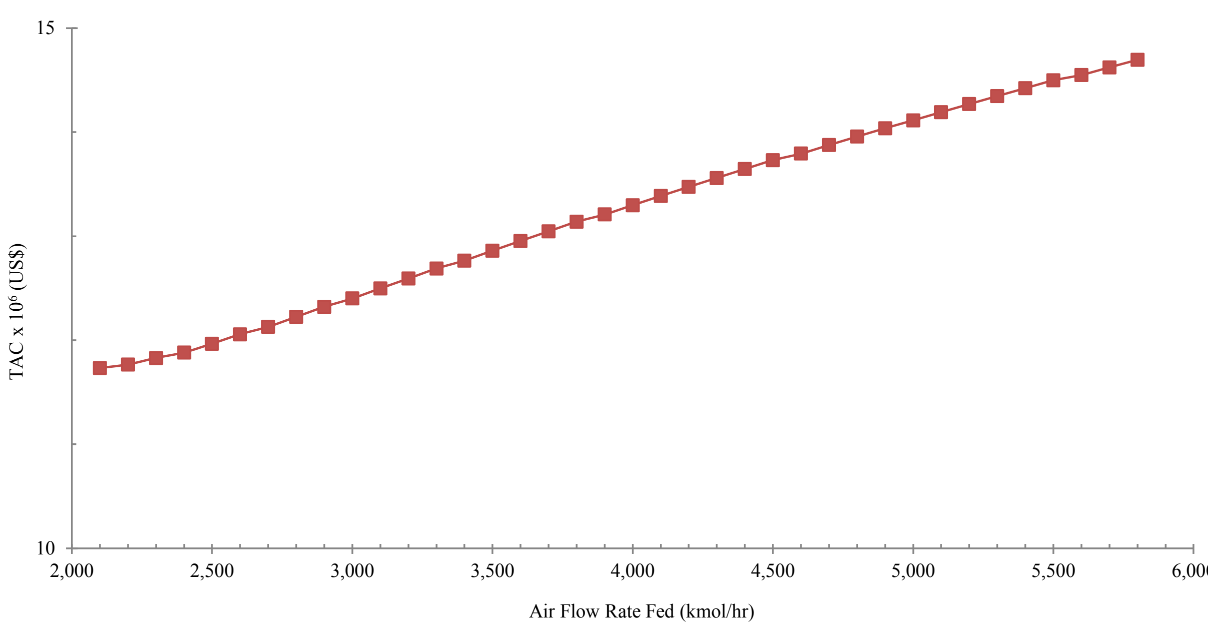

Fig. 19 – The Total Annual Cost of the Process for the configuration in counter current arrangement.

13.5% compared to the conventional configuration. This reduction in cost is an important incentive for further study on the use of ITM membranes in countercurrent with sweep streams (as a mass exchanger). In turn, it is expected that in the near future ITM membranes (if their industrialization occurs) will improve their permeability and will substantially reduce their cost.

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