

## 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.

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#### Introduction

In previous papers we worked on hydrogen exchange between different streams of the same process  $[1-4]$  $[1-4]$  $[1-4]$  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 [\[5\]:](#page--1-0) 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 [\[6\]](#page--1-0). 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 [\[7,8\]](#page--1-0), 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% [\[8\],](#page--1-0) 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 [\[9\]\)](#page--1-0).

#### Overview of oxygen separation

The most commercially suitable alternative for obtaining oxygen, has long been the cryogenic distillation of air [\[10\]](#page--1-0). 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% [\[11\]](#page--1-0). 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 [\[8\].](#page--1-0) 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 [\[9\]](#page--1-0). 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 [\[12\]](#page--1-0).

Presently, ITM (Ion Transport Membrane), or also named MCM (Mixed Conducting Membrane) have been developed, that efficiently perform the oxygen separation from air [\[13,14\]](#page--1-0). 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) [\[15\]](#page--1-0). 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 [\[16\]](#page--1-0).

The ITM membranes were proposed by Researchers of Air Products and Chemicals Inc. [\[10,15,17\]](#page--1-0), 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. [\[18,19\]](#page--1-0), Möller et al. [\[16\],](#page--1-0) Foy and Yantovski [\[20\],](#page--1-0) developed zero emission cycles based on these membranes. Yantovsky et al. [\[18,19\]](#page--1-0) 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

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