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Thermodynamic analysis of a hard coal oxyfuel power plant with high temperature three-end membrane for air separation

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ABSTRACT

Cryogenic air separation is a mature state-of-the-art technology to produce the high tonnage of oxygen required for oxyfuel power plants. However, this technology represents an important burden to the net plant efficiency (losses between 8% and 12%-points). High temperature ceramic membranes, associated with significantly lower efficiency losses, are foreseen as the best candidate to challenge cryogenics for high tonnage oxygen production. Although this technology is still at an embryonic state of development, the three-end membrane operation mode offers important technical advantages over the four-end mode that can be a good technological option in the near future.

This paper analyzes the influence of both, the cryogenic and three-end high temperature membrane air separation units on the net oxyfuel plant efficiency considering the same boundary conditions and different equivalent thermal integrations. Moreover, the oxygen permeation rate, heat recovery, and required membrane area are also evaluated at different membrane operating conditions. Using a state-of-the-art perovskite BSCF as membrane material, net plant efficiency losses up to 5.1%-points can be reached requiring around 400,000 m^2 of membrane area. Applying this membrane-based technology it is possible to improve the oxyfuel plant efficiency over 4%-points (compared with cryogenic technology); however, it is still necessary to develop new ceramic materials to reduce the amount of membrane area required.

1. Introduction

The reduction of anthropogenic carbon dioxide emissions resulting from the use of coal is one of the most important challenges in addressing global climate change. Carbon capture and storage (CCS) is a technology which captures carbon dioxide from power plants for further storage under the ground. Broadly, three different types of CCS technologies exist: oxyfuel combustion, pre-combustion, and post-combustion [1]. This paper is focused on the oxyfuel combustion concept, where the fuel is burned with pure oxygen instead of air and mixed with a recycled part of the flue gas to maintain the combustion temperature level. Thus, the carbon dioxide concentration in the product flue gas increases dramatically, facilitating its capture. Cryogenic air separation (C-ASU) is the only available mature state-of-the-art technology to produce thousand of tons of oxygen per day (beyond 8000 TPD) at high purity (>95%) for commercial-scale oxyfuel power plants [2] and represents an important burden to the total plant efficiency. Compared with conventional air combustion plants, total efficiency drops between 8 and 12 percentage points (%-points) can be expected [3]. Nowadays, high temperature ceramic membranes (HTM), which are associated with significantly lower efficiency losses, are foreseen as the best candidate to challenge cryogenics for high tonnage oxygen production [4]. Other technologies to separate oxygen such as: pressure and temperature swing adsorption (PSA, TSA), vacuum pressure swing adsorption (VPSA) or polymeric membranes are economically suitable only for low tonnage production (250–350 TPD) at low oxygen purities (<95%). Considering even future technological improvements (materials, and energy efficiency), they are not expected appropriate for large-scale oxyfuel applications [2,5,6].

Normally, membrane modules can be operated in four-end or three-end mode. In the four-end membrane operation mode, the flue gas (mainly CO_2) is recycled at high temperature (membrane temperature) and used as sweep gas on the permeate side, whereas in the three-end mode, there is no sweep gas and the differential oxygen partial pressure through the membrane, necessary for the oxygen transport, is given by a vacuum pump (see Fig. 1). The four-end concept cannot be deployed in the near future because it still faces several technological challenges such as the need to develop: a high temperature flue gas cleaning unit (700–1000 °C) to remove dust and impurities incompatible with the membrane material, a high temperature recirculation fan (500–700 °C), and a stable ceramic membrane capable of keeping its oxygen transport properties in the presence of flue gas contaminants (e.g. CO_2

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Nomenclature

Abbreviations

ASU	air separation unit
BSCF	ceramic perovskite material $Ba_{0.5}Sr_{0.5}CO_{0.8}Fe_{0.2}O_{3-\delta}$
C-ASU	cryogenic air separation unit
CCS	carbon capture and storage
CO_2 COMP	CO_2 compression and purification unit
COP	coefficient of performance
DENOX	denitrification unit
ESP	electrostatic precipitator unit
FG COND	flue gas condenser unit
FGD	flue gas desulphurization unit
HTM-ASU	high temperature membrane air separation unit
HX GG	regenerative gas/gas heat exchanger unit
HX RECOV	recovery heat exchanger unit
LCA	life cycle assessment
MIEC	mixed ionic electronic conducting
PSA	pressure swing adsorption
TIT	turbine inlet temperature
TPD	tons per day
TSA	temperature swing adsorption
VPSA	vacuum pressure swing adsorption

Symbols

A	membrane area, m^2
C_{Wagner}	Wagner conductivity constant, mol/(cm s K)
d	thickness, mm
F	Faraday constant, C/mol
JO_2	oxygen permeation rate, ml/(min cm^2)
k_{Wagner}	Wagner temperature constant, K
$\dot{m}$	mass flow, kg/s
P	pressure, bar
PO_2	oxygen partial pressure, bar
Ps	CO_2 separation pressure, bar
P_{vacuum}	vacuum pressure, mbar
R	ideal gas constant, J/(mol K)
$\dot{Q}$	heat, MW
SR_CO_2	CO_2 recovery rate, %
SR_O_2	oxygen separation ratio, %
T	temperature, °C
Ts	CO_2 separation temperature, °C
$\dot{W}$	power, MW
XCO_2	carbon dioxide purity, %
XO_2	oxygen molar fraction, dimensionless

Greek symbols

β	turbo-group compression ratio, dimensionless
Π	oxygen partial pressure ratio, dimensionless
σ	conductivity, S/m^2

Subscripts

e	electronic
el	electric
$feed$	feed side
i	ionic
$memb$	membrane
$perm$	permeate side
ret	retentate side
th	thermal

and SO_2). A comparative thermodynamic analysis between four-end membrane-based and cryogenic oxyfuel plants have been performed by Pfaff and Kather [7]. The three-end concept, on the other hand, does not present these technological problems, because no sweep gas is required, and membrane surfaces remain chemically stable without any contact with the contaminants present in the flue gas. Moreover, conventional flue gas cleaning and recirculation fan materials can be used because high flue gas temperatures are not required to heat the membrane unit.

Although the three-end concept can be a good technological option for membrane-based oxyfuel plants in the near future, the advantages of this process with respect to the cryogenic oxygen separation are still unclear for a variety of reasons. First of all, differing plant assumptions were considered in previous thermodynamic analysis found in the literature [8,9]. For accurate comparison to be possible, the following parameters should be similar in both technologies: the adiabatic combustion temperature, which has a direct influence on the amount of recycled flue gas and the energy required by the recirculation fan; the air infiltrations, which affect the flue gas composition and CO_2 purification and compression requirements; and the thermal plant integration degree, which has an important influence on the total plant efficiency. Secondly, an appropriated study considering the effect of the plant performance on the required membrane area, which represents an important criterion to evaluate the viability of this technology for oxyfuel power plants, is missing.

For these reasons, the purpose of this investigation is to compare the influence of the cryogenic and three-end HTM air separation units on the thermal performance of the oxyfuel power plant considering equivalent thermal integration and boundary conditions, as well as to analyze the impact of the plant operating conditions on the membrane unit design.

2. Oxyfuel power plants

In an oxyfuel process, air has to be treated in an air separation unit (ASU) to take out the oxygen required for combustion. Oxygen purity and recovery (O_2 separation ratio) have important influences on plant performance and depend on the ASU technology applied. Considering coal combustion with 15% of oxygen in excess, the amount of oxygen to be supplied by this unit is around 97 kg/s, whereas the total amount of flue gas recycled to the furnace corresponds to an adiabatic combustion temperature of 2120 °C. The cryogenic and membrane-based oxyfuel power plants presented are based on the advanced supercritical 600 °C coal-fired power plant Nordrhein-Westfalen concept (net efficiency of 45.9% and net power of 555 MW) [10], and were modeled using Aspen Plus® process simulation software. Plant performance calculations were done at ISO-conditions (ambient at 15 °C, 1.013 bar, and 60% relative humidity) and considering the common characteristics presented in Table 1. Kleinkopje hard coal was considered as fuel and its composition is shown in Table 2.

Simplified plant arrangement valid for both oxyfuel plants is depicted in Fig. 2. After combustion, the flue gas exiting the boiler at 350 °C is cooled down to 250 °C to heat the primary and secondary recycled flue gas streams via a regenerative gas/gas heat exchanger (HXGG unit). Then, the fly ash content is removed by a cold side electrostatic precipitator (ESP unit), which has a collecting efficiency of 99.9%.

Afterwards, the dedusted flue gas is separated into two streams: the secondary recycle flue gas which is heated up to 300 °C and sent back to the furnace, and the remaining flue gas which is further denitrified (DENOX unit), desulphurized (FGD unit), dehydrated and cooled down to 25 °C (FG COND unit). The cleaned flue gas is then split again into a primary recycled flue gas and a