Optimization of CO_2 Capture from Natural Gas Combined Cycle with Hydrogen-Assisted Exhaust Gas Recirculation

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Abstract. CO_2 capture and storage (CCS) is crucial for reducing emissions in areas with limited renewable energy or to balance their intermittent nature. CCS, however, reduces the efficiency of gas turbine combined cycles due to the high air-to-fuel ratio, resulting in low CO_2 concentration in exhaust gases. Exhaust Gas Recirculation (EGR) can increase CO_2 levels but is limited to about 35% due to combustion stability issues. The EU project TRANSITION explores using hydrogen to pilot burners, allowing higher EGR rates without compromising combustion stability. Experimental data on emissions and Lean Blowout resistance were used to model the impact of hydrogen-assisted EGR on CO_2 capture and overall efficiency. Results indicate that higher EGR rates reduce CO_2 capture energy requirements and equipment sizes, with minimal additional energy costs for EGR systems. The study aims to optimize hydrogen use for maximum efficiency gains while considering costs, safety, and auxiliary energy use. Additionally, the integration of hydrogen-assisted EGR shows promise in enhancing the overall performance of gas turbines, making them viable for low-carbon power generation. This approach could significantly contribute to the energy transition by enabling more efficient and stable CCS in gas turbine systems.

1 Introduction

The combustion of fossil fuels significantly increases CO_2 levels in the atmosphere, contributing to global warming and its adverse effects on weather, biodiversity, marine environments, and food security. The COP28 conference emphasized the need to transition away from fossil fuels. CO_2 capture and storage (CCS) is a key strategy for mitigating global warming [1]. Natural gas power plants with CCS are seen as a short-term solution for climate-friendly energy. However, integrating CO_2 capture in gas turbine systems reduces efficiency and increases area requirements. A major source of the energy penalty is the intrinsically high air-to-fuel ratio at which gas turbines operate, leading to low CO_2 concentration in the exhaust gases. The established, although not commercial, method to counteract this shortcoming is to implement Exhaust Gas Recirculation (EGR) technology by recirculating a portion of exhaust gases back into the compressor to the gas turbine, increasing the CO_2 content [2,3]. The higher the EGR, the larger the CO_2 concentrations, however a combustion stability barrier quickly limits the EGR rate to approximately 35% [2].

EGR also decreases NOx emissions and improves part-load efficiency, but lowers O_2 concentration, affecting combustion stability and efficiency [4]. Hydrogen piloting can mitigate these issues, allowing higher EGR levels. The study performed under the Horizon Europe project TRANSITION (grant number 101069665) evaluates the potential and challenges of integrating H_2-assisted EGR with CO_2 capture in gas turbines, focusing on energy requirements, power demand, and equipment sizes, which are crucial for operating and capital costs [5]. This analysis forms the basis for further economic evaluations. Adding hydrogen for piloting the combustion represents a cost, a safety concern, and an auxiliary energy use, therefore an objective of the study is to optimize the effort of adding H_2 against its benefit in terms of overall (power cycle + capture) efficiency gain. In addition, an extensive combustion study covers the design of H_2 piloting to an existing commercial combustor, numerical CFD simulations, and testing of the prototype at various power and pressure scale.

2 Procedures

2.1 Experimental research

A first experimental campaign was conducted at the THT Lab of the University of Florence on an industrial burner tested at atmospheric pressure. The rig, operating under relevant gas turbine conditions, is equipped with an optical access to visualize the flame and employing oxidizer compositions mimicking EGR operation. The

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aim was to characterize the burner performance, with CO emissions as the primary performance metric under the investigated conditions, since they critically increase with high EGR rates. NOx emissions were also measured, and the dynamic behavior of the flame was monitored to assess the presence of thermoacoustic oscillations. Additionally, advanced optical diagnostic techniques were employed to analyze flame behavior, enabling a deeper understanding of how operating conditions influence dynamics, stability, and pollutant formation.

The single-sector tubular combustion chamber consists of a quartz cylindrical liner enclosed in an outer vessel, as shown in Fig. 1. The investigated burner is an aero-derivative concept from Baker Hughes, currently installed on medium sized plants fuelled with natural gas. With the goal of retrofitting existing systems and gas turbines, the first phase of the study focused on an established design, introducing the addition of a small hydrogen amount with the purpose of increasing the flame reactivity. Additionally, different burner configurations were tested to assess the influence of various geometric parameters, and the optimal operating point was identified in terms of the pilot-to-premixed fuel ratio and hydrogen content, in order to improve burner performance under the challenging conditions imposed by EGR.

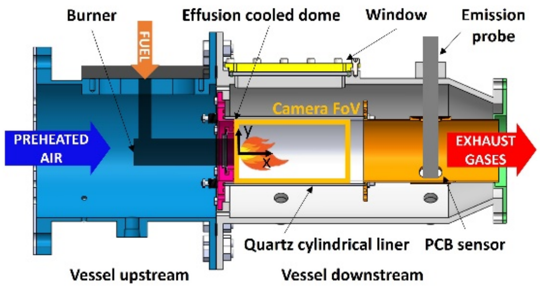

Fig. 1. Optical reactive test rig employed for the experimental activity at ambient pressure.

Recreating real EGR composition in lab scale is quite challenging, and CO_2 enriched mixtures have been considered a good trade-off in this investigation phase. A restricted set of tests was also repeated with nitrogen addition to observe the effect of the oxidizer composition on the reactive process. In the experiments, combustion air is diluted with CO_2/N_2 to lower the oxygen level, injecting the inert gases in the preheated airflow upstream of the reactive rig. EGR levels are represented by the inlet oxygen mass fraction (YO_2), and tests have been conducted at constant total thermal power to compare different operating conditions with the dual-fuel configuration and different oxygen levels. The hydrogen fraction was maintained below 10% of the total thermal power to improve combustion stability without drastically decreasing the CO_2 concentration at the exhaust because of the combustion of a carbon-free fuel, as it will be also shown in the Results section. Further details on the experimental setup, operating conditions and main findings can be found in previous works [6,7].

2.2 Process simulations

A fit-for-purpose model of a GT, along with an in-house tool to model the CO_2 capture process, were employed to capture the key effects of EGR. The general scheme of the system considered in this study is shown in Fig. 2. The reference GT considered is the MS9001 from GE [8], modeled through the ESMS code developed by Carcasci et al. [9,10], an in-house modular tool based on a Gauss-Jordan solver. The developed fit-for-purpose model of the GT together with EGR, with and without H_2 piloting, allows to evaluate the system at several EGR rates, defined as the ratio between the flowrate of the recycled stream and the flowrate of the exhaust gases, as in Equation (1):

$$EGR = \frac{\text{Recycled stream flowrate}}{\text{Exhaust gas flowrate}} \quad \text{Eq. (1)}$$

The EGR rate is varied between 0 and 0.65 (higher EGR rates would arguably be unfeasible due to severa lack of oxygen and combustion instability). The results obtained, in terms of exhaust gas compositions and flowrates, represent then the inputs for the CO_2 capture system, an MEA-solvent based absorption process simulated using the iCCS tool developed by SINTEF Energy [11].

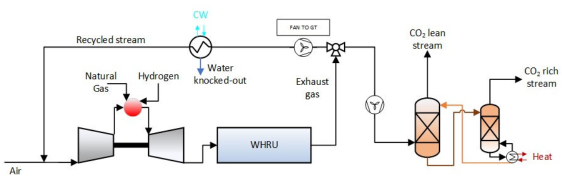

Fig. 2. General scheme of the power plant process.

Simulations have been performed at 0% H_2 piloting, 5% H_2 piloting and 10% H_2 piloting (in line with the experimental campaign), where the percentage indicated represents the energy contribution associated with the H_2 addition. Consequently, the H_2 piloting accounts for 0%, 5% and 10% of the GT total power outlet, respectively, keeping the total power constant in the three cases.

A parametric study is carried out to evaluate the EGR effect on CO_2 concentration in the exhaust gases and O_2 level at the combustor, which respectively influence the CO_2 capture energy requirement and sizes, and the combustion operability.

The additional effect of the EGR on the system can be evaluated in terms of the global power demand, considering additional power demands in the system (pumping and fan power).

In this study, the performance of a power plant based on the commercially available MS9001E engine is examined. The MS9001E heavy-duty gas turbine is General Electric's, specifically the 9E.04 type [8]. The engine is of the single-shaft type. The compressor is a 17-stage, axial flow design. The combustion chamber is of the multi-can type. The turbine is four stages and its blades and nozzles are air-cooled. The developed ESMS code [9] is able to assess the plant's performance under various operational conditions. Its modular nature allows to describe the power plant configuration by connecting elementary components, representing diverse unit operations like compressors, combustion chambers, mixers, etc. Each component operates as a black box capable of simulating specific chemical and thermodynamic transformations. The resultant set of nonlinear equations defining the power plant is linearized, with coefficients updated during calculations. Employing a classic matrix method, all equations are solved simultaneously, reflecting the essence of a fully implicit linear approach. Design and off-design simulation is a two-step process, where off-design performance simulation involves fixed geometry obtained during the design study, thus reducing the number of input data. In the off-design simulations, the fuel flowrate is regulated to keep the Turbine Inlet Temperature (TIT) constant at the design value, i.e., 1146 °C.

The first technology selected for CO_2 capture is by absorption as it is a mature and commercially available technology. The CO_2 capture scheme is presented in Fig. 3. The exhaust gas is first cooled to an appropriate temperature through a Direct Contact Cooler (DCC) and then sent to the absorption column. In the absorber column, the exhaust gas is contacted with an amine-based solvent (MEA). The CO_2 is absorbed from the gas to the liquid solvent, and the cleaned gas is vented from the top of the absorber. The solvent is sent from the absorber to the desorber, where the solvent is regenerated thanks to higher temperatures, and the CO_2 passes in gas phase, while the regenerated solvent is sent back to the absorber. The resulting CO_2-rich gas stream is ready for conditioning and ultimately transport to a sequestration site. The heat is provided by a steam reboiler at the bottom of the desorber, representing the main energy consumption of the capture. At the top of the desorber, a condenser provides the liquid return to the column.

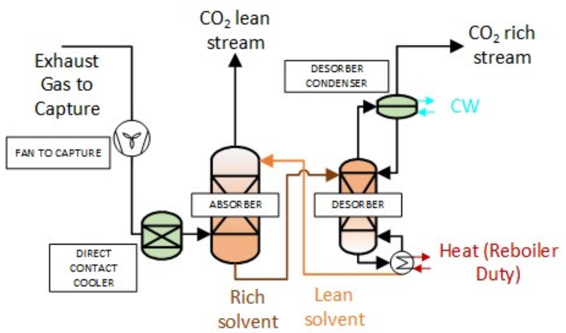

Fig. 3. CO_2 capture scheme based on solvent absorption.

The amine loading is defined as the amount of CO_2 in the solvent (units are molCO_2/molMEA). When the solvent has been regenerated, it is considered lean, and the amount of CO_2 contained in the lean solvent is defined as lean loading. When the solvent has absorbed CO_2, it is defined as rich, then the amount of CO_2 contained in the rich solvent is defined as rich loading. Both are calculated as a function of the CO_2 concentration in the EG. The liquid flow ratios of lean and rich solvents are calculated through the molar balance, considering the amount of CO_2 entering the absorber, the rich and lean loading, and the capture ratio. The L/G ratio is consequently the ratio between the lean solvent flowrate and the exhaust gas flowrate. As the CO_2 process design is optimized for each CO_2 concentration, the liquid to gas ratio in the absorber and the lean amine loading are allowed to vary accordingly. The CO_2 capture rate, defined as the percentage of CO_2 captured with respect to the CO_2 contained in the exhaust gases, is instead constant and equal to 90% for all cases.

As an alternative, a 2-stage membrane system is also considered. Capture processes based on polymeric membranes are relatively mature [12]. The technology is power-driven as it relies on compressors and vacuum pumps to generate pressure gradients for CO_2 separation. They show competitive performance from moderate CO_2 concentrations (> 10%) [12]. Accordingly, they could become an attractive solution at high EGR rates. The capture scheme is presented in Fig. 4.

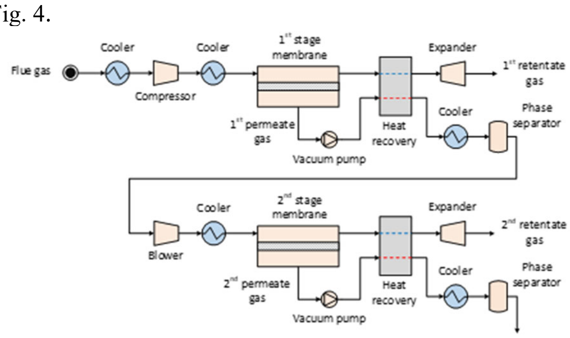

Fig. 4. CO_2 capture scheme based on membrane system.

For the absorption-based processes and at each EGR case, the CO_2 capture section has been simulated with the use of the iCCS tool developed by SINTEF Energy Research [11]. The tool includes first principle calculations (e.g. for pressure drops) and simplified calculations (e.g. regressions) to model each sub-section, like direct contact cooler, absorber, desorber, etc. The tool is based on ASPEN Hysys simulations for a wide range of CO_2 concentrations and flue gas flow rates. For each of these simulations, the L/G ratio was varied between suitable ranges to identify the optimal capture energy requirement. As ASPEN Hysys is a commercial software for process simulation with widely verified and validated correlations, the results are validated. The process simulations allow calculating the capture energy requirement, defined as the Specific Reboiler Duty (SRD) on the desorber, which is the reboiler duty divided by the CO_2 captured. The main equipment diameters (DCC, Absorber and Desorber) are also calculated.

For the membrane-based process and at each EGR case, the CO_2 capture section has been simulated with an in-house modelling framework for the design and optimization of post-combustion capture units [12]. The membrane properties are tuned to reflect the reported performance of an existing polymeric membrane, i.e., MTR Polaris [13]. The design is optimized to minimize the power requirements, considering as variables the pressure levels of the two membrane stages. In addition, an internal recycle scheme is implemented where the retentate gas from the second stage is recycled at the inlet of the first stage. The recovery rate of the second membrane stage is also subject to optimization.

3 Results and discussion

3.1 Combustion study

The results of the experimental campaign were used to identify key operating points to simulate for the entire power plant. Initially, the baseline design was tested without the addition of hydrogen, in order to establish the starting point of the study.

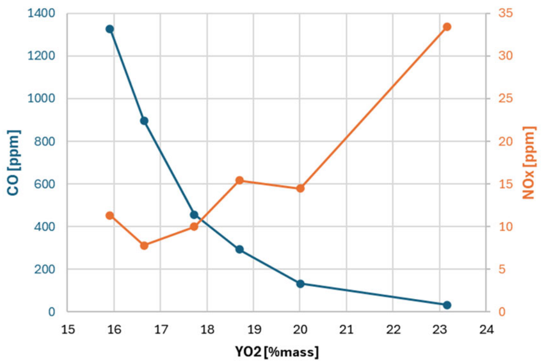

Fig. 5 Effect of lower inlet oxygen level caused by EGR conditions on CO and NOx emissions

Fig. 5 shows the CO and NOx emissions as the inlet oxygen level decreases, with a fixed fuel split and the burner fully fed with natural gas. CO emissions increase exponentially as the inlet oxygen level decreases, i.e., as the EGR rate increases, since the oxidation reaction is slowed down by the lower reactivity. It is observed during experiments that the reaction zone lifts and elongates significantly when lowering the inlet oxygen content, becoming widespread [2]. This shift towards a distributed combustion regime helps to reduce NOx emissions, which decrease with higher EGR levels, only to increase again at very low oxygen levels. For this reason, the oxygen level of 16.6% was identified as optimal, considering the trade-off between increasing CO and decreasing NOx, and also because it is close to the minimum oxygen level that the flame can maintain under stable conditions, which is slightly over YO_2=15%, depending on the burner configuration and fuel split.

The effect of adding hydrogen to the pilot line was tested to evaluate the impact of the enhanced hydrogen reactivity in reducing CO emissions with high EGR rates. Fig. 6 shows CO and NOx emissions as the hydrogen fraction increases, with constant inlet oxygen level. Positive effects are observed, with CO levels decreasing as the hydrogen fraction rises. The benefit is already significant at 5% of the thermal power, while increasing the hydrogen content further has a limited effect, with only a very small gain. Although part of the reduction is due to the lower carbon content in the fuel, the hydrogen mass fraction remains low and this effect remains marginal. Interestingly, even NOx emission drops increasing the hydrogen content. This is due to the lifted configuration of the pilot jets that enhances the mixing among reactants favoring a reduction of NOx [14]. Increasing the hydrogen content, the higher and higher mean velocity of the jets favors such reduction mechanism.

From the visualization of the reaction zone, it has been observed that hydrogen addition allows the flame to stabilize closer to the burner outlet, improving flame stability and reducing high CO levels due to EGR conditions [7]. Hydrogen's effects vary depending on whether it is injected as pure jets or blended with natural gas. Pure hydrogen jets offer significant benefits, limiting the hydrogen amount and maximizing the outcomes, opening the possibility for new, improved burner designs to operate under these unconventional conditions.

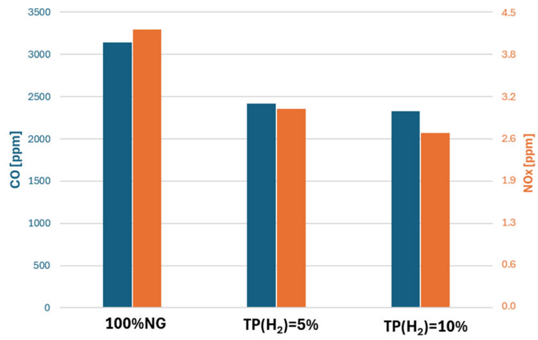

Fig. 6. Effect of hydrogen amount on CO and NOx emissions with constant total thermal power

The results of the experimental study allowed for the identification of some geometric solutions that improve performance, and the selection of relevant operating conditions. Although the results were obtained at ambient pressure, they still provide useful insights for determining optimal operating conditions. Dynamic behavior and emission levels differ when operating under engine-like conditions, and for this reason, pressurized tests on the single sector are planned for the next experimental campaign.

3.2 Process study

The power plant process with CO_2 capture was simulated to identify the main impacts of the EGR and H_2 piloting, focusing on energy requirements, power demand, and equipment sizes, which are crucial for operating and capital costs. Fig. 7 shows the molar concentrations of the CO_2 in the exhaust gases, of the O_2 at the combustor outlet and of the O_2 at the combustor inlet, as a function of the EGR rate and of the H_2 piloting. As expected, results confirm that EGR causes an increase in the CO_2 concentration in exhaust gases and a decrease in the O_2 concentration for the combustion. It is interesting to notice that these trends are not impacted by the addition of hydrogen, as the flowrate of hydrogen is relatively small. Fig. 7 also shows that the EGR rate and the CO_2 concentration are directly proportional, so they can be used in an equivalent way in the current analysis. Based on this, results regarding the overall power plant are shown as a function of the EGR rate (Fig. 7 and Fig. 10. Compression power, gas turbine power and GT net power vs. EGR rate and hydrogen piloting.). The results regarding the CO_2 capture are instead shown as a function of the CO_2 concentration in the exhaust gases, as it is the main variable for the CO_2 capture section (Fig. 8 and Fig. 9). The reference results (Fig. 7 to Fig. 10) consider the MEA-based capture technology.

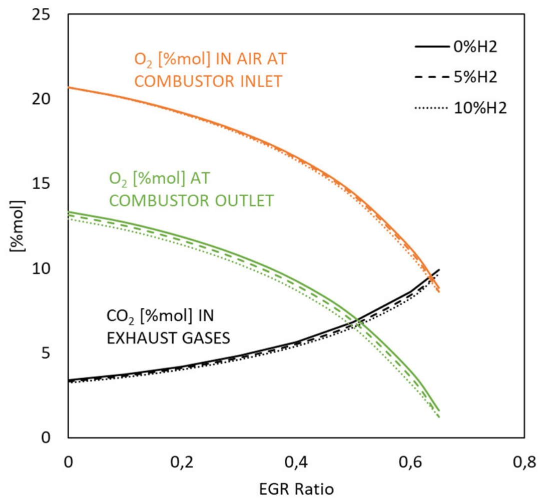

Fig. 7. GT exhaust CO_2 and O_2 molar concentrations vs EGR rate and hydrogen piloting

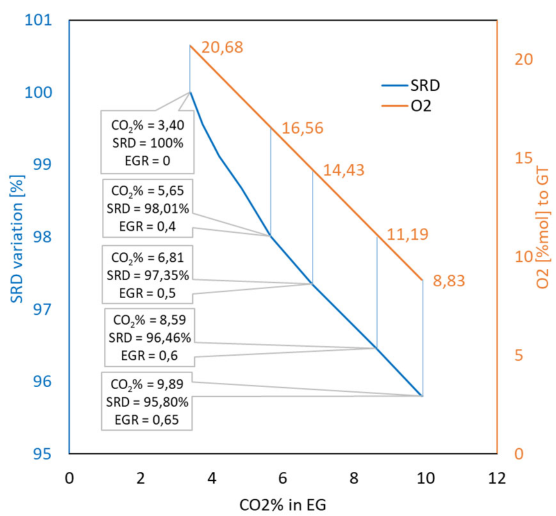

Fig. 8. Specific Reboiler Duty and O_2 molar concentration to the gas turbine vs. the CO_2 concentration in the GT exhaust gases.

The increase of the CO_2 concentration is translated to an energetic benefit for the CO_2 capture section, as represented by the simulation results in Fig. 10. The SRD, the main energy requirement in the system, consistently decreases as the CO_2 concentration increases. As the O_2 concentration limits the EGR rate, it is also represented in the figure to easily show how much the SRD can be decreased before impairing the combustion quality. To maintain combustion stability and avoid operating issues in the combustor, the optimal O_2 concentration is experimental found to be 16.6 % (cf. §3.1), which is in good agreement with previous reported studies around 16 % [2,3]. According to our results, this value corresponds to a maximum EGR rate of 40%, to a CO_2 concentration of 5,65%mol and a reduction of the SRD of 2%. This figure shows explicitly how hydrogen piloting can be interesting to push these limitations towards the right side of the graphics, that is, towards a lower energy demand for the CO_2 capture.

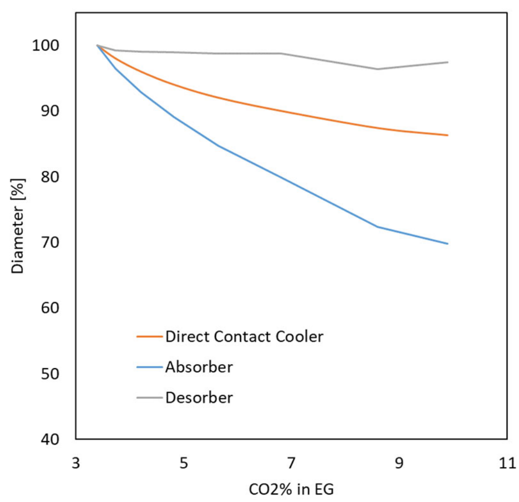

Fig. 9. CO_2 capture main equipment diameters vs. CO_2 concentration in the GT exhaust gases.

The decrease in energy demand is not the only benefit of implementing EGR, as it also determines a significant decrease in the volumetric flowrate of the exhaust gases to be treated in the CO_2 capture, with a significant decrease in the equipment sizes, that is a relevant factor for the capital costs. Fig. 9 shows the main equipment sizes of the MEA-based CO_2 capture unit as a function of the CO_2 concentration in the EG. As the Direct Contact Cooler and the Absorber column process directly the exhaust gases, their size is strongly impacted. The desorber is impacted less as it processes only the solvent and not the exhausts, even if there is still a slight benefit on its dimensions.

Fig. 10 illustrates the effect of EGR, showing how the compression power in the gas turbine, the turbine power, and the net power all vary with the EGR rate. As observed, both the compression power and turbine power increase with higher EGR. This behavior can be mainly attributed to the change in fluid composition (i.e., higher CO_2 content), in turn resulting in a larger mass flow rate swallowed by the compressor and in a decrease of the specific heat ratio γ, as described in Equation (2):

$$P_K = \frac{1}{\eta_{is}} \frac{\gamma}{\gamma - 1} RT \left[\left(\frac{P_{out}}{P_{in}} \right)^{(\gamma-1)/\gamma} - 1 \right] \dot{n}_{in}$$

Where γ is the gas specific ratio, defined as the ratio of the specific heat of the gas at a constant pressure to its specific heat at a constant volume, η_{is} is the isentropic efficiency, in this work assumed equal to

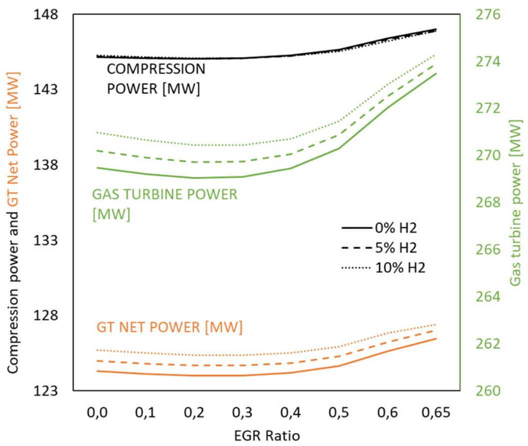

Fig. 10. Compression power, gas turbine power and GT net power vs. EGR rate and hydrogen piloting.

0.85, $\dot{n}_{in}$ is the molar flowrate of the gas

The expansion power is also affected by a larger mass flow rate and by the decrease of γ, following the same equation but with an opposite (negative) sign. Indeed, as the CO_2 concentration increases, the γ of the air entering the compressor slightly decreases—from 1.403 at 0% EGR to 1.392 at 65% EGR—leading to a rise in the absolute values of both compression and expansion power. Additionally, the compression power remains unaffected by the H_2 pilot injection, since hydrogen is introduced directly into the combustor.

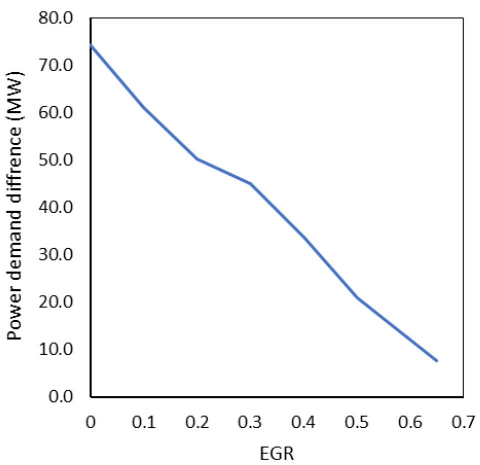

Fig. 11. Power demand difference between membrane and MEA absorption technology vs. EGR rate.

As an alternative CO_2 capture solution, a membrane technology has been tested. Fig. 11 shows the difference in power demand between the membrane and MEA absorption solutions, as a function of EGR rate. To enable a fair comparison, the heat requirement from the MEA process has been converted into power by means of a conversion factor, which relate steam extraction with reduced expansion power in the steam turbine [15]. In accordance with existing knowledge, the membrane process shows poor performances at low EGR rates, i.e., low CO_2 inlet concentrations. At the same time the performance rapidly improves as the EGR rate increases, gradually diminishing the gap against the absorption processes. However, at the highest EGR rates, the difference is still in favour of solvent absorption. Arguably, the results indicate that membrane will hardly be competitive for CO_2 capture from the flue gas a NGCC.

4 Conclusions

The presented study explored how CO_2 capture implementation in a Natural Gas Combined Cycle can be improved by combining EGR and H_2 piloting to maximize CO_2 concentration in the exhaust gases to be treated. The system has been analyzed specifically for a large size gas turbine both from a combustion perspective and integrated process level.

The results can be summarized as follows:

- 5% power equivalent of H_2 piloting allows to extend the operating EGR rates, which is otherwise limited to 40%.
- Further increase in H_2 pilot does not impact significantly the performance.
- CO_2 capture is positively impacted by EGR with a reduction in energy requirement up to approximately 3% and in equipment sized up to approximately 20%.
- From the CO_2 capture point of view, the optimal EGR ratio is the highest achievable before combustion limitations arise.
  - Gas turbine net power is also positively impacted by EGR, as well as that of the overall combined cycle, mainly due to the reduced steam extraction.

The on-going TRANSITION project will further progress the technology by improving the combustion performance through high pressure testing, advanced numerical simulation, and a techno-economic analysis of the whole power plants.

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