



Hybrid membrane/cryogenic separation of oxygen from air for use in the oxy-fuel process

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ABSTRACT

The process of oxy-fuel combustion requires the separation of oxygen from air on a large scale for use in the combustion chamber. This separation is currently done through energy intensive cryogenic distillation. To reduce the overall energy requirements for air separation it is examined whether a hybrid membrane and cryogenic process be utilized instead. The examined process uses an O₂/N₂ permeable membrane to create oxygen enriched air. This enriched air is then turned into high purity oxygen using cryogenic distillation. Several arrangements of such a system are investigated and compared on a practical and thermodynamic level to the current cryogenic process in use. It is found that using a vacuum pump arrangement to draw air through the membrane has potential to reduce energy requirements from the current standard. It is also found that the hybrid system is more productive in small to medium scale applications than in large scale applications because of the increased irreversibilities in the cryogenic process at smaller scales.

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1. Introduction

Oxygen production from air separation is used in many applications worldwide such as medical devices, steel and chemical manufacturing, and most recently carbon capture on a large scale. Historically, the size of the operation and the required purity of oxygen determine the method of separation [1].

Presently, a widely discussed process for carbon capture is the oxy-fuel process, in which the oxidizer for the power plant is pure oxygen mixed with power plant exhaust [2]. Thus, the final exhaust is essentially only carbon dioxide and water, which can be easily separated; the carbon dioxide is then stored in underground caverns, e.g., depleted oil or gas fields. The process of oxy-fuel combustion requires high purity oxygen in large quantities. For oxy-fuel processes such as the combined cycle, Graz cycle [3] and Water cycle [4] the oxygen separation process, however, would be an addition to an existing power plant and the work required to separate oxygen from air becomes an energy penalty for the power plant to operate with sequestration [5]. Oxy-fuel cycles which require no additional energy for air separation such as Chemical Looping Combustion [6] are also proposed but still require significant development. Kvamsdal et al. [7] show that a gas turbine combined cycle power plant installed with the oxy-fuel process will reduce the overall plant

efficiency from 56.7% to 47%. Approximately 8.8% of the efficiency loss comes from the work required to produce and compress oxygen for combustion through cryogenic distillation in the Linde process. Thus, if the separation process can be improved by even a small margin, the oxy-fuel process will benefit substantially.

The three branches of technologies that currently exist to separate oxygen from the air include distillation, adsorption and membranes [8]. Distillation is the most mature of the three technologies and allows for both high purities (>99%) and large scale productions [9]. Adsorption is able to reach purities of up to 95% oxygen but the requirement of solvents limits its size capacity due primarily to capital costs [1]. Membrane technology is the most recent of the three and includes polymeric and high temperature ion transport membranes. While polymeric membranes can produce oxygen enriched air of various concentrations, ion transport membranes can produce purities of close to 100%. Both membranes however have yet to be built for large scale gas separations [1]. A combination of the above technologies for the production of high purity oxygen has been suggested [10] but no detailed research has been completed discussing either energy costs or integration to the oxy-fuel cycle. Several papers though have discussed the possibility of combining these technologies for various other purposes such as the production of argon [11] and the recovery of CO₂ [12].

The hypothesized design to create pure oxygen using less energy utilizes both membrane and cryogenic technologies. With an induced pressure difference, an O₂/N₂ selective polymeric membrane can partly separate oxygen from air by having properties that allow

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oxygen molecules to pass through the membrane easier than nitrogen molecules. However, the membrane cannot fully separate oxygen from the air; it can only be used to increase the concentration of oxygen as some nitrogen molecules will also flow through the membrane. The enriched air can then be processed in a cryogenic stage; this will require less work than if ambient air is used, since a smaller volume of gas needs to be processed in the cryogenic process to obtain the same amount of product, i.e. pure oxygen. Enrichment of the air using a membrane, however, introduces extra work irreversibilities in the form of additional compressors, turbines and blowers, and irreversibilities for the membrane flow. Depending upon the actual value of the various irreversibilities, using a combined membrane and cryogenic process may provide a smaller energy penalty than the cryogenic process alone; in the following, several arrangements are considered to test this hypothesis.

A compressor on the feed side of the membrane coupled with a recovery turbine for the waste gas can be used to create the required membrane pressure difference. Alternatively, a vacuum pump on the permeate side of the membrane can be used to create the pressure difference. Both configurations are thermodynamically modeled to determine the best method of air separation. The primary parameter in the system is the membrane selectivity which is a property of the membrane. The selectivity is varied to study the effect it has on the separation work. The effects of adding heat to the turbine inlet and varying the efficiencies of the compressor and turbine are examined as well. For large oxygen production rates (thousands of tons O₂/day) only the vacuum pump arrangements show improvement over the existing (i.e., cryogenic alone) design, while both variants of the hybrid design show improvements in small to medium scale productions (hundreds of tons O₂/day).

The remainder of this paper is organized as follows: Section 2 discusses the basic processes in membranes and cryogenic processes for air separation. In particular a detailed discussion of membrane separation is presented. In Section 3, various configurations for hybrid systems are introduced, including a discussion of the methodology behind the processes. The numerical results for the separation work of the various arrangements are graphed and discussed in Section 4 for both large and small scale productions rates, including the resulting effect on the oxy-fuel process. In the last Section, the effect of varying certain parameters is investigated and recommendations regarding the future of the hybrid system are discussed. Integration of the hybrid system to the oxy-fuel process and comparisons to ceramic membrane separation are also included in the discussion.

2. Current technologies for air separation

2.1. Membranes

The use of selective membranes for partial air separation progressed rapidly in the 1980s as a promising alternative to cryogenics and adsorption [13]. These membranes allow the passage of certain molecules easier than other molecules. For an O₂/N₂ permeable membrane, both oxygen and nitrogen can permeate through the membrane, but oxygen does so with more ease. Fig. 1

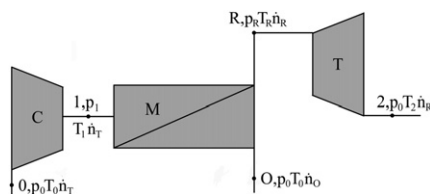


Fig. 1. Simple Membrane Process with a Compressor coupled with a Recovery Turbine.

shows the set-up of the basic process. To induce a flow of molecules across the membrane, a pressure difference must exist, which is created by a compressor. A recovery turbine is then placed downstream, so that the portion of the gas that did not pass through the membrane can be expanded and some of the compression work can be recovered. The oxygen enriched gas that did pass through the membrane is assumed to be at atmospheric pressure.

An alternative set-up to create the pressure difference is a vacuum pump on the permeate side as seen in Fig. 2. This configuration requires a blower to keep fresh, undepleted air flowing past the membrane. Otherwise oxygen depleted air would pool along the membrane reducing the amount of oxygen that passes through.

In the following, the subscript *R* indicates the nitrogen rich waste gas while the subscript *O* indicates the oxygen rich product gas. All gases are assumed to be ideal gases with constant specific heats.

Membrane permeability is different for each type of molecule and determines its ability to flow through the membrane. For a component γ , the mole flow through the membrane is given by

$$\frac{dn_\gamma}{dt} = Q_\gamma (p_{\gamma,0} - p_{\gamma,1}) \quad (1)$$

where Q_γ is the permeability of the membrane for a given component, and $p_{\gamma,0}$ and $p_{\gamma,1}$ are the partial pressures of the component before and after the membrane, respectively.

Technical membrane separation processes are continuous processes, where the source gas flows along the membrane while some of it penetrates to the other side. For the computation it is convenient to follow an initial amount n_T of source gas as it travels along the membrane and its components traverse the membrane. Then Q_γ is a parameter that depends on the initial amount considered and on the size of the membrane.

In an O₂/N₂ permeable membrane the ratio of the membrane permeabilities of oxygen and nitrogen, Q_{O_2} and Q_{N_2} , is the membrane selectivity, α , which is independent of the size of the system considered,

$$\alpha = \frac{Q_{O_2}}{Q_{N_2}} \quad (2)$$

The selectivity is dependent upon the type of membrane, the membrane material and the membrane configuration.

In order to gain independence of the system size, it is convenient to introduce a dimensionless permeation time scale as

$$\tau = tp_0 Q_{N_2} / n_T \quad (3)$$

The dimensionless permeation time τ measures the time that the gas is allowed to permeate through the membrane; it can be controlled by choosing the flow rate of the gas through the system or by altering the length of the membrane. However, sufficient air flow must be required to ensure that the gas in front of the membrane is well mixed. The flow rates of oxygen and nitrogen across the membrane are determined by the difference in the partial pressures of each gas on the feed and permeate sides. Since the concentration of each gas changes over time, the mole flows are time dependent. Using Eqs. (1–3) and the ideal gas relation $p_\gamma/p = X_\gamma$, the mole flows of oxygen and nitrogen obey the equations

$$\frac{dn_{1f}}{d\tau} = \alpha \left(X_{1p} - X_{1f} \frac{p_1}{p_0} \right) = -\frac{dn_{1p}}{d\tau} \quad (4)$$

$$\frac{dn_{2f}}{d\tau} = \left((1 - X_{1p}) - (1 - X_{1f}) \frac{p_1}{p_0} \right) = -\frac{dn_{2p}}{d\tau} \quad (5)$$

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