

# ENERGETIC ANALYSIS OF OXYGEN-ENHANCED COMBUSTION WITH AIR SEPARATION BY POLYMERIC MEMBRANES

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## **Abstract.**

*This paper presents a study of an oxygen enhanced combustion process where the oxidant stream is obtained after air separation by a membrane set. A theoretical analysis is performed over a generic combustion chamber fed by an oxidant stream, initially at atmospheric conditions. The oxygen concentration is ranged from 21% up to 30% in volume, known as oxygen enhanced combustion, keeping the equivalence ratio constant. The oxygen enhanced stream is obtained out a membrane set, capable to deliver O<sub>2</sub>/N<sub>2</sub> with different concentrations and flow rates according to the operational feeding pressure. Polymeric membranes with three permselectivities are accessed for several operational pressure values. The energy balance of the coupled system displays an increase about 6,0% in available energy when compared to the standard case (21% O<sub>2</sub> in the oxidizer by volume). It is also observed a reduction up to 45,0% in the specific emission of CO in some of the conditions studied.*

**Keywords:** oxygen-enhanced combustion, polymeric membranes, gas separation

## 1. INTRODUCTION

Combustion systems usually use air as the oxidant stream, which is composed of approximately 21% O<sub>2</sub> and 79% N<sub>2</sub> on volume. This amount of nitrogen carries out much of the thermal energy obtained from the combustion process, decreasing its efficiency and producing large amounts of exhaust gases.

The oxygen-enhanced combustion (OEC) is a technology that can be applied to any combustion process. It comprises the use of an oxidizer with higher oxygen concentration if compared to the atmospheric air. Thus, for a given oxidant / fuel ratio, the amount of O<sub>2</sub> in the oxidizer can be kept constant while the amount of N<sub>2</sub> in the air is reduced by different techniques. The benefits of EOC on combustion processes include the increase on the thermal efficiency, the reduction on the amount of generated flue gas and consequent impact on industrial process performance (Baukal Jr., 1998).

Low enrichment levels of combustion (O<sub>2</sub> < 30%) are attractive due to the low cost of retrofitting. In addition, methods of oxygen production with high efficiency can be used in these cases, including the N<sub>2</sub> adsorption processes (PSA and TSA) and separation by membrane.

The search for more efficient energy processes and the advances on oxygen production have motivated the study of the application of OEC technology. Qiu et al., 2008, analyzed experimentally the application of OEC in burners of the porous fiber type and porous reticulated ceramic. Results showed a 5% increase in efficiency of the reticulated ceramic burner and 6% in the porous fiber burner. The authors concluded that the gain in heating efficiency was due to the increase in flame temperature.

In a detailed study, Baukal Jr., 1997, examined the thermal power emitted by a gas burner for several combustion enrichment levels, for different flame positions and for equivalence ratios  $\Phi$  between 0.25 and 1.45. The higher thermal power was obtained for equivalence ratios near stoichiometric and oxygen concentrations of approximately 100%. All experimental measurements were performed near the centerline of the burner. An important feature detected during the study was the increase in non-luminous flame obtained with the combustion enrichment.

With this background, this paper investigates the application of OEC technology to a generic combustion chamber for burning gas. The processes to increase the concentration of O<sub>2</sub> in the oxidizer and the combustion chamber were modeled using thermodynamic equations and diffusive mass transport. This article intends to be a proof of concept application of oxygen-enhanced combustion obtained with the aid of polymeric membranes.

## 2. METHODOLOGY

### 2.1 Description of the case analyzed

The EOC system is depicted in Fig. 1, where a generic combustion chamber is coupled to an air separation set at stream 5. The air separation set is composed by an atmospheric air compressor, a heat exchanger and a membrane.

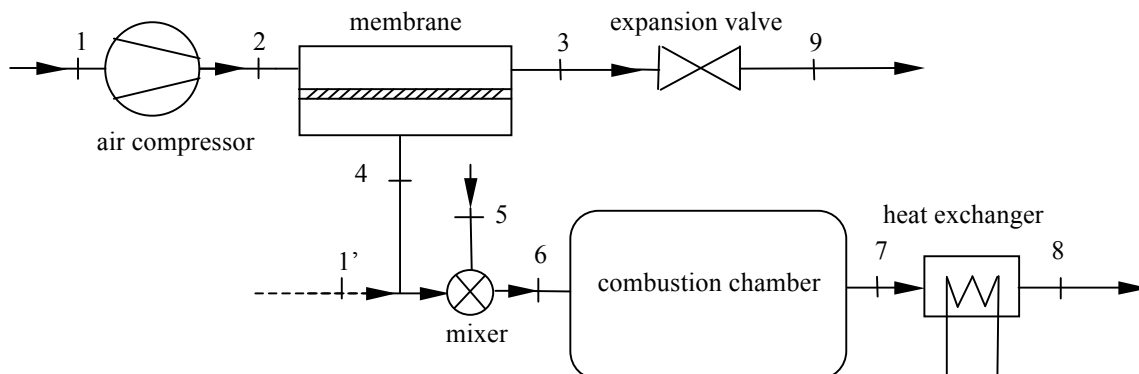


Figure 1. Schematic representation of the combustion system with a membrane air separation set for oxygen enhancement

Atmospheric air is admitted from the environment at point 1 with 21% O<sub>2</sub> and 79% N<sub>2</sub>, and separated into two different streams. The enhanced oxidizer stream at point 5, with O<sub>2</sub> content varying up to 30%, also called the permeate stream, and a retentate stream (point 4), vented back to the environment by expansion valve. The oxidizer reacts with the injected fuel (point 5) at the combustion chamber. The generated flue gases are discharged out of the chamber at point 8, after going through the heat exchanger.

To compare the conventional combustion process with the OEC, is considered that for the normal combustion (21% de O<sub>2</sub> in point 5), atmospheric air is injected through the point 1' of Fig. 1. In this case, the rate flow in point 1 is assumed to be zero.

In the Fig. 1, energy is exchanged with the environment by compressor and the heat exchanger. The other processes are considered energetically insulated.

Three types of polymeric membranes for air separation were chosen, and their characteristics are shown in Table 1:

Table 1. Main physico-chemical properties of polymeric membranes selected for the air separation study.

Polymeric Membrane (Material)	O <sub>2</sub> Permeability (kmol μm m <sup>-2</sup> kPa <sup>-1</sup> s <sup>-1</sup> )	Seletivity (O <sub>2</sub> / N <sub>2</sub> )
Etyl cellulose	3,696x10 <sup>-9</sup>	3,4
Poly (4-methyl-1-pentene) (TPX)	9,900x10 <sup>-9</sup>	4,2
Polyimide	2,607x10 <sup>-9</sup>	6,2

All processes are modeled as ideal, with pressure head loss occurring only at the membrane device. It was also considered that the physico-chemical properties of membranes and the thermophysical properties of the stream gases of system are constant under the conditions studied.

## 2.2 Mathematical modeling

Mass, energy and chemical species balances were established for each one of the devices of the complete system. All processes were simulated for steady state regime, and fluids followed ideal gas behavior, as it is a common assumption for combustion process modeling.

Atmospheric air was compressed from point 1 to point 2, and its flow rate was a function of the imposed O<sub>2</sub> concentration of the oxidizer stream (stream 5), to be injected into the combustion chamber. Regular devices as compressors and heat exchangers operate below 100% efficiency, for different reasons. The former one will never reach that condition due to the friction of the working fluid, meanwhile the last one would need an infinite surface area to perform total heat exchange. Nevertheless, the compressor efficiency  $\eta_c$  and the heat exchanger effectiveness  $\varepsilon$  were both taken as 100% to bring results independent of any technologic choice.

In polymeric membranes, the separation process is modeled by Fick's Law (Baker, 2004). The molar flow rate  $\dot{n}_i$  of a given specie  $i$  was calculated by Eq.(1):

$$\dot{n}_i = P_i(A/L)(p_{i,H} - p_{i,L}) \quad (1)$$

where  $P_i$  is the permeability of specie  $i$ ,  $p_{i,H}$  and  $p_{i,L}$ , are the partial pressures of  $i$  at low-pressure side and high-pressure side of membrane, respectively. For the area  $A$  and thickness  $L$  were initially admitted the values of 10,000 m<sup>2</sup> and 1.0 μm, respectively. To facilitate convergence of the calculations, the partial pressure at the high level flow side was

admitted as being equal to the pressure of O<sub>2</sub> at retentate side (Lopez, 2010). The low-pressure side (point 4) was set at 101.325 kPa.

Fuel at point 6 was taken as pure CH<sub>4</sub>, which reacts with the oxidizer (stream 5) in stoichiometric proportion (equivalence ratio of  $\Phi = 1$ ). The overall combustion reaction is given by Eq. (2):



where the variables  $a$  and  $b$  depend on the combustion enrichment level. Traces are formed by the gases such as CO, NO and O<sub>2</sub> in chemical equilibrium with the other exhaust gases. The exhaust gases are cooled in heat exchanger of the adiabatic flame temperature (point 7) up to environment temperature (point 8), which value was set at 25°C.

The energy balance, applied to Fig. 1, is calculated by Eq. (3):

$$\dot{E}_s = \dot{W}_c + \sum_e \dot{n}_e \bar{h}_e - \sum_s \dot{n}_s \bar{h}_s \quad (3)$$

where  $\dot{E}_s$  is the net rate of energy obtained for each mole fraction of O<sub>2</sub> allowed for the stream 5. The quantities  $\dot{W}_c$  the power demanded by the compressor. The sub-indexes  $e$  and  $s$  refer, respectively, the gas streams entering and leaving the processes shown in Fig. 1.

The balance mass applied to each of the species  $i$  in the considered problem is calculated by Eq. (4):

$$\sum_e \dot{m}_{e,i} - \sum_s \dot{m}_{s,i} = 0 \quad (4)$$

where  $\dot{m}$  is the mass flow rate of each species. The sub-indices  $e$  e  $s$  follow the same notation adopted in Eq (3).

For all the cases analyzed, the net energy gain (or lost)  $EG$  with a given enrichment level of combustion is compared with the conventional combustion by the following ratio given by Eq. (5):

$$EG = \left( \left| \dot{E}_{s,OEC} \right| - \left| \dot{E}_{s,CC} \right| \right) / \left| \dot{E}_{s,CC} \right| \quad (5)$$

being  $\dot{E}_{s,OEC}$  and  $\dot{E}_{s,CC}$  are the net energy obtained for a given enrichment level of combustion and for conventional combustion, respectively, in the predefined conditions of equivalence ratio and area.

The analysis of the influence of physical quantities with values initially pre-defined (separation area and equivalence ratio), is given by the variable energy gain (or lost) ratio  $VEG$ , defined by Eq. (6) below:

$$VEG = \left[ \left| \dot{E}_{s,0.30}(A, \Phi) \right| - \left| \dot{E}_{s,CC}(A, \Phi) \right| \right] / \left| \dot{E}_{s,CC}(A, \Phi) \right| \quad (6)$$

where  $\dot{E}_{s,0.30}(A, \Phi)$  is the net rate energy obtained for molar fraction of O<sub>2</sub> in point 4 of 0.30 and  $\dot{E}_{s,CC}(A, \Phi)$  for conventional combustion, both as a function of area,  $A$ , and equivalence ratio,  $\Phi$ .

The relative specific emission of main flue gases generated by the combustion chamber (CO, CO<sub>2</sub> and N<sub>2</sub>), considering the pre-defined values of  $A$  and  $\Phi$ , is given by the ratio given by Eq. (7):

$$RSE_i = \left( se_{i,8,OEC} - se_{i,8,CC} \right) / se_{i,8,CC} \quad (7)$$

and for variable values of  $A$  and  $\Phi$ .

$$VRSE_i = \left[ se_{i,8,0.30}(A, \Phi) - se_{i,8,CC}(A, \Phi) \right] / se_{i,8,CC}(A, \Phi) \quad (8)$$

where  $RSE_i$  is the relative specific emission associated to flue gas  $i$  calculated in the stream 8. The quantity  $VRSE_i$  represents the variation of  $RSE_i$  with area and equivalence ratio, for a given enrichment level of combustion. The variable  $se$  is the specific emission of gas  $i$ , which is defined by Eq. (9):

$$se_i = \dot{m}_{8,i} / \left| \dot{E}_s \right| \quad (9)$$

where  $\dot{m}_{8,i}$  is the mass flow rate of flue gas  $i$  in the stream 8.

### 3. RESULTS

The complete set of equations was simulated by the aid of the Engineering Equation Solver software (EES), and the variable notation refers to the one presented at Fig. 1. Results were obtained for constant values of separation area and equivalence ratio. It was also analyzed the influence of variation of these values in the net rate of energy obtained with the combustion enrichment.

#### 3.1 Net energy

The Fig. 2 shows the behavior of the energy gain (in absolute percentage), given by Eq. (5), as a function enrichment level, for constant molar flow rate of fuel:

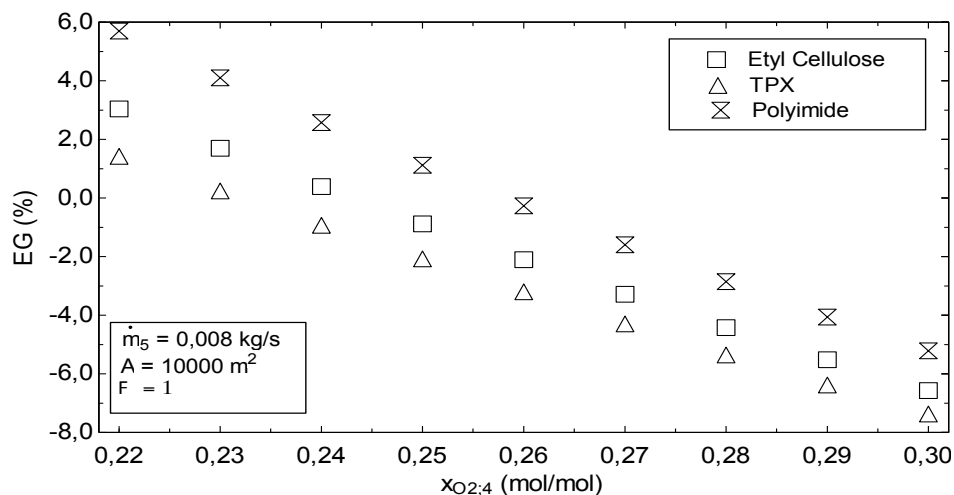


Figure 2. Percentual gain of energy by the system,  $EG$ , as a function of  $O_2$  level in the stream 5 to the three types of membranes analyzed.

The behavior displayed at Fig. 2 was obtained by applying Eq. (5) to various levels of combustion enrichment and for the membranes selected. The results show an increase (for values of  $x_{O_2,4}$  up to 0,26) of net rate of energy released by the system of Fig. 1 for constant values of fuel flow rate, separation area and equivalence ratio. The greatest gains in energy with the OEC process were obtained with the membrane of polyimide, mainly due to higher enthalpy obtained for the point 2 of the system. For the three membranes, the value of  $EG$  decreases with the enrichment level of combustion due to increase of the power consumption by the compressor.

The values obtained for  $VEG$  as a function of quantity  $A$  are shown in Fig. 3, respectively:

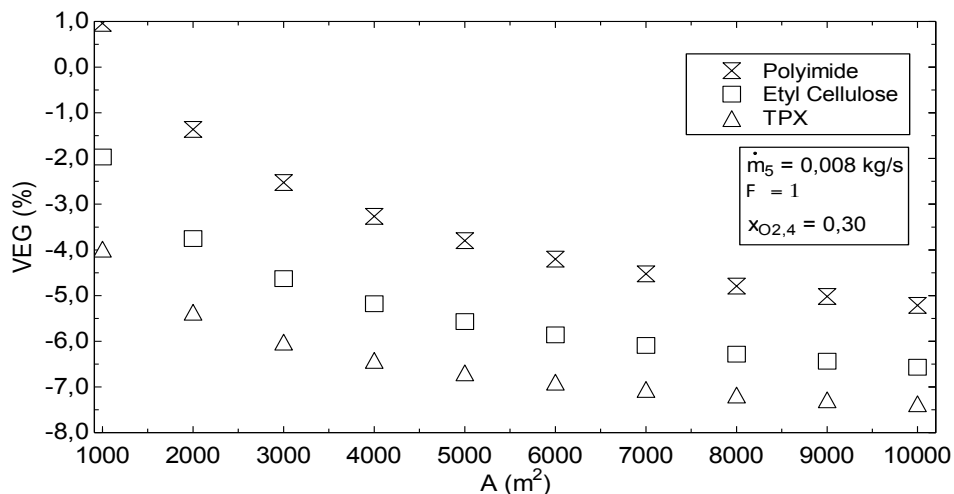


Figure 3. Percentual variation of net energy of the system,  $VEG$ , as a function of separation area of membrane. Results obtained for 30% of  $O_2$  level in the stream 4.

The results of Fig. 3 show that, compared with those shown in Fig 2, there is an increase in energy gained with the OEC process, as the quantity  $A$  increases. This behavior is due mainly to reduce the air flow rate necessary to increase the area of separation membranes, thereby reducing the power demanded by the compressor.

The Fig. 4 shows the value of  $VEG$  as a function of equivalence ratio,  $\Phi$ , for the three selected membranes:

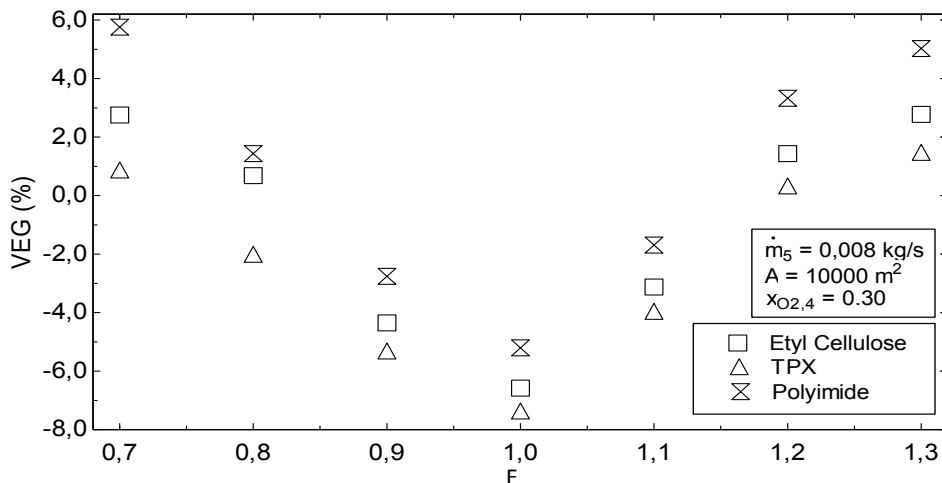


Figure 4. Percentual variation of net energy of the system,  $VEG$ , as a function of equivalence ratio  $\Phi$ . Results obtained for 30% of  $O_2$  level in the stream 4.

The results of Fig. 4 show that there is a reduction of net energy gain with the OEC process for equivalence ratios near 1.0 for all analyzed membranes. This behavior is mainly due to reduction of the specific enthalpy,  $h_4$ , and as a consequence, of the temperature  $T_8$  as the combustion become stoichiometric.

### 3.2 Emissions

From Fig. 5 to Fig. 7 shows the behavior of emission reduction (in percentage), given by Eq. (7), as a function enrichment level, for constant molar flow rate of fuel and for the pollutants  $CO_2$ ,  $CO$  and  $NO$ :

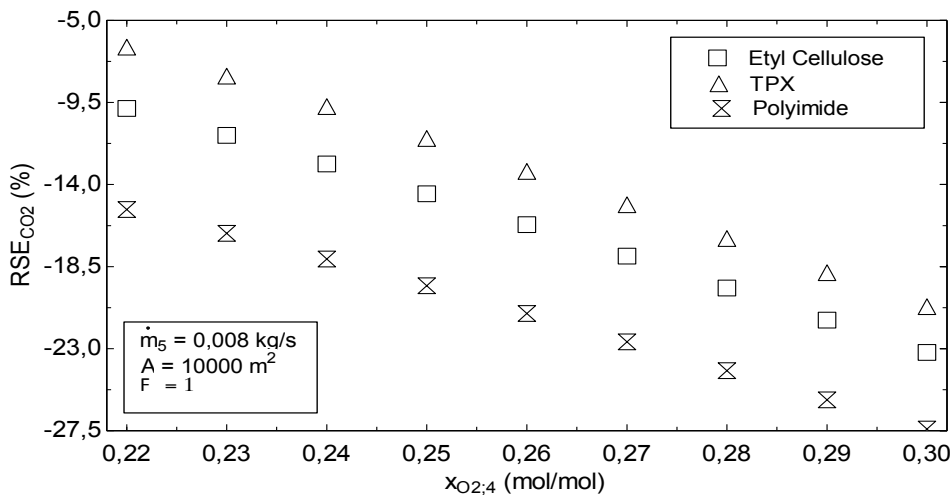


Figure 5. Relative specific emission of the flue gas  $CO_2$ ,  $SE_{CO_2}$ , as a function of  $O_2$  level in the stream 4 to the three types of membranes analyzed.

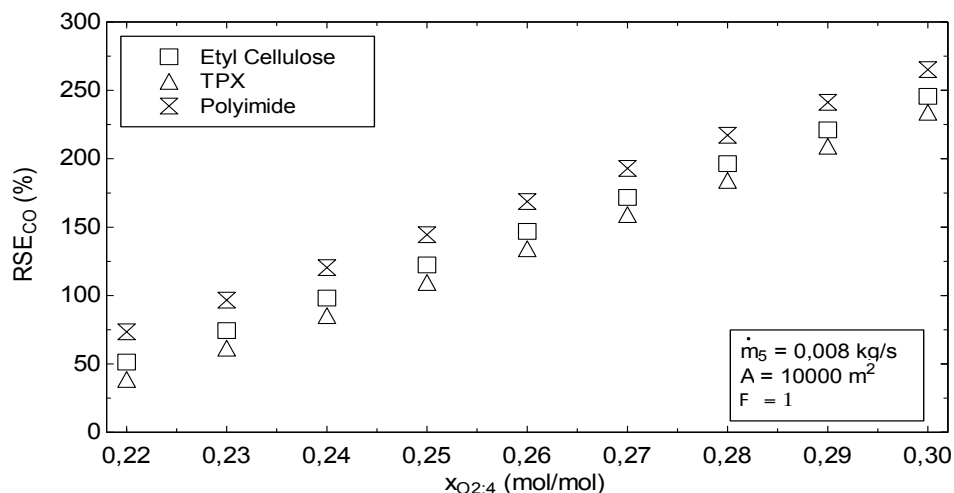


Figure 6. Relative specific emission of the flue gas CO,  $RE_{CO}$ , as a function of O<sub>2</sub> level in the stream 4 to the three types of membranes analyzed.

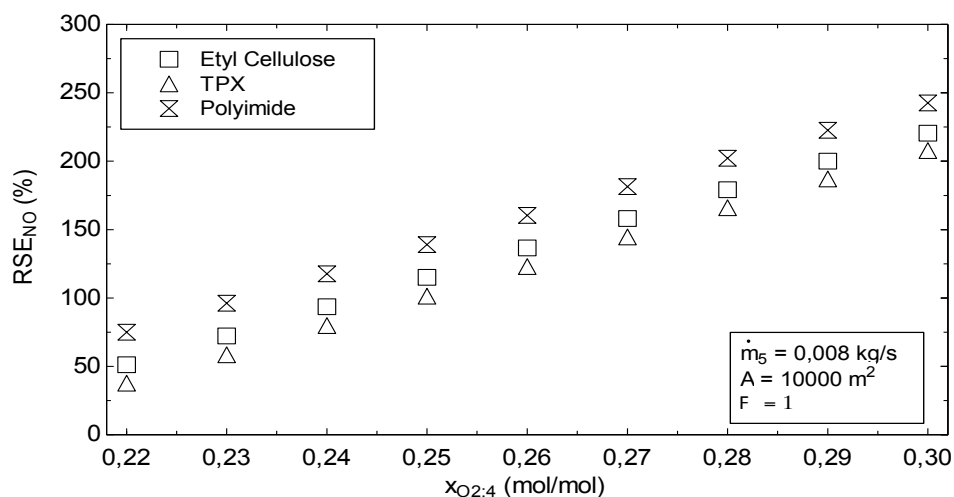


Figure 7. Relative specific emission of the flue gas NO,  $RE_{NO}$ , as a function of O<sub>2</sub> level in the stream 4 to the three types of membranes analyzed.

As in Fig. 2, the results shown from Fig. 5 to Fig. 7 were obtained for constant values of fuel flow rate, separation area and equivalence ratio. In all cases occurs a reduction with the application of the OEC process of the specific emission of all considered pollutants. The values of  $RSE$  the behavior presented mainly due to the dissociation reactions of the combustion gases to increase the combustion enrichment. The system with the TPX membrane showed the highest percentage value of  $RSE_{CO}$  and  $RSE_{NO}$ , due to lower temperature of flue gas (point 7).

Similarly the Fig. 3, from Fig. 8 to Fig. 10 are shows the behavior of variation of emission reduction,  $VRE_i$  as a function membrane separation area. The results are obtained for conditions for constant molar flow rate of fuel and level of enrichment ( $x_{O_2,4}$  of 0.30):

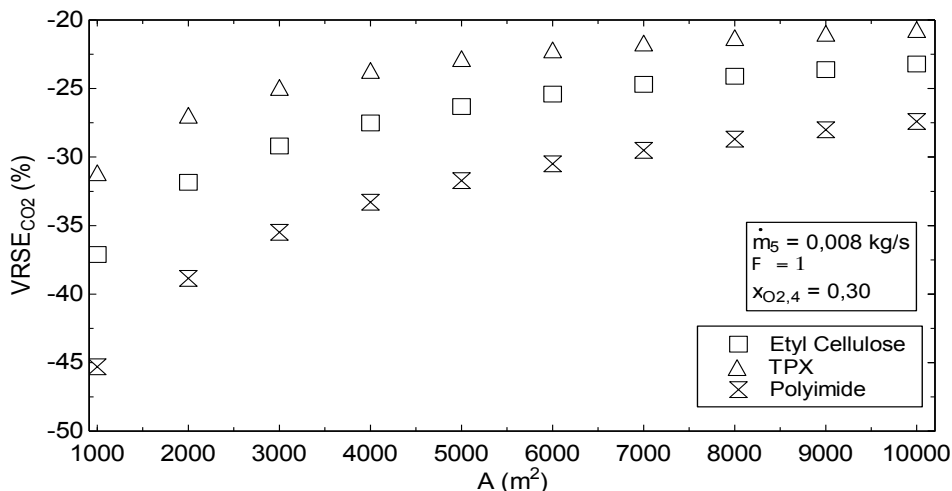


Figure 8. Percentual variation of relative emission of the flue gas CO<sub>2</sub>,  $VRSE_{CO_2}$ , as a function of separation area of membrane. Results obtained for 30% of O<sub>2</sub> level in the stream 4.

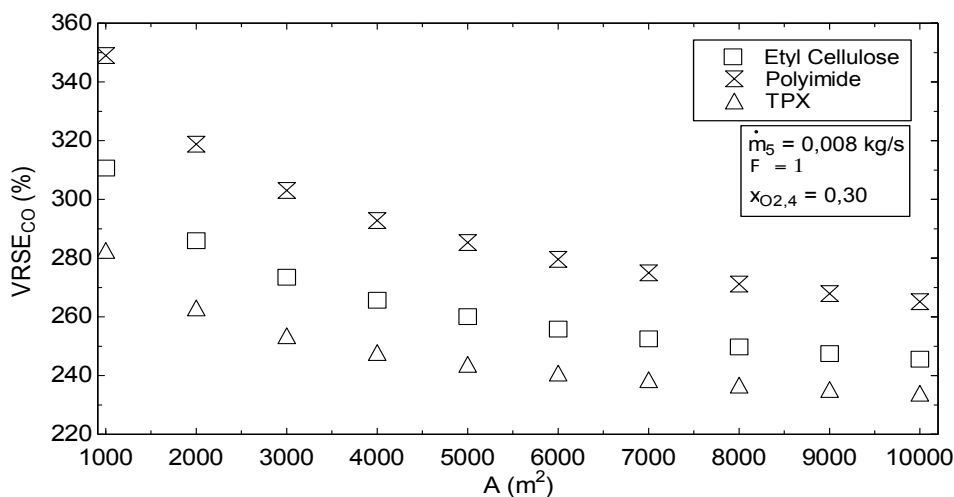


Figure 9. Percentual variation of relative emission of the flue gas CO,  $VRSE_{CO}$ , as a function of separation area of membrane. Results obtained for 30% of O<sub>2</sub> level in the stream 4.

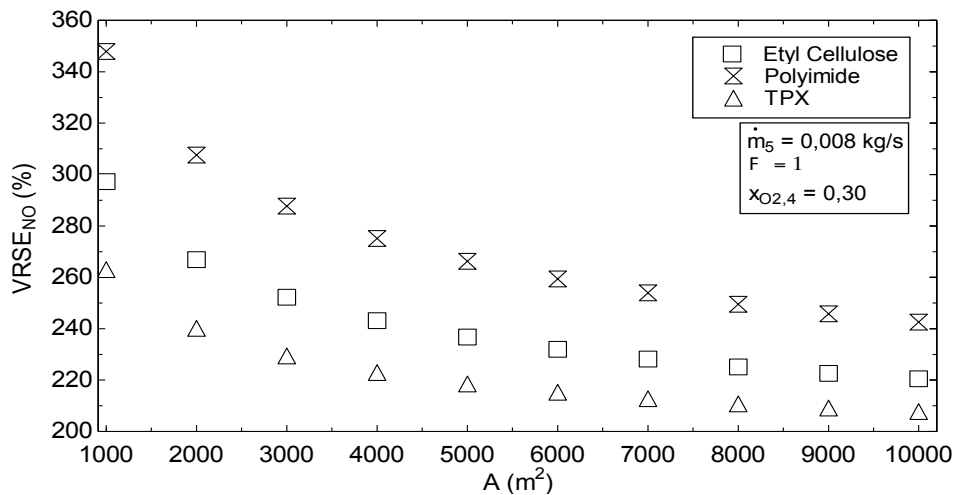


Figure 10. Percentual variation of relative emission of the flue gas NO,  $VRSE_{NO}$ , as a function of separation area of membrane. Results obtained for 30% of O<sub>2</sub> level in the stream 4.

As shown, there is a reduction in specific emissions of CO and NO with the increase of the separation area of membrane, and consequently, an increase in CO<sub>2</sub>. This is mainly due to increase of  $T_8$  with decrease of the area  $A$ .

In the Fig. 11 is shown the behavior of  $RSE_{CO}$  as a function of  $\Phi$ , for the constant area  $A$ , given by Eq. (8):

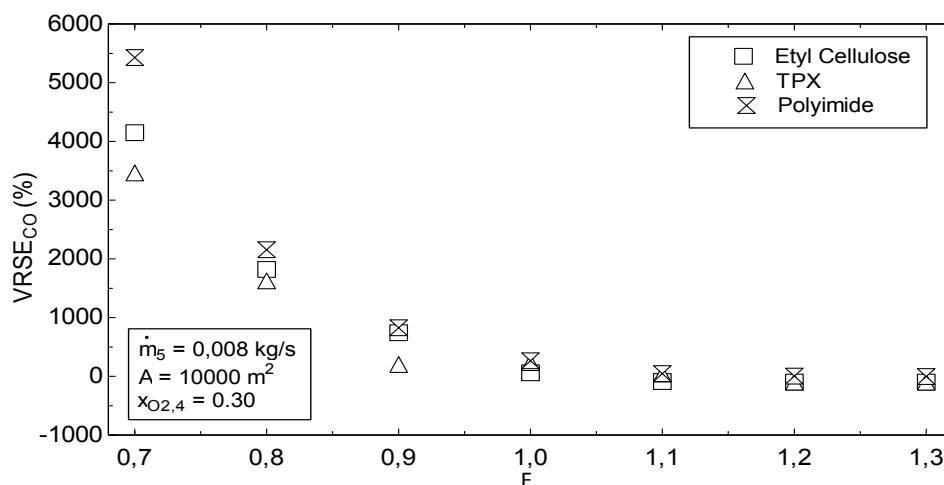


Figure 11. Percentual variation of relative emission of the flue gas CO,  $VRSE_{CO}$ , as a function of equivalence ratio. Results obtained for 30% of O<sub>2</sub> level in the stream 4.

Besides the variation of rate flow of CO, the behavior shown in Fig. 11 also occurs due to the pattern variation of the net energy available of the system with the equivalence ratio, as shown in Fig. 4.

### 3. CONCLUSIONS

In this paper, a preliminary analysis of the feasibility of using the technology of oxygen-enhanced combustion (OEC) was investigated. For this, a system comprising a combustion chamber coupled to the processes that comprise the step of enrichment of the oxidizer was modeled thermodynamically.

The results show that for all cases tested, there was an increase in energy provided by the system with lower levels of enrichment combustion (values up to 1.0% with the membrane of polyimide).

For the conditions initially pre-fixed, CO and NO increased with increasing oxygen concentration (up to 250% compared to conventional combustion). However, there was a reduction in specific CO (up to 98%) with  $\Phi$  greater than 1.0.

The present theoretical study shows that it is possible to obtain fuel saving or increase in available energy with the use of OEC technology. Higher gains of available energy can be obtained with the OEC process using more complex systems based on polymeric membranes. However, an experimental analysis is needed to determine effectively the actual gains.

### 4. ACKNOWLEDGEMENTS

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