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PII: S1875-5100(17)30445-6

DOI: [10.1016/j.jngse.2017.09.012](https://doi.org/10.1016/j.jngse.2017.09.012)

Reference: JNGSE 2356

To appear in: *Journal of Natural Gas Science and Engineering*

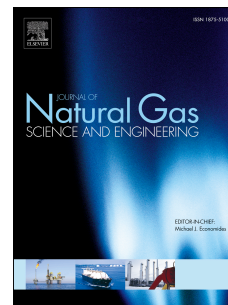
Received Date: 4 April 2017

Revised Date: 13 September 2017

Accepted Date: 26 September 2017

Please cite this article as: Darabkhani, H.G., Jurado, N., Prpich, G., Oakey, J.E., Wagland, S.T., Anthony, E.J., Design, process simulation and construction of a 100 kW pilot-scale CO₂ membrane rig: Improving in situ CO₂ capture using selective exhaust gas recirculation (S-EGR), *Journal of Natural Gas Science & Engineering* (2017), doi: 10.1016/j.jngse.2017.09.012.

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Design, Process Simulation and Construction of a 100 kW Pilot-scale CO₂ Membrane Rig: Improving in situ CO₂ Capture Using Selective Exhaust Gas Recirculation (S-EGR)

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Abstract

Carbon capture and storage (CCS) from natural gas fired systems is an emerging field and many of the concepts and underlying scientific principles are still being developed. Preliminary studies suggest this approach can boost the CO₂ content in the feed gas up to 3 times compared to the 'no recycle' case (CO₂ concentration increased to 18% vs. 6%), with a consequent reduction in flow to the post-combustion capture unit by a factor of three compared to conventional, non S-EGR. For this project, Cranfield developed a pilot-scale 100 kW CO₂ membrane rig facility in order to investigate simultaneously EGR and S-EGR technologies, the latter being achieved by using a CO₂ sweep air polymeric membrane. A bench-scale membrane rig has also been developed to investigate the permeability and selectivity of different polymeric membranes to CO₂. Currently a small-scale polydimethylsiloxane (PDMS) membrane module is also being investigated to study its selectivity/permeability. The tests include exploring the performance improvement of the PDMS membrane using different operating conditions with a view to developing scale-up procedures for the membrane unit for the actual 100 kW pilot-scale rig.

Process simulations were performed using Aspen Plus software to predict behaviour of the pilot-scale rig using a model developed based on empirical parameters (i.e., mass transfer coefficient of CO₂ through the membrane and permeance), measured in the bench-scale membrane test unit. The results show that CO₂ concentrations of up to 14.9% (comparable to CO₂ level in coal combustion) can be achieved with 60% EGR, with a 90% CO₂ removal efficiency of the membrane units. However, the results generated with the membrane model in which specific permeance values to PDMS were applied, predicted concentrations of CO₂ in flue gases up to 9.8% (v/v) for a selective recycle of 60%. The study shows that the S-EGR technique is an effective method that can provide similar conditions to that of a coal-fired power plant for the post-combustion capture system operating on natural gas-fired units, but also highlights the fact that more research is required to find more suitable materials for membranes that optimise the CO₂ removal efficiencies from the flue gas.

Keywords: Carbon dioxide capture; Gas-CCS; Exhaust gas recirculation (EGR); CO₂ selective membrane, Selective EGR (S-EGR)

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1- Introduction

Recent EU regulations, such as the Large Combustion Plant Directive (Council of the European Union, 2001) and the Industrial Emissions Directive (Council of the European Union, 2010) have reduced the number of thermal power plants in operation in the EU (Bonjean Stanton et al., 2016). However, pressure from the EU regulation to promote a greater share of renewable energy in the energy mix (Council of the European Union, 2009) has led to demand for secure and flexible supplies of electricity that can complement renewable energy's intermittency. The uptake of renewable energy has changed the way conventional plants operate (Eser et al., 2016). Though projections for natural gas usage in the EU vary ((Böhringer et al., 2017; Hills and Michalena, 2017; Smith, 2013)), it remains a vital contributor to the EU energy mix. Challenges facing the natural industry include the renewable energy targets, the highly variable cost of natural gas, and increasingly stringent emissions targets. As part of the drive towards a more sustainable energy portfolio there remains a pressing need to capture the CO₂ emitted by gas-fired plants to meet EU emission targets (Hills and Michalena, 2017).

To date, carbon capture research has been largely focused on coal-fired power plants and only a few studies are available that describe carbon capture from gas turbines (Belaissaoui et al., 2014, 2013). In addition, the research has been dominated by solvent technologies, like amine-based solvents (Arshad et al., 2016; Chavez and Guadarrama, 2016; Liang et al., 2015; Xiao et al., 2017).

Membrane separation has shown promise as a means of selectively removing CO₂ from post-combustion gas streams due to its lower energy demand, technological maturity, and relatively straightforward installation compared to equilibrium systems (Pires et al., 2011). In the context of power generation, membranes are used to separate CO₂ from flue gas to create high-concentration CO₂ streams that can be sent to secondary capture units or recirculated (Carapellucci et al., 2015). The ability of a gas to transfer across a membrane is limited by membrane selectivity (for CO₂) and its permeability. Considerable research into the development of new membrane materials continues (Fu et al., 2016; Karimi et al., 2017; Kim and Lee, 2015, 2013; Yan et al., 2015). However, pilot-scale membrane studies are often restricted to commercially available membrane products that have fixed selectivity and permeability. Future developments for gas separation at larger scale will rely upon improvements to operational parameters, specifically control of the difference in partial pressure across the membrane. This can be adjusted by increasing the feed-side pressure via compression, increasing the feed-side concentration via exhaust gas recirculation (EGR), decreasing the permeate-side pressure via vacuum, or a combination thereof.

Much of the knowledge being generated is via simulation, such as the study by Carapellucci et al., 2015. Numerous studies have been conducted showing that carbon separation using membrane

systems in natural gas-fired turbines is feasible (in terms of cost and energy penalties) when compared to amine-based systems (Ramírez-Santos et al., 2017; Zhai and Rubin, 2013). In general, there are few studies that can provide empirical evidence to support and validate these simulations (Yuan et al., 2016). Moreover, there is a lack of fundamental knowledge about the practicalities of operating membrane systems at scale (Adewole et al., 2013; Dalane et al., 2017; Kang et al., 2017). For example, experimentation is needed to study the effects of dynamic gas mixtures and moisture content on membrane aging and performance.

The concentration of CO₂ in flue gases has direct effects on energy consumption in post-combustion capture (PCC) and CO₂ compression systems (Ali et al., 2016; Li and Ditaranto, 2012; Tola and Finkenrath, 2015). From the chemical absorption point of view, it has been proven that the reboiler heat requirement in solvent regeneration is sensitive to the CO₂ levels in the flue gas (Li and Ditaranto, 2012). As reboiler duty is the most energy-consuming part of the chemical absorption capture system, application of EGR and S-EGR (selective EGR using a CO₂ selective membrane in the recirculation loop) could noticeably cut the reboiler duty (Canepa et al., 2012; Carapellucci et al., 2015; Li and Ditaranto, 2012). With EGR/S-EGR, the exhaust gas is partially substituting for air in the combustion chamber and consequently, CO₂ concentration in the flue gas increases as the EGR ratio increases. However, stable operation on a DLN F-class turbine has been reported with a 35% EGR ratio (17% O₂ inlet concentration) producing flue gas with 10% CO₂ (ElKady et al., 2009). However, simulations run at 40% and 30% EGR ratio, respectively, reported oxygen concentration in the combustion air to be 16% O₂, and 6-8% CO₂ in exhaust (Canepa et al., 2012; Johnshagen, Klas; Sipocz, 2010; Li and Ditaranto, 2012).

2- Experimental Facility

A 100 kW rig was designed, constructed and commissioned to study the effects of exhaust gas recirculation and selective recirculation (using membranes) on the performance of natural gas-fired combustion with carbon capture capability. A process flow diagram for the systems is shown in Fig. 1. This system is based on the concept that by recirculating a CO₂-rich permeate stream into the feed gas of the burner, the CO₂ concentration in the exhaust stream would increase compared to non-recirculated systems (Canepa et al., 2012; Johnshagen, Klas; Sipocz, 2010; Li et al., 2011) and that the addition of a membrane separation unit could increase the CO₂ concentrations even further. Simulations have shown that the CO₂ concentration could be increased from 6% to 18% using the membrane system, which would reduce the flow of gas to the post-combustion capture unit by a factor of three compared to the non-S-EGR system. The system was designed to operate in various modes (e.g., recirculation, recirculation + membrane, no recirculation), and to accommodate the direct injection of gases (e.g., CO₂, O₂, steam) to simulate different carbon capture conditions (e.g., oxyfuel, recirculation). It was also designed to accommodate the use of different membrane separation

materials and, potentially, other carbon capture techniques. To our knowledge, this is the only example of a 100 kW gas burner equipped with a membrane separation unit.

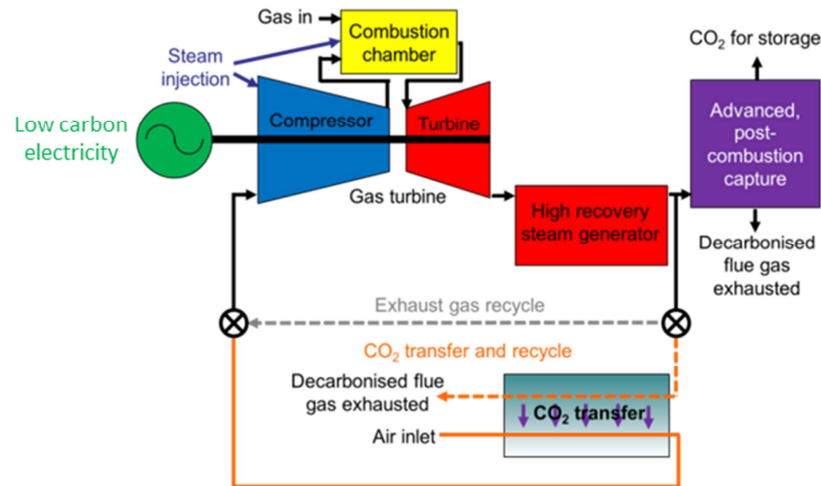


Fig. 1. Flow diagram of EGR and S-EGR (EGR with membrane) in the Gas-FACTS project

The CO₂ membrane combustion system operates at atmospheric pressure and uses a 100 kW MP4 Nu-Way burner, which can achieve a high degree of stability over a wide turndown range for relatively low service inlet pressures, allowing the rig to operate over a range of power outputs (e.g. 20-100 kW). City natural gas was fed to the system using mains pressure and controlled by a mass flow meter, and combusted with ambient air supplied by a centrifugal fan at a feed rate of up to 360 m³/h at a gauge pressure of 3.4 kPa. Combustion gas mixture was controlled via an electronic gas valve that maintained stoichiometric ratios. This design configuration enables flexibility in the turndown range of the gas burner, and the composition of the combustion gas mixture, enables tests to be performed under different recirculation conditions (i.e., variable CO₂ levels). The diagram of the process is presented in Fig. 2.

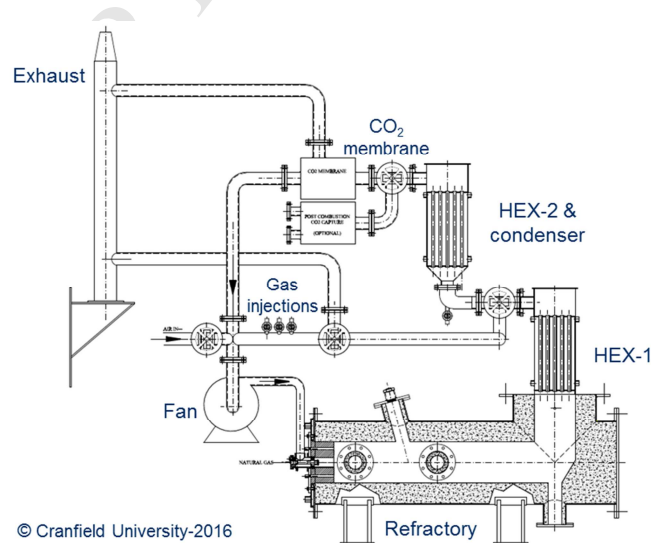


Fig. 2. Process diagram of 100 kW CO₂ selective membrane natural gas-fired combustion rig (HEX: heat exchanger)

Combustion parameters for standard operating conditions of the natural gas-fired burner are provided in Table 1.

Table 1: Combustion parameters of a 100 kW natural gas-fired burner

Combustion Parameter	Value	Units
Natural Gas Flow rate	9.30	m ³ /h
Caloric Value (CV)	39.40	MJ/m ³
Air/Fuel Ratio	9.70	by volume
Air flow rate	90.21	m ³ /h
Inlet Temperature	15	°C
Combustor Efficiency	99	%
Heating Power	360.92	MJ/h (100.26 kW)
Fuel Density	0.712	kg/m ³ @ 15°C
Air Density	1.205	kg/m ³ @ 15°C

Combusted flue gases flow through a refractory cylinder of 2.2 m length and an inner diameter of 0.6 m. The combustion chamber has three viewing ports that provide optical access to the flame. Flame stability and temperature can be measured by processing images of the flame captured via a high-speed camera.

The air/fuel ratio conventionally varies from 60:1 to 120:1 for simple cycle gas turbines in aircraft (Saravanamuttoo et al., 2001). To control NO_x emissions, combustion in a gas turbine system normally occurs under ultra-lean premixed conditions with air-fuel ratios up to 200:1 (IIT Kanpur, n.d.). With recirculation of the flue gas only, the concentration of CO₂ in the natural gas combined cycle (NGCC) flue gases would be expected to increase from 3–4% (Wei et al., 2011) to an upper level of approximately 6-7% (Merkel et al., 2013). The stoichiometric air-fuel ratio for natural gas combustion is 17.3:1 by mass (9.59:1 by volume) (Porpatham et al., 2008). Operation in S-EGR mode is expected to increase CO₂ levels in the gas burner feed even higher (up to 18.6%), which should limit the dilution of O₂ in the combustion air by reduction of N₂ in the recirculation stream (Wei et al., 2011). Gas feed ports were installed on the recirculation loop and these can be used to permit the injection of supplementary gases (e.g., CO₂, O₂) should it be required to simulate different S-EGR conditions or to accommodate future partial oxy-firing mode.

To operate under S-EGR conditions, the temperature of the flue gases must be controlled and this is achieved using two heat exchangers. The first heat exchanger has an overall size of 1282 mm x φ168 mm and is used to reduce the temperature of the flue gas from 700°C (max) to 300°C. After the heat exchanger, flue gases are directed to either recirculation, the membrane (S-EGR), or to exhaust via a splitter. Gases that are directed to the S-EGR need to be cooled further, to between 60 and 80°C, before entering the membrane unit to avoid degradation of the membrane (exceeding the maximum allowable temperature for the silicon fibres). The second heat exchanger is 1192 mm x φ114 mm. Both heat

exchangers use coolant water, operate in a counter-current flow, and have a maximum working water pressure of 4 bar, and a maximum working temperature of 110°C. The maximum working exhaust gas pressure is 0.5 bar with a maximum working exhaust gas temperature of 700°C.

Flue gases can be recycled via two different routes. The first route is a simple recirculation, whereby a fraction of the flue gas is returned to the air feed of the gas turbine. The second route is recirculation via the CO₂ selective membrane whereby a fraction of the permeate stream from the membrane, which has a high CO₂ content, can be recycled back to the air feed of the gas turbine. To recycle the flue gases through the membrane units and push the sweep air through the membrane, two small brushless fans have been installed with a maximum flow rate of 147.9 m³/h and sealed pressure of 421 mbar.

The S-EGR system uses a commercially available, polydimethylsiloxane (PDMS) organic polymer membrane that was purchased from PermSelect Ltd (USA). PDMS is also referred to as silicone, which is among the most gas permeable dense polymeric membrane materials available (PermSelect 2016). Expected permeability coefficients for common gas species are given in Table 2. To size the membrane an Aspen Plus simulation was performed (see Section 3.0) that used empirical data from a small-scale experimental setup to estimate a global mass transfer coefficient for CO₂. Based on this analysis, two 35 m² membrane units were manufactured to provide a total of 70 m² of PDMS membrane surface area. The wall thickness of the silicon fibres was 20 µm and this was chosen to provide the greatest permeability. Pressure drop across the membrane was estimated to be near 0.1 MPa at a flow rate of 100 dm³/min.

Table 2: PDMS permeability coefficient for different gases [PermSelect 2016 and Robb, 2006]

Gas Species	Permeability Coefficient (Barrer)* for PDMS
Nitrogen	280
Carbon monoxide	340
Oxygen	600
Nitric oxide	600
Methane	950
Carbon dioxide	3250
Nitrous oxide	4350
Nitrogen dioxide	7500
Sulphur dioxide	15000
Water	36000

*1 Barrer = 10⁻¹⁰ cm³ (STP) · cm / cm² · s · cm-Hg

Fresh air was used as the sweep gas and was provided via recirculating fan. The two membrane units could be configured to operate in co-current or counter-current feed/sweep gas modes. A vacuum pump was attached to the shell side of the membrane to enhance the partial pressure gradient across the membrane. The two membrane units also enable operation under various arrangements, e.g., in series or parallel, and additional membrane units can be added to increase surface area. The process and instrumentation (P&I) diagram for the system is presented in Fig. 3.

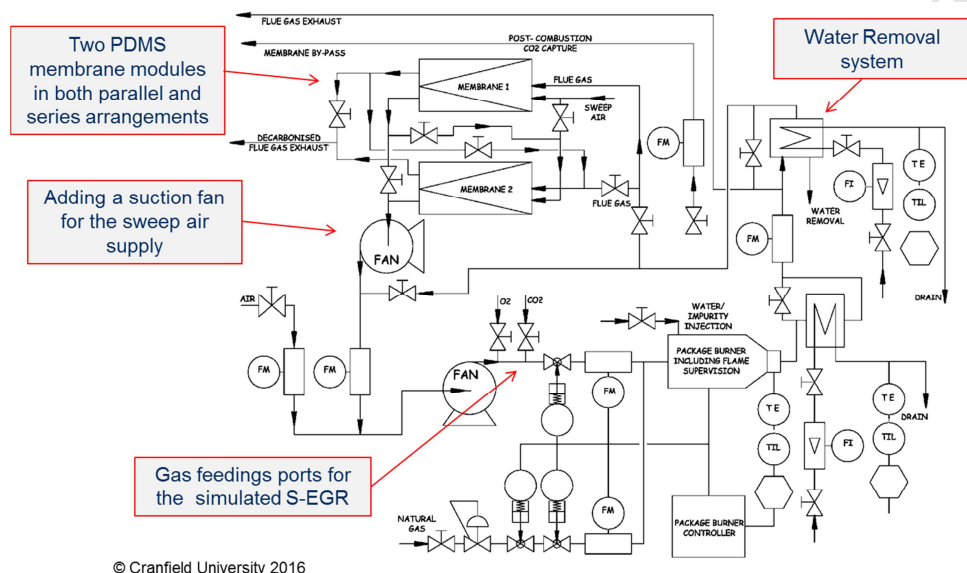


Fig. 3. P&I diagram of 100 kW CO₂ selective membrane natural gas-fired combustion rig facility

3. Modelling procedure

Process simulation, using ASPEN Plus, was used to calculate the mass and energy balances of the system for equipment selection and design. A visual description of the simulation is shown in Fig. 4. Combustion of the natural gas is performed with air using an RStoic, which is based on known fractional conversions or extent of reactions. The generated flue gas stream passes through a heat exchanger that cools the gas temperature down to 300°C. Exiting gas enters the splitter and the fraction of flue gas that goes to the membrane treatment is set. The remainder of the gas is diverted to secondary capture. Gas exiting the splitter enters a second heat exchanger where the gas temperature is reduced to 60°C to meet the requirement of the membrane material. This temperature reduction also enables the condensation of the water vapour contained in the flue gas, which can impact membrane performance. The permeate stream exiting the membrane is CO₂-rich and is recycled and re-supplied to the combustor. The CO₂-depleted stream is then discarded by venting to the atmosphere.

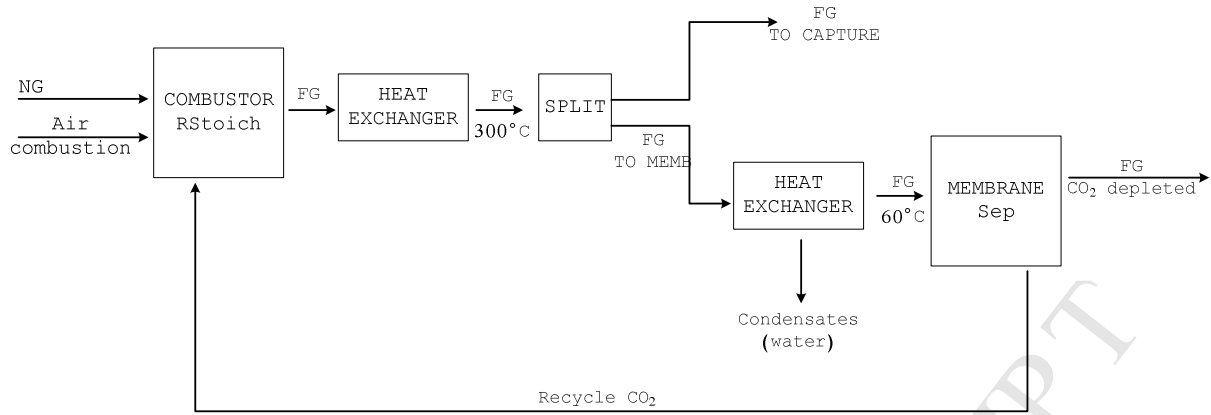


Fig. 4. Schematic of the pilot-scale simulation model

For the CO₂ capture unit, during the first stage of the model development, the membrane was assigned an Aspen Separator block. For the second phase of the model, the Separator block was replaced with a User Model block. This User Model block communicated with an Excel file, where a sub-model to simulate the mass transfer occurred inside the membrane was run, was used to account for differences in membrane performance given changes in CO₂ permeability and selectivity. Fig. 5 shows the interface of the simulation model developed using Aspen Plus and Excel.

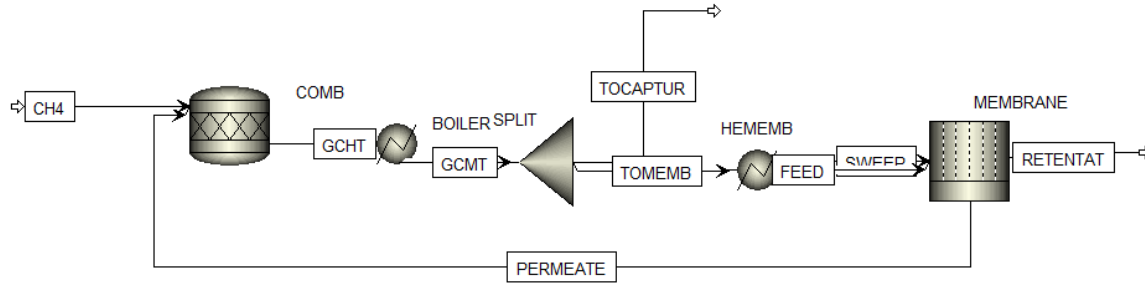


Fig. 5. Aspen Plus interface for membrane model

The User Model calculated the flux of CO₂ transferred across the membrane from the flue gas to the sweep air stream. This relationship is described by Fick's laws of diffusion at steady state conditions. Developing Fick's first law and taking into consideration the suggestions given by Tremblay et al., 2006 (Tremblay et al., 2006), of introducing boundary conditions for a planar sheet, and replacing concentrations with gas partial vapour pressures (for a gas system), we obtain equation 1:

$$J = P \frac{p_0 - p_1}{l} \quad (2)$$

where P is the permeability coefficient, p_0 and p_1 are the gas partial vapour pressures on either side of the membrane wall, and l is the membrane thickness. Specifying this expression to the diffusion of CO₂ through the membrane, we can write equation 2.

$$J_{CO_2} dA = \frac{P_{CO_2}^*}{l} \cdot [xP_f - yP_p] dA \quad (2)$$

To complete the User Model the global mass transfer coefficient (K_L) for the membrane, as well as the permeance ($P^*_{CO_2}/l$) of the membrane, needed to be determined. The permeance is defined as the permeability divided by the membrane wall thickness (P/l) (Baker, n.d.; Baker et al., 2010). Equations 3 and 4 show the parameters responsible for calculating the mass transfer of CO_2 through the wall of the membrane (Heile et al., 2014).

$$A_{\text{membrane}} = \frac{Q_{\text{Sweep air}}}{K_L} \cdot \frac{Y_{\text{Sweep air,CO}_2}|_{\text{final}} - Y_{\text{Sweep air,CO}_2}|_{\text{initial}}}{\Delta Y_{\log \text{ mean}}} \quad (3)$$

$$\Delta Y_{\log \text{ mean}} = \frac{\Delta Y_{CO_2}|_B - \Delta Y_{CO_2}|_A}{\ln \left[\frac{\Delta Y_{CO_2}|_B}{\Delta Y_{CO_2}|_A} \right]} \quad (4)$$

A series of experiments using a small-scale PDMS membrane system were conducted to empirically estimate the global mass transfer coefficient for CO_2 across the membrane, as well as the permeance of the PDMS membrane. Tests were conducted using a 1 m² PDMS membrane (PermSelect, USA) in a continuous cross-flow setup. The separation capability of the membrane was determined by feeding a binary gas mixture of CO_2/N_2 into the lumen and by varying y_{CO_2} (5% to 40%). A sweep gas comprised of pure N_2 was supplied to the shell side of the membrane. Gas flow rates into the lumen and shell side were maintained at 10 dm³/min and were measured using a rotameter. A Fourier Transform Infrared Spectrometer (FTIR) was used to analyse gas compositions at all membrane inlets and outlets once the system reached steady state. All experiments were carried out at room temperature (21°C) and atmospheric pressure. Average values for the mass transfer coefficient and permeance of CO_2 through the membrane were calculated to be $2.851 \cdot 10^{-2} \text{ CO}_2/\text{s} \cdot \text{m}^2$ and $2.862 \cdot 10^{-8} \text{ m}^3/(\text{s} \cdot (\text{N}/\text{m}^2) \cdot \text{m}^2)$, respectively.

4. Results and discussion

4.1. Preliminary experimental results: 100 kW CO_2 membrane rig

The experimental conditions explored in this work are shown in Table 3.

Table 3: The operational conditions in a full set of experimental work in the pilot scale membrane rig

Power (kW)	Fuel (m ³ /h)	Fuel (kg/h)	Air (m ³ /h)	Air (kg/h)	RFG %
40	3.71	2.64	35.99	43.37	20, 40, 60, 75
50	4.64	3.30	44.99	54.21	20, 40, 60, 75
70	6.49	4.62	62.99	75.90	20, 40, 65
100	9.28	6.60	89.98	108.42	20, 40, 65

In the initial phases of this work the burner power output was set to 70 kW. The temperature profile results at the inlet and outlet ports of the heat exchangers are presented in Fig. 6. The set temperature

point for the inlet of the flue gases to the first heat exchanger (H.X.1) was 640°C. The commissioning results show that the burner could adjust itself to maintain a temperature of around 540°C and the second heat exchanger drops the temperature down to 54°C achieving a total temperature drop 580°C. In another experiment with adjustment of the water coolant flow rate the flue gas temperature after the second heat exchanger dropped the final temperature down to 30°C. This removes much of the water content from the flue gases in a specially designed water trap system after the second heat exchanger. This set of heat exchangers are essential to ensure the temperature of flue gases is kept well below 80°C which is the upper limit of temperature for the silicon PDMS fibres.

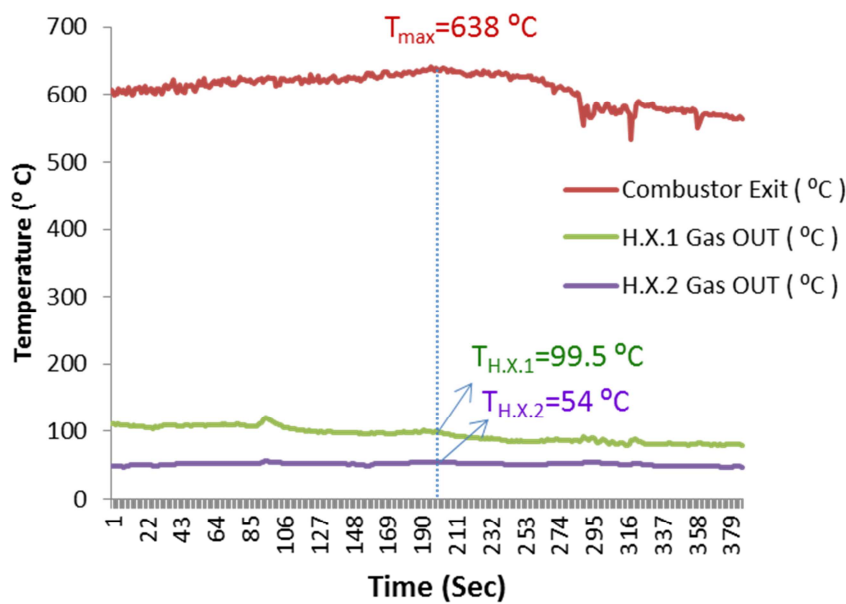


Fig. 6. Flue gas temperature profiles at the inlet and outlet ports of the heat exchangers in the pilot scale 100 kW membrane rig

The major flue gas components in the membrane rig commissioning experiments are shown in Fig. 7. The original volume concentration of CO₂ was about 5%, however, a maximum concentration of 7.3% was achieved during initial tests. This is equivalent to the results for a simulation with 20% EGR when the efficiency of CO₂ removal in the membrane units are as low as 11%. The average concentration of H₂O is about 4% with minimum of 2.5% when a high level of water removal is in place. The NO_x level fluctuated with an acceptable average of 32 ppm.

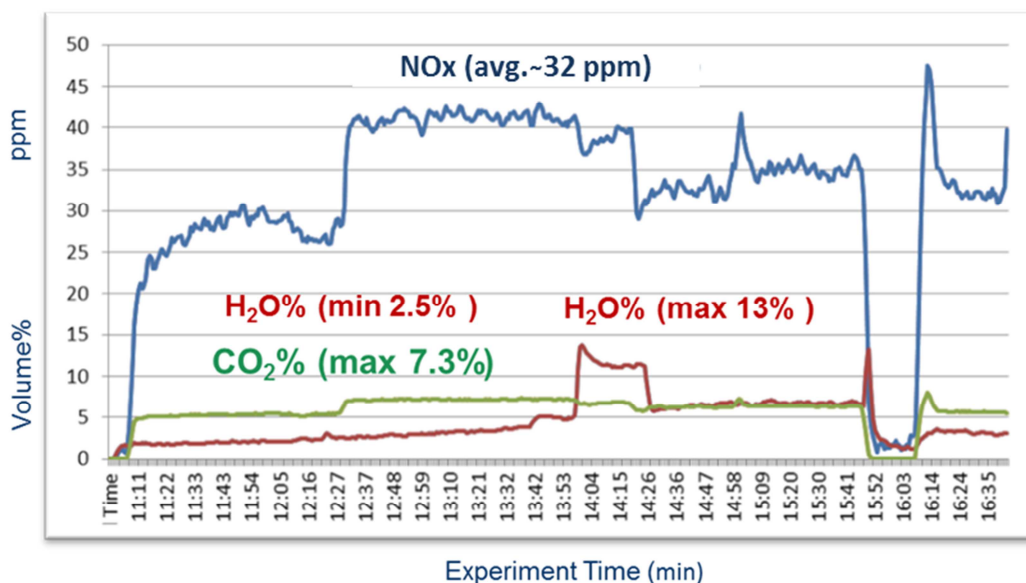


Fig. 7. Major gas compositions in the flue gases in the stoichiometric combustion in the membrane rig during the commissioning experiment

After the successful commissioning stage an experimental campaign to study the effect of selective EGR using one of the membrane units with 35 m² of PDMS polymer surface was undertaken. Experimental trials explored the effect of varying flue gas to membrane flow rate and sweep air flow rate. Both of these variables were manipulated using the fans installed in the corresponding lines. All flow rate measurements taken from the rig, were obtained using orifice plates. In the cases that further sweep air is used for better membrane separation of the CO₂, the concentration values were adjusted in such a way that the total air supply was constant; meaning that if the sweep air was increased, the main air supply fan of the burner (ID fan) was decreased accordingly, this way the sweep air does not add to the air flow rate, but replaces atmospheric air from the ID fan. This was done by multiplying the value of the CO₂ concentration by the ratio of the new total flow to the initial flow.

Effect of Sweep Air Flow Rate

The CO₂ concentration of the flue gas (raw data and corrected values) change as the sweep air flow rate was increases (see Fig. 8 and Fig. 9 for experiments 2 and 4, respectively). A constant flow rate sent to the membrane -this was approximately 66 dm³/min-, which means that around 6.6% of the flow only was sent to the membrane for separation. There are two noticeable trends in these graphs; the sweep air flow rate is directly proportional to the amount of CO₂ in the permeate, and the increase in CO₂ concentration is very low compared to the increase in flow rate through the membrane. To obtain the values available in the corrected graph, a control scheme needs to be introduced into this system, to allow a fixed total of air into the combustion chamber, or allow more gas in to keep constant the air to fuel ratio, and hence isolate the CO₂ concentration and study them closer.

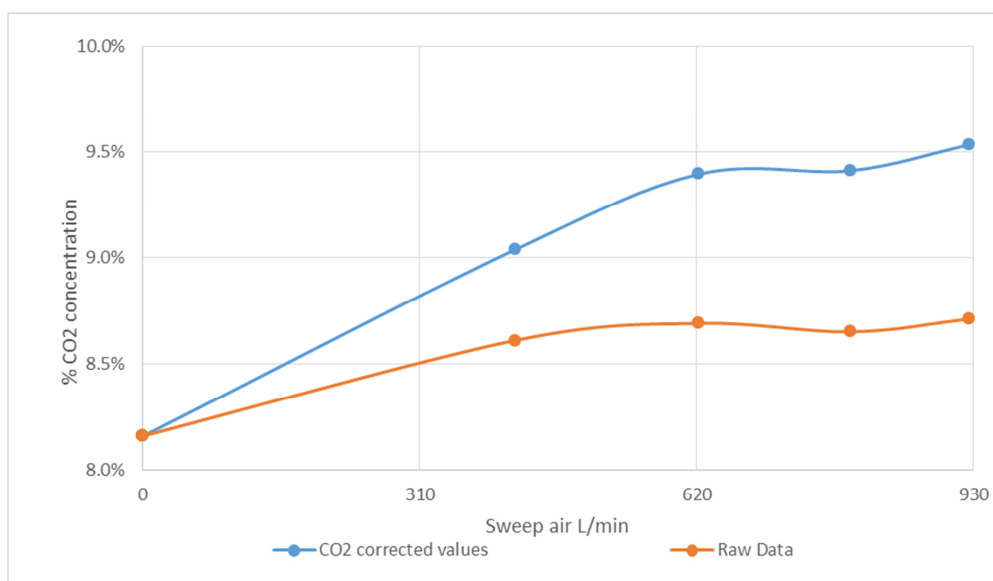


Fig. 8. Sweep Air Flow Rate vs. CO₂ Concentration (Experiment 2)

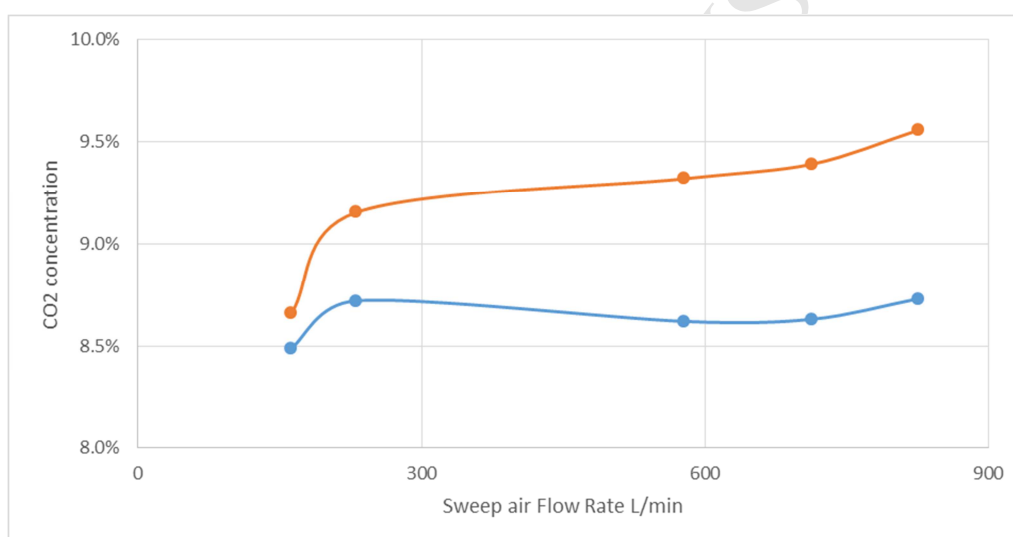


Fig. 9. Sweep Air Flow Rate vs. CO₂ Concentration (Experiment 4)

Effect of Flue Gas to Membrane Flow Rate

During this trial the sweep air flow rate was kept constant and the flue gas to membrane was manipulated. A correction factor was also included to monitor the CO₂ concentration if the air to fuel ratio was kept constant. The experiment was done on a constant sweep air flow rate of 140 dm³/min. It can be noticed that the higher flow rate of gas allowed through the membrane, the higher amount of CO₂ that can be extracted and hence recirculated (see Fig. 10 and Fig. 11).

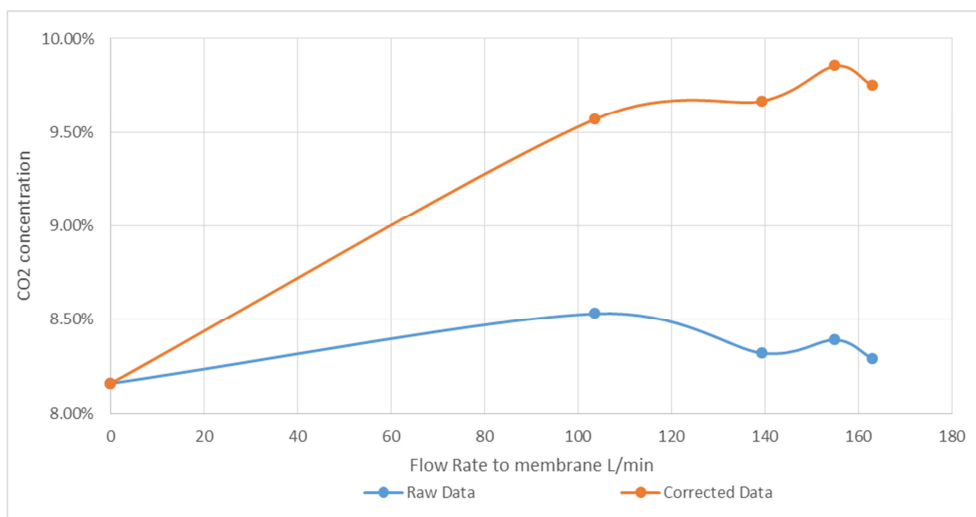


Fig. 10. Flue Gas to Membrane vs. CO₂ concentration (Experiment 2)

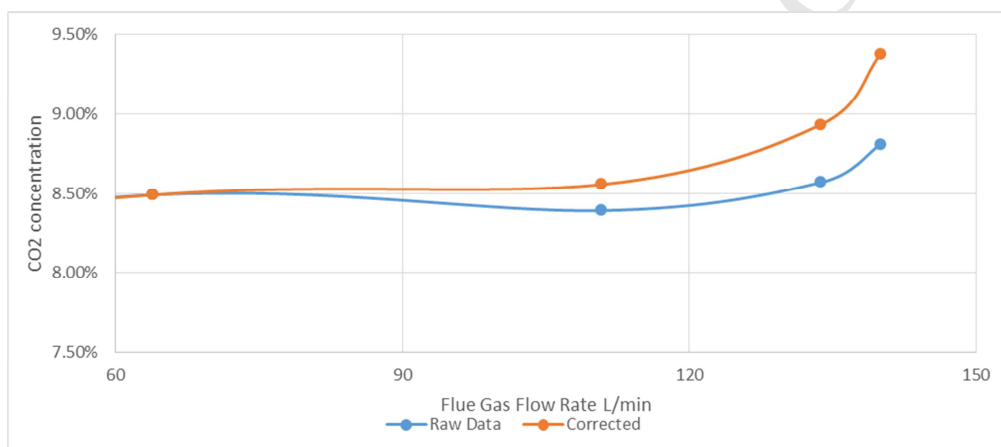


Fig. 11. Flue Gas to Membrane vs. CO₂ concentration (Experiment 4)

Effect of Increase in Both Flow Rates

Since there is a noticeable increase in both cases, when the flow rate in both sides is increased independently, another experiment was conducted to find the maximum concentration that would be possible to reach within the limitation of this setup. This experiment was done by increasing both flows in equal percentage of the maximum flow simultaneously. The below graphs show the details of these experiments (see Fig. 12 and Fig. 13).

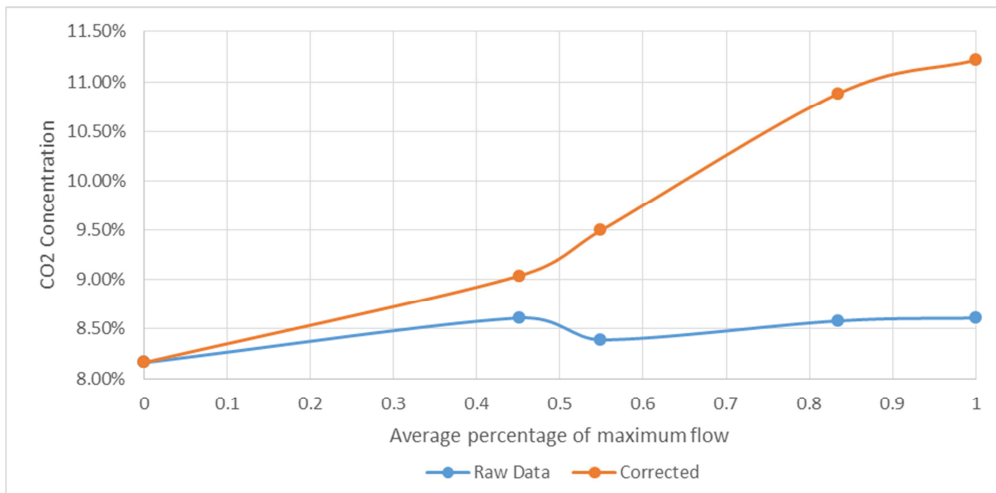


Fig. 12. Average Percentage of maximum Flows vs. CO₂ concentration (Experiment 2)

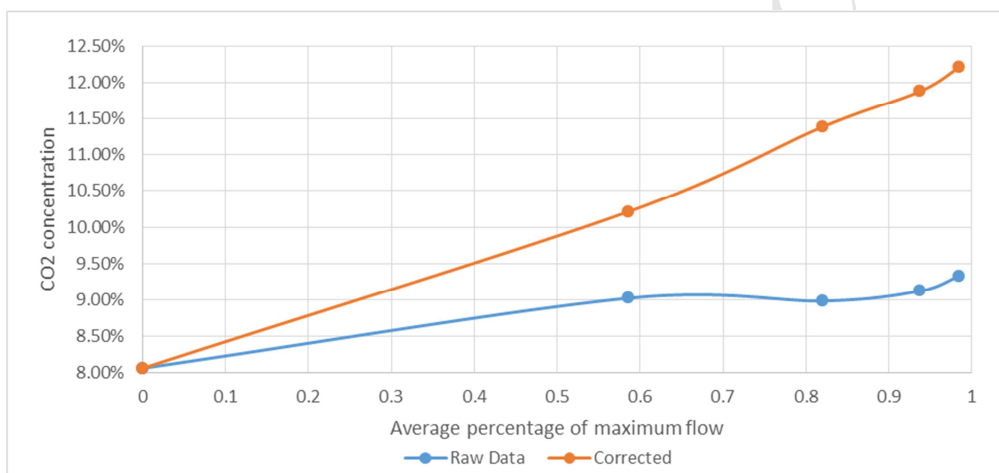


Fig. 13. Average Percentage of maximum Flows vs. CO₂ concentration (Experiment 4)

A similar pattern is observed where adding sweep air and EGR will increase the level of CO₂ concentration. However, through this experiment the maximum CO₂ percentage achieved, was 12%). These represent a 15% increase in case the air to fuel ratio is not fixed, and using the correction the values could increase up to 50% higher. The values are presented in percentage since the initial composition was different during the experiments. Fig. 14 clarifies the operational area of the experiments of the corrected figures. The CO₂ percentages are the area between the two graphs.

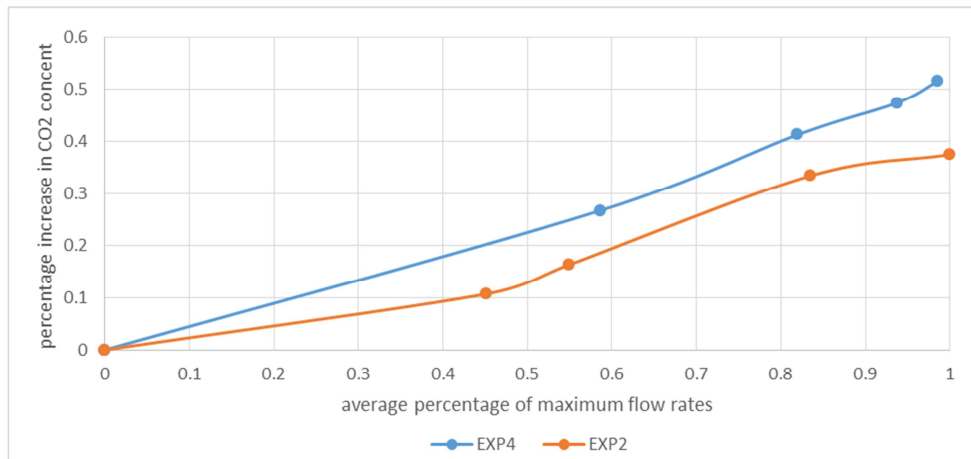


Fig. 14. Operational Area of Corrected Data

From the above graphs, it can be deduced that if the setup is adjusted to allow a constant air to fuel ratio, the percentage increase in CO₂ concentration can vary between 40 to 50%, considering the flow rate limitations in which a maximum of 15% of the exhaust flow only can be sent to the membrane.

Membrane Overall performance

The membrane performance is affected by both the flue gas flow rate, and the sweep air flow rate (See Fig. 15). However, it is clear that the higher the ratio between sweep to flue, the better separation performance achieved.

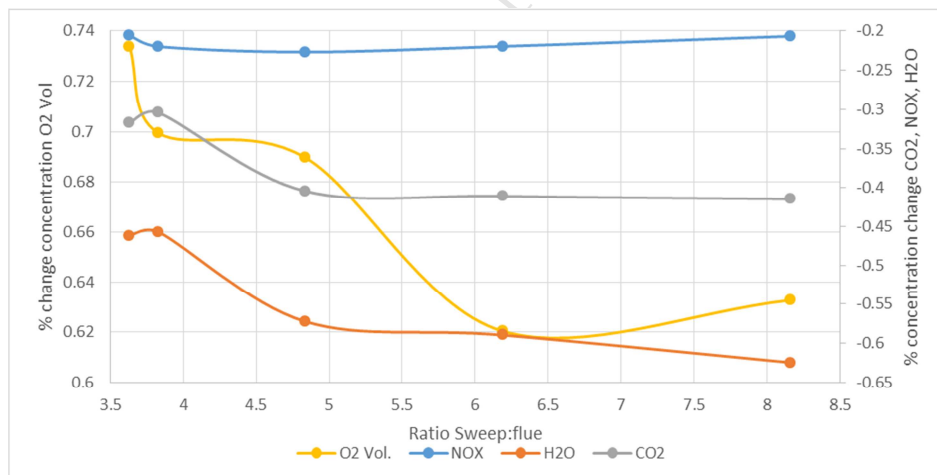


Fig. 15. Effect Sweep to Flue Ratio on composition

A higher sweep air to flue gas to membrane flue ratio causes the membrane to achieve a better separation on the flue gas side. The highest separation achieved for CO₂ was at ratios between 5 and 9, allowing separation to reach values from 40 to 45%.

4.2. Simulation results

A sensitivity analysis was performed on the pilot-scale model, as shown in **Error! Reference source not found.**, to understand the effect that CO₂ removal efficiencies of the membrane, and the

percentage of recycled flue gas, might have on CO₂ levels in the flue gas. Membrane efficiencies were set at 11, 20 and 90% under stoichiometric conditions and using an excess of air of 5% (molar) over the stoichiometric. The values set for the percentage of flue gas recycle (FGR or EGR) to the process were: 20, 30, 40, 50 and 60%. Table 4 shows the selected results extracted from this sensitivity analysis, for a thermal power generation of 100 kW.

Table 4: Selected results for CO₂ level in the flue gas from Aspen Plus simulation for the pilot-scale model varying CO₂ removal efficiency and percentage of recycled flue gas

<i>CO₂ Removal efficiency</i>	<i>20% EGR</i>	<i>30% EGR</i>	<i>40% EGR</i>	<i>50% EGR</i>	<i>60% EGR</i>
11%	7.6	7.7	7.8	7.9	8.0
20%	7.8	7.9	8.1	8.2	8.4
90%	9.0	10.0	11.2	12.8	14.9

Another parameter that was varied for this study was the load of the combustor, to look at how the process would behave at partial loads. The values defined with this aim were: 40, 50, 60, 75 and 100% of the total load (100 kW) of the combustor.

The membrane model developed as part of this work, using empirically deduced operating parameters such as the global mass transfer coefficient and the permeance detected for the studied membrane (PDMS), helped to specify some key aspects in the design of the pilot-scale membrane rig. The information extracted from the bench-scale membrane plant together with the membrane model, allowed the calculation of the area of membrane needed for the flue gas stream generated in the 100 kW combustor. This area was calculated to be 230 m², considering flue gas: sweep gas ratio of 1, combustion at atmospheric pressure and an excess of air supplied to the combustion of around 5%. The other key factor was to define the efficiency of CO₂ removal for the membrane, which here was 12.6%. An example explaining the physical meaning for the efficiency of the membrane is provided by a case study where stoichiometric air is supplied for combustion and a value of 60% of the flue gas is treated through the membrane section; in this situation, calculations suggest the membrane would allow one to produce a CO₂ concentration at the exit of the combustor of 9.8% (v/v).

5. Conclusions

A flexible pilot-scale CO₂ membrane combustion system (100 kW) with exhaust gas recirculation and selective exhaust gas recirculation, has been designed, installed and commissioned at Cranfield University. Selective recirculation of the flue gases was achieved using a CO₂-selective membrane unit with PDMS polymeric tubes.

Two models were developed in Aspen Plus, one focused on the mass and energy balance of the pilot-scale plant and the other focused on realistically simulating the membrane performance. The second model was an improved version of the first one where empirical mass transfer parameters were applied to predict the CO₂ levels that can be reached at the exit of the combustor after selectively recycling part of the flue gas generated. A bench-scale membrane rig was designed and commissioned to study aspects related to the mass transfer of CO₂ through the membrane and its removal efficiency. The empirical values obtained from the operation with the bench-scale rig: global mass transfer coefficient and permeance of the membrane were used to develop a model in Aspen Plus. The results are used for scale-up studies and process verification of commercial-scale simulations based on the results of pilot plant operation. The process simulation shows that the membrane unit can enhance the concentration of CO₂ in flue gases up to 9.8% (v/v), given the removal efficiency of the PDMS membrane of 12.6%. Other values have been predicted for the CO₂ concentration for cases when using membranes with higher removal efficiencies (20 and 90%). It can be concluded from these simulation results that more research is required to find more suitable materials for membranes that optimise the CO₂ removal efficiency from the flue gas.

Preliminary experimental results show a CO₂ level of 7.3% in the flue gases which is equivalent to a recycle ratio of 20%. The research demonstrates that the concept of EGR and S-EGR can improve the efficiency of post-combustion capture in gas-fired power plant by a factor of four or more.

Acknowledgements

This work was carried out as part of a UK consortium called Gas - Future Advanced Capture Technology Systems (Gas-FACTS) on enhancing the performance in capturing CO₂ from natural gas-fired power plants. Mr. Howard Smith is acknowledged for the major assistance he provided in building and operating the test rig.

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