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Study of a Natural Gas Combined Cycle with Multi-Stage Membrane Systems for CO_2 Post-Combustion Capture

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Abstract

Carbon capture based on membrane process is receiving an increasing attention, due to advances in membrane performances. The aim of this paper is to investigate the integration of a post-combustion capture system based on membrane separation into a natural gas combined cycle (NGCC) power plant. A sensitivity analysis is carried out in order to evaluate the effect of membrane technology and operating conditions on permeate purity, membrane area and energy requirement for CO_2 capture. Hence, energy and environmental performances of the NGCC integrated with the separation unit are assessed and compared to a baseline power plant without carbon dioxide removal.

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Keywords: Carbon capture; Combined cycle; Polymeric membranes; EGR; CO_2 purity; Capture ratio.

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1. Introduction

Carbon capture and storage (CCS) is considered a key option for mitigating carbon dioxide emissions from industrial facilities and fossil fuel power plants, that represent the major emitters of greenhouse gases related to global climate changes [1].

Amine-based absorption represents the reference technology for CO_2 post-combustion capture, since it is capable to treat huge flue gas flows, ensuring a high level of carbon dioxide removal [2]. This process is well-known as it has been utilized in natural gas treatment, food processing and chemical industries for decades [3].

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However, many challenging problems still hinder the deployment of such technology for CO_2 removal. The most important is undoubtedly the remarkable energy requirement for solvent regeneration, that states at about 3.5 MJ/kg_{CO2} for the conventional MEA-absorption process, corresponding to 50-60% of the steam in the low-pressure turbine of a power plant. Hence, this issue leads to a penalization of power plant efficiency, as well as a reduction of rated capacity. In addition, deeper investigations are required in order to evaluate all the issues related to thermal and oxidative solvent degradation, equipment corrosion and environmental hazards [4]. An alternative option to amine absorption is represented by membrane separation process. Such technology is faced with different issues, including the low CO_2 concentration, the need of cooling flue gas to avoid membrane degradation and the energy requirement to create the driving force for separation process [5]. In spite of these issues, carbon capture based on membrane process is receiving an increasing attention, mainly due to advances in materials technology, that are pushing membrane performances beyond the trade-off limit between permeability and selectivity. Other important advantages include no regeneration, a modular design, operational simplicity and low environmental impact.

CO_2 capture based on membrane processes has been the subject of many studies. However, many of these have focused the attention on the membrane system only [5-9], with the aim to explore the effect of membrane system layout and operating conditions on capture system performances. Such studies have highlighted that a single-stage system cannot achieve the target for CO_2 purity (90%) and capture ratio (90%) with the current state of membrane material technology. On the other hand, few investigations have dealt with the integration of membrane processes for carbon capture into power generating systems, limiting the investigation to coal fired power plants [10-11].

This paper aims to investigate the integration of a post-combustion carbon capture based on membrane separation processes into a natural gas combined cycle (NGCC) power plant, having a three pressure and reheat heat recovery steam generator (HRSG). In order to reduce the gas flow to be treated and increase the carbon dioxide concentration, the steam-gas power plant is provided with a partial recirculation of flue gas. Due to the thermodynamic conditions of the flue gas to be treated, the driving force for CO_2 separation is artificially created by using a feed compression with an energy recovery system, that also allows to reduce the temperature of flue gas down to the maximum allowable temperature of polymeric membranes [12].

In this paper two types of energy analysis are carried out. The first one examines the effect of the membrane system technology and operating conditions on different figures of merit, including permeate purity, membrane area and specific energy requirement for compressing the flue gas at membrane inlet. The second analysis assesses the impact of carbon capture system integration on energy and environmental performances of the NGCC, considering commercial and next generation membrane technologies.

Nomenclature

Symbols

A	membrane area, m^2
E	Energy, kWh
m	Mass flow rate, kg/s
p	Permeability, Barrer
P	Power, MW
x	Molar fraction, %

Greek letters

α	Selectivity
β	Pressure ratio
η	Efficiency, %
φ	CO_2 capture ratio, %

Subscripts

ECS	Exhaust compression system
EXH	Exhaust
PCS	Permeate compression system
tot	Total

Acronyms

CCS	Carbon capture and storage
EGR	Exhaust gas recirculation
HRSG	Heat recovery steam generator
LHV	Lower heating value
MEA	Monoethanolamine
NGCC	Natural gas combined cycle

2. Simulation of a CO_2 removal system based on a multi-stage membrane separation process

The membrane system is simulated through a one-dimensional hollow-fiber model, assuming steady state conditions and a countercurrent mode of operation. The model allows to evaluate, through mass balances on discretized membrane elements, the permeate purity, the membrane area and the energy expenditure for exhaust compression system (ECS), given the capture ratio (φ) and membrane properties, i.e. CO_2 permeability (p_{CO2}) and selectivity over O_2 ($\alpha_{CO2/O2}$) and N_2 ($\alpha_{CO2/N2}$) [13]. Figure 1 shows the layout of the membrane system investigated. The flue gas to be treated is first dewatered using a flash tank and then compressed (β=20) in a two-stage compression unit with intercooling, integrated with an energy recovery system; hence, the compressed flue gas passes through a dual-stage membrane system based on the enricher concept [7]. In this configuration, the permeate of the first membrane is compressed, sent as feed gas to the second membrane and then to the compression section for the CO_2 transport and storage. The retentate of the second stage is recirculated back to the feed side of the first stage, with the aim to increase the CO_2 concentration at feed inlet and then the permeate purity with the same membrane parameters and operating conditions. The retentate of the first stage is sent to the atmosphere, after expanding in a two-stage turbine that allows to reduce the compressors power requirement.

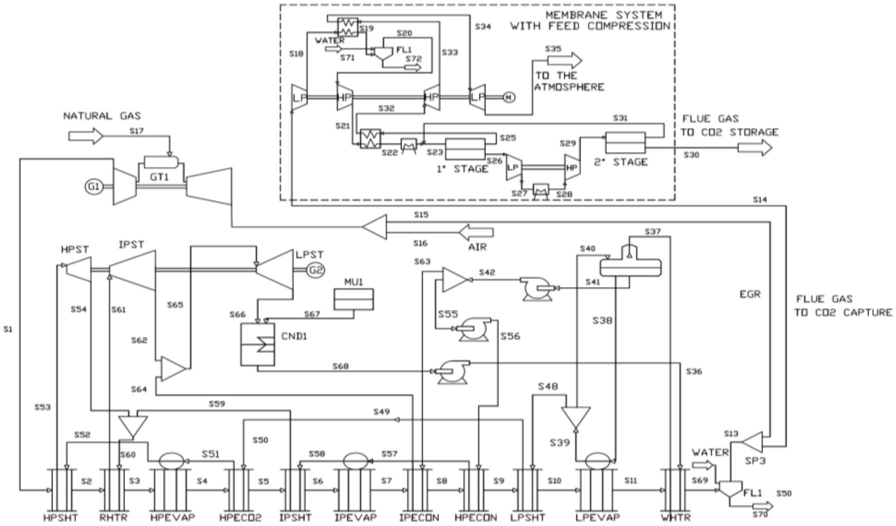

Figure 1. Layout of NGCC integrated with the CO_2 capture system based on membrane separation process.

In this paper a sensitivity analysis is carried out to explore the impact of CO_2 concentration at feed inlet ($x_{CO2,F}$), membrane material properties (p_{CO2}, $\alpha_{CO2/O2}$, $\alpha_{CO2/N2}$) and number of stages (N_s) on permeate purity, membrane area and specific energy requirement for the exhaust gas compression, varying the targeted carbon capture ratio.

2.1. Effect of CO_2 concentration at membrane inlet

The low concentration of carbon dioxide in the flue gas of a NGCC power plant requires special precautions in order to increase its molar fraction, thus enhancing the driving force for CO_2 separation. This objective can be achieved through a partial recirculation of the exhaust flue gas (EGR) at gas turbine inlet, that simultaneously allows to reduce the mass flow to be treated, with beneficial effect on membrane surface. The effect of EGR on membrane system performances have been quantitatively assessed, assuming p_{CO2}=325 Barrer, $\alpha_{CO2/O2}$=17.5 and $\alpha_{CO2/N2}$=42.5. Table 1 summarizes the flue gas composition and mass flow rate at membrane system inlet, varying EGR from 0 to

Table 1. Flue gas composition at membrane system inlet varying EGR.

	m_{EXH} (kg/s)	$x_{CO2,F}$ (%)	$x_{O2,F}$ (%)
EGR=0%	590.5	4.3	13.2
EGR=20%	470.1	5.4	11.2
EGR=40%	350.8	7.4	7.6
EGR=60%	233.4	11.1	0.9

60%. Increasing EGR, the molar fraction of CO_2 increases from 4.3 to 11.1%; conversely, O_2 concentration reduces from 13.2 to 0.9%, positively affecting the permeate purity achievable through the membrane separation process.

Figure 2a shows that the increase of permeate purity due to EGR is more and more pronounced at increasing of capture ratio. For instance, at EGR=40%, the increase of permeate purity with respect to EGR=0% passes from 4 (97%) to 11 percentage points (89%), as φ increases from 60 to 90%. Simultaneously, the membrane area decreases, due to concurrent effects of the reduced flue gas mass flow rate and the increased driving force for CO_2 separation. As shown in Fig. 2b, the membrane area decreases of about 40% at φ=90%, passing from 394.7*10^3 m^2 (EGR=0%) to 220.4*10^3 m^2 (EGR=40%). The exhaust gas recirculation positively affects the power consumption for feed compression (Fig. 2c), due to the reduced exhaust flow rate. Hence, increasing EGR from 0 to 40%, the specific energy requirement reduces of about 40%, passing from 0.71 to 0.45 kWh/$kg_{CO2,P}$ at φ=90%.

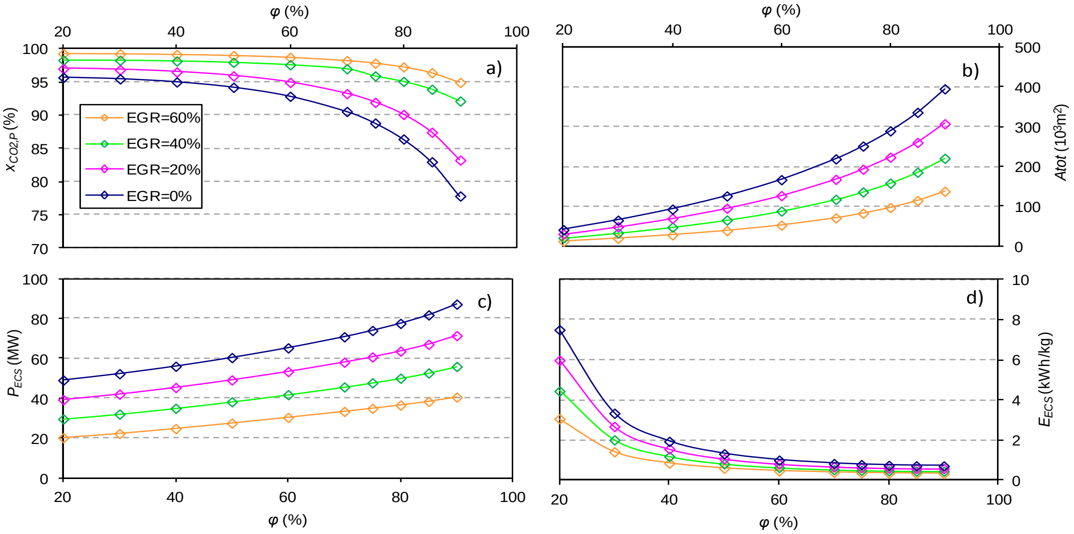

Figure 2. Effect of exhaust gas recirculation on membrane system performances.

It is noteworthy to observe that the increase of EGR is constrained by the O_2 concentration at the combustor inlet, which cannot fall below 16%, in order to ensure the flame stability and then a high combustion efficiency [14]. Hence, the EGR cannot exceed 35%, unless of technical adaptations of combustion chamber to allow an oxygen air enrichment.

2.2. Effect of membrane technology

Performances of gas separation processes are strictly related to membrane material technology. Suitable candidates for CO_2 removal in post-combustion processes are polymeric membranes, that allow to separate CO_2 through a solution-diffusion mechanism [12]. Structure and property studies on polymeric membranes have highlighted the existence of a trade-off limit between permeability and selectivity. Hence, membranes with higher permeability show a low selectivity and vice versa. In this paper, the impact of membrane properties on separation performances is investigated considering four types of polymeric membranes, that differ in terms of CO_2 permeability and

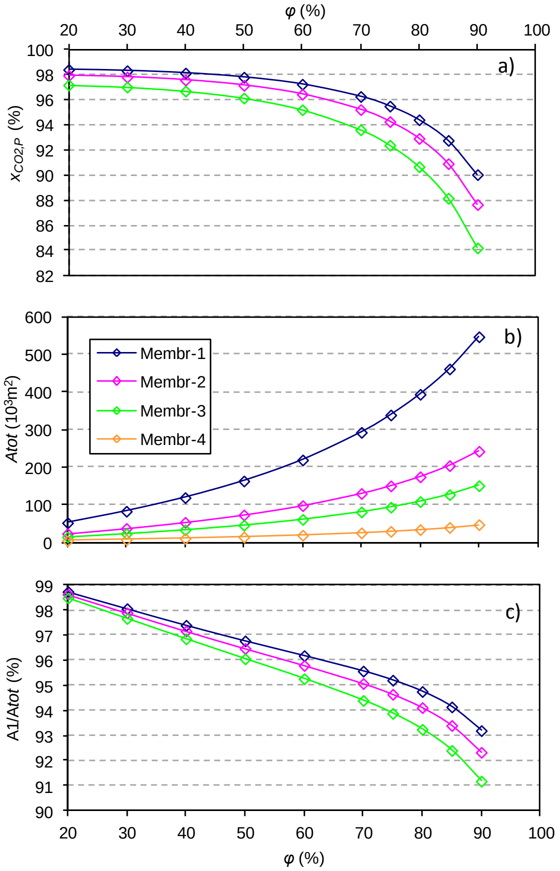

Figure 3. Effect of membrane technology on CO_2 purity (a), total membrane area (b) and incidence of the first membrane stage on total membrane area (c).

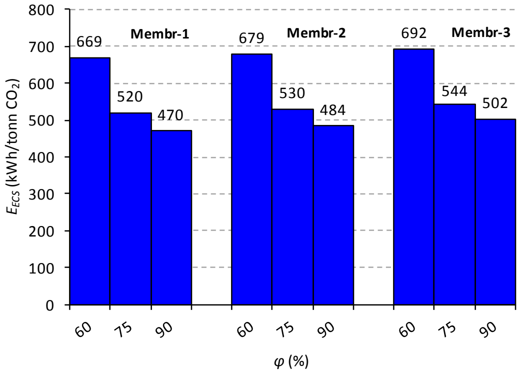

Figure 4. Effect of membrane technology on the specific energy requirement for feed compression.

Table 2. Membrane properties.

	p_{CO2} (Barrer)	$\alpha_{CO2/O2}$	$\alpha_{CO2/N2}$
Membr-1	150	20	50
Membr-2	325	17.5	42.5
Membr-3	500	15	35
Membr-4	1600	15	35

selectivity over O_2 and N_2. As shown in Table 2, passing from *Membr-1* to *Membr-4*, CO_2 permeability increases from 150 to 1600 Barrer, while selectivities over O_2 and N_2 reduces from 20 to 15 and from 50 to 35 respectively. In addition, *Membr-4* has the same $\alpha_{CO2/O2}$ and $\alpha_{CO2/N2}$ of *Membr-3*, but with a higher p_{CO2}, being representative of a cutting-edge membrane technology.

Figure 3 summarizes the sensitivity of capture system to membrane properties, having set EGR to 35%. The simulation results show that the permeate purity, while reducing at increasing of φ, remains higher than 80% in all cases investigated (Fig. 3a). As expected, *Membr-1* performs the best permeate purity, due to the higher values of $\alpha_{CO2/O2}$ and $\alpha_{CO2/N2}$. Passing from *Membr-3* to *Membr-1*, the permeate purity gain increases with capture ratio; hence, rising φ from 60 to 90%, the permeate purity reaches about 97% (+2 percentage points) and 90% (+6 percentage points) respectively. On the other hand, due to the low CO_2 permeability, *Membr-1* requires the highest membrane area, that exceeds $300*10^3$ m^2 for φ>70% (Fig. 3b). In addition, it is noteworthy to observe that *Membr-3* and *Membr-4* achieve the same level of CO_2 purity, having the same values of $\alpha_{CO2/O2}$ and $\alpha_{CO2/N2}$, but *Membr-4* allows to drastically reduce the membrane area, that passes from $151*10^3$ to $47*10^3$ m^2 at φ=90%. Finally, Fig 3c highlights that the first membrane stage has the highest impact on the total membrane area, contributing to over than 90%, even for the higher values of φ.

Figure 4 compares the specific energy required for feed compression of *Membr-1*, *Membr-2* and *Membr-3*, varying the capture ratio from 60 to 90%. *Membr-4* has been omitted, having the same E_{ECS} of *Membr-3*. It is highlighted that E_{ECS} has an asymptotic decreasing trend with capture ratio, being its reduction less pronounced for φ>75%. In addition, for a fixed capture ratio, E_{ECS} reduces from *Membr-3* to *Membr-1*, due to the rise of selectivity values that increase the power production from retentate expansion; for instance, if φ is set at 90%, the specific energy requirement passes from 502 to 470 kWh/tonn$_{CO2}$.

2.3. Effect of number of membrane stages

The effect of membrane system layout on gas separation performances has been evaluated, assuming *Membr-2* (p_{CO2}=325 Barrer, $\alpha_{CO2/O2}$=17.5 and α_{N2}=42.5) as reference technology. In this respect, Fig. 5 compares the performances of CO_2 removal system, varying the number of membrane stages. As expected, a single-stage configuration does not allow to ensure a satisfactory permeate purity, regardless of targeted capture ratio. Conversely, the CO_2 purity is nearly 100% when a three-stages membrane system is used (Fig. 5a).

The increase of membrane area is noticeable in the passage from 1- to 2-stage configuration for higher values of capture ratio, while it is negligible with the addition of a third stage. For instance, at φ=90%, passing from 1- to 2-stage configuration, membrane area rises to 242*10^3 m^2 (+20%), reaching 250*10^3 m^2 (+3%) in the case of 3-stage configuration (Fig. 5b). In addition, the first stage always plays the most important role, accounting for over than 90% of the total membrane area, regardless of the number of membrane stages.

According to Fig. 5c, the power consumption for exhaust gas compression increases, due to the need for recompressing the permeate arising from the first and the second stage. However, the power required by the additional stages is more contained due to the smaller fraction of exhaust flow rate processed by the compressors (Fig. 5c). As a result, the increase of specific energy requirement for exhaust compression reduces with the capture ratio, stating at about +60% (0.57 kWh/kg$_{CO2,P}$) with φ=90%.

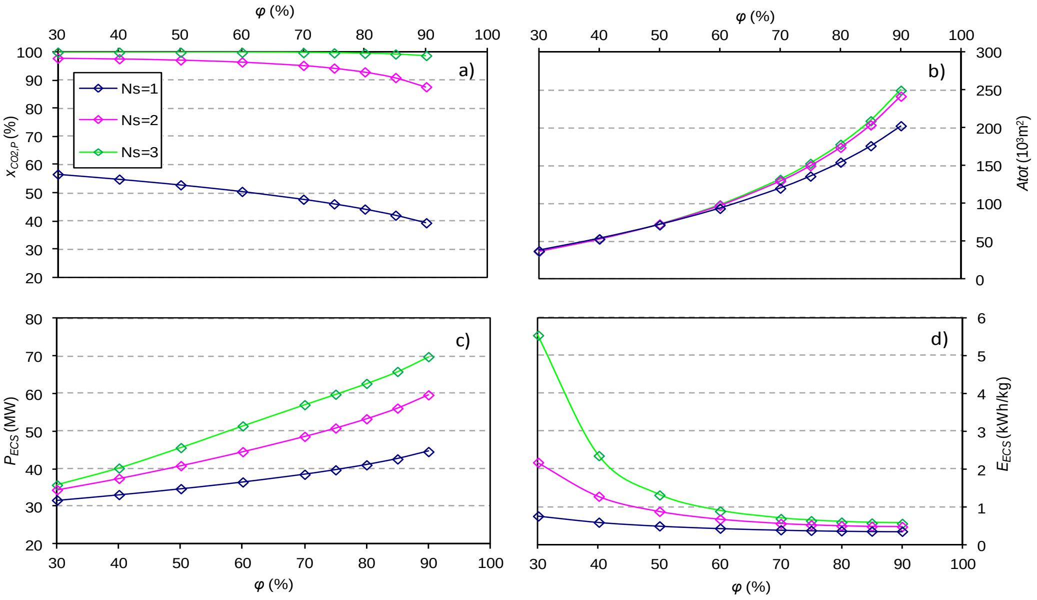

Figure 5. Effect of stages number on membrane system performances.

3. Effect of membrane system on NGCC energy performances

The capture system based on membrane separation process has been integrated into a natural gas combined cycle with a three pressure and reheat HRSG. The integrated system (Fig. 1), is composed of two main blocks; the power

Table 3. Main operating parameters and energy performances of gas turbine and combined cycle.

Operating parameters	Value
Gas turbine	
Model	GE PG9351(FA)
Compressor pressure ratio	14
Exhaust gas flow, kg/s	622.8
Exhaust gas temperature, °C	608.5
Power output, MW	254.5
Net efficiency, %	36.9
Combined cycle	
HP steam pressure, bar	130
IP steam pressure, bar	30
Dearator pressure, bar	5
HPEVAP ΔTpp, °C	10
IPEVAP ΔTpp, °C	10
LPEVAP ΔTpp, °C	10
Condenser pressure, bar	0.05
NGCC power output, MW	382.5
Net efficiency, %	55.5
Specific CO_2 emissions, g/kWh	358.7
T_{STACK}, °C	94
$x_{CO2,\ STACK}$, %	3.94

block, represented by a combined cycle power plant with exhaust gas recirculation and the membrane system with feed compression. As shown in Fig. 1, the exhaust flue gas at stack is cooled down to 25°C in a flash tank, thus allowing the separation of the excess condensed water. Hence, a fraction of flue gas (EGR=35%) is recirculated and mixed to the air at gas turbine inlet, while the remainder is sent to the feed compression system, that produces the driving force for CO_2 separation required by the two-stage membrane system configuration. Due to the intercooled compression, the temperature of compressed flue gas at membrane inlet is reduced to 50°C, in order to ensure the stability of membrane material properties [15]. The CO_2-enriched permeate stream at membrane outlet is then compressed at 100 bar by a 5-stages compression unit with intercooling and sent to CO_2 storage. All components of the integrated system have been simulated using the GateCycle software of General Electric [16], with the exception of two-stage membrane system that has been modeled and integrated into the power system using the Excel-Visual Basic environment.

At baseline conditions (EGR=0%, φ=0), the NGCC has a rated capacity of about 380 MW, an efficiency of 55.5%, and specific CO_2 emissions of 358.7 g/kWh. Table 3 summarizes the main operating conditions of gas turbine and combined cycle, as well as their energy performances at baseline conditions.

Performances of NGCC with CO_2 removal system have been evaluated and compared to the baseline power plant, considering different membrane materials and pressure ratio of feed compression system.

3.1. Effect of membrane technology

The energy performances of NGCC with CO_2 capture system have been evaluated for three types of membrane materials [5], whose properties are summarized in Table 4. The membranes selected for investigation include two commercial membranes, Polyactive® and PEBAX®/PDMS-PEG, having permeability of 1590 Barrer and 326 Barrer respectively and a next generation membrane based on thermally rearranged polybenzimidazole (TR-PBI), having the same permeability of Polyactive, but with a selective layer thickness of 0.5 μm. As shown in Fig. 6, the Polyactive® membrane allows to achieve a permeate purity higher than 80% with φ=90%, being the total membrane area of about $50*10^3$ m^2; in the case of PEBAX®/PDMS-PEG membrane, the permeate purity is slightly higher, in spite of a substantial increase of the total area ($x_{CO2,P}$=87%, $Atot$=$238*10^3$ m^2). On the other hand, the permeate purity with TR-PBI is quite unsatisfactory for φ>70%, falling below 80%, while the membrane area drastically reduces to about 10-$20*10^3$ m^2, due to the rise of CO_2 permeability.

Figure 7 shows the effect of capture system integration on the energy performances of NGCC, varying the type of membrane technology. As shown in Fig. 7a, the efficiency decreases with capture ratio, due to the increase of power consumption required for the compression of the exhaust gas at membrane inlet and the CO_2-enriched permeate stream. At φ=90%, the efficiency penalty compared to the baseline power plant (without carbon capture system) ranges from -13 (TR-PBI) to -11 percentage points (PEBAX®/PDMS-PEG and Polyactive®), corresponding to a reduction of power plant capacity of -24% and -19% respectively (Fig. 7b). It is noteworthy to observe that the total specific energy requirement (E_{TOT}) shows the minimum value at

Table 4. Properties of membranes selected for investigation.

	CO_2 permeance (GPU)	$\alpha_{CO2/O2}$	$\alpha_{CO2/N2}$
Polyactive®	1590	12.8	36
PEBAX®/PDMS-PEG	326	17.1	40.1
TR-PBI	3185	4.6	25

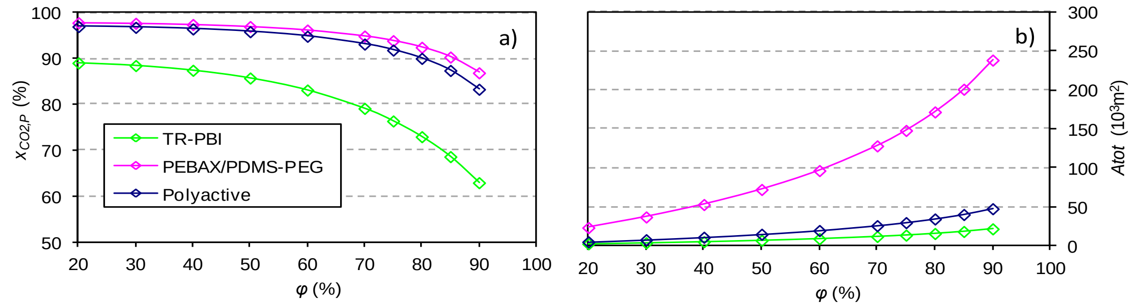

Figure 6. CO_2 purity (a) and total membrane area (b) of membrane selected for investigation.

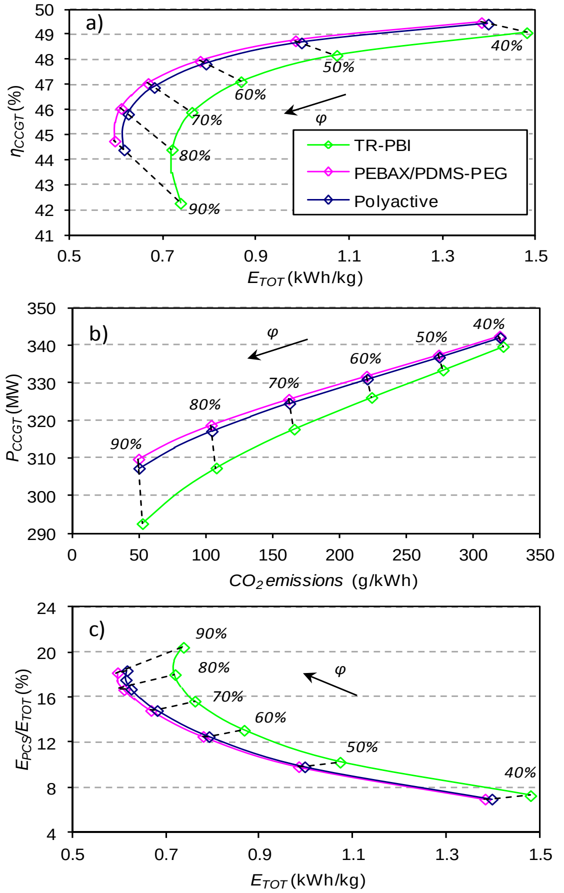

Figure 7. Effect of membrane technology on CCGT efficiency (a), rated capacity (b) and incidence of CO_2 compression on total power consumption (c).

capture ratio of about 80-90% (Fig. 7a), corresponding to the lowest values of specific CO_2 emissions. For instance, in the case of TR-PBI, the CO_2 emission reaches about 100 g/kWh, while it further reduces to 50 g/kWh in the case of PEBAX®/PDMS-PEG and Polyactive® (Fig. 7b).

The contribution for the compression of CO_2-enriched stream ranges from 8 to 20% of the total specific energy consumption, being the higher values achieved at higher capture ratio (Fig. 7c).

The low permeate purity of TR-PBI suggests to investigate a membrane system with a higher number of stages. In this respect, Fig. 8 highlights that, at increasing of capture ratio, the increase of permeate purity with a 3-stages configuration is more pronounced, at the expense of a higher efficiency reduction, related to the increase of energy consumption of feed compression system. Hence, at φ=90%, $x_{CO2,\,P}$ rises from 63% to about 80% (Fig. 8a); as shown in Fig. 8c, the reduction of power consumption for CO_2 compression (-21%) due to the increased purity is more than offset by the increase of energy expense (+22%) related to the recompression of permeate from the second stage. Hence the net efficiency reduces from 42.3 to 40.6%, that corresponds to an efficiency penalty of about 15 percentage points.

3.2. Effect of pressure ratio of the exhaust compression system

Simulation results have shown that the Polyactive® membrane allows to achieve a good compromise between permeate purity, membrane area and energy performances of NGCC. Hence, focusing the attention on this technology, a further investigation has been carried out with the aim to explore the influence of pressure ratio of exhaust compression system. In this respect, Fig. 9 shows that the increase of pressure ratio has a beneficial effect on permeate purity and membrane area, that is more and more pronounced at increasing of φ.

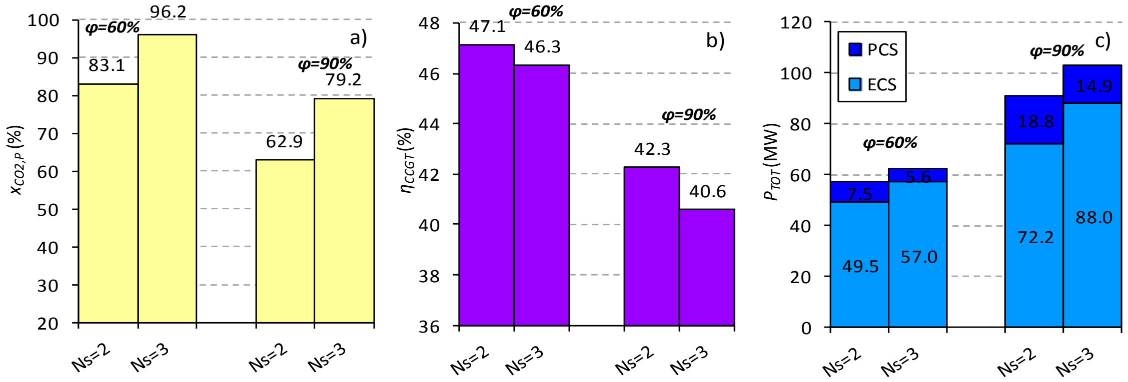

Figure 8. Comparison of 2 and 3-stages configurations for TR-PBI membrane: effect on permeate purity (a), NGCC efficiency (b), relative contribution of power consumption for the compression of feed at membrane inlet and CO_2-enriched stream (c).

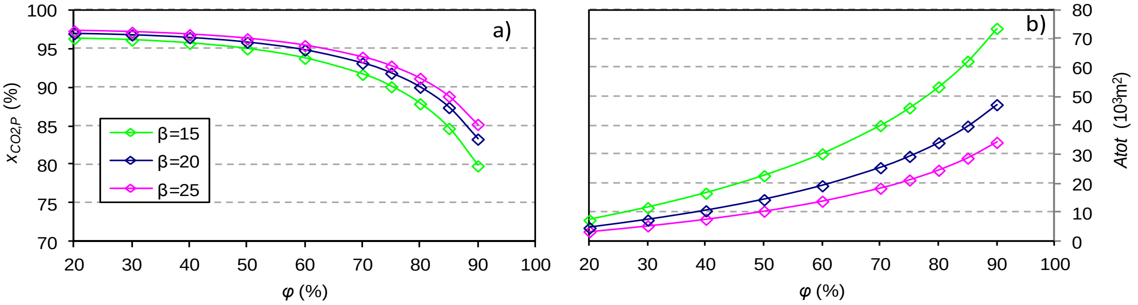

Figure 9. Effect of pressure ratio on CO_2 purity (a) and total membrane area (b) of Polyactive® membrane.

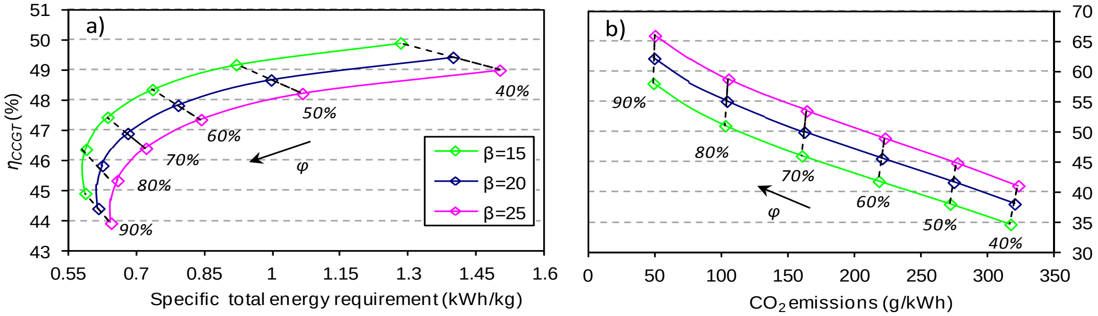

Figure 10. Effect of pressure ratio on NGCC efficiency (a), and power consumption for the exhaust compression (b).

For instance, increasing β from 15 to 25 at φ=90%, the permeate purity rises from 80 to 85%, while membrane area drastically reduces, passing from $73.5*10^3$ to about $34*10^3$ m^2. As expected, due to the increase of power consumption of exhaust compression system (Fig. 10b), the net efficiency reduces of about 1 percentage point at a fixed value of capture ratio, stating at 44% for φ=90% (Fig. 10a); on the other hand, the specific energy consumption increase becomes less pronounced at increasing of φ, due to the increase of permeate purity that positively affects the power consumption for the compression of CO_2-enriched stream. Hence, increasing β from 15 to 25, the specific energy consumption increases of 14.5% and 9.7% for φ=60% and 90%, stating at 0.84 and 0.64 kWh/kg respectively. From the environmental point of view, it is noted that the increase of pressure ratio produces a negligible increase of specific CO_2 emissions (Fig. 10b).

4. Conclusions

The aim of this paper has been the energy analysis of a natural gas combined cycle integrated with a post-combustion CO_2 removal system based on membrane separation process. The membrane system is represented by a dual stage configuration with feed compression and recirculation of retentate at first stage inlet.

Focusing on membrane system only, simulations results have highlighted that EGR positively affects the permeate purity and membrane area, due to concurrent increase of CO_2 concentration and the reduction of flue gas flow to be treated. The increase of CO_2 permeability allows to further reduce membrane area, in spite of a decrease of permeate purity arising from the lower selectivity over N_2 and O_2. On the other hand, the specific energy requirement for CO_2 compression is virtually unaffected by membrane properties, with a maximum variation of about 7% at φ=90%. Passing from 2- to 3-stages configuration, permeate purity substantially rises, in spite of an increase of specific energy expense for feed compression of about 20% at φ=90%.

The energy analysis of the NGCC integrated with the capture system has shown that the Polyactive® membrane (p_{CO2}=1590 Barrer, $\alpha_{CO2/O2}$=12.8 and α_{N2}=36) allows to achieve the best compromise between membrane performances and the energy expense for the compression of exhaust flue gas and the CO_2-enriched permeate stream. Assuming φ=90%, the permeate purity is about 83%, against a membrane area of $47.1*10^3$ m^2 and a total specific energy requirement of 0.62 kWh/kg, being the contribution for CO_2 compression less than 20%. Hence, the efficiency penalty of NGCC compared to the baseline power plant without CO_2 removal system is about 11 percentage points, corresponding to the lowest specific CO_2 emissions (49.6 g/kWh). Finally, the increase of feed compression pressure ratio from 20 to 25 allows to improve membrane performances both in terms of CO_2 purity and membrane area, against to an increase of specific energy requirement of about 5%, leading to an efficiency penalty increase less than 1 percentage point.

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