A Novel Gas Switching Combustion Reactor for Power Production with Integrated CO₂ Capture: Sensitivity to the Fuel Type

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Abstract

This paper reports further experimental demonstration of the Gas Switching Combustion (GSC) concept with a focus on the concept sensitivity to the fuel type. The GSC concept offers an alternative to the conventional chemical looping combustion (CLC) process for power production with integrated CO₂ capture. In this concept, oxidation and reduction of the oxygen carrier take place in the same reactor by alternating air and fuel gas feeds into the reactor. This completely avoids any need for oxygen carrier circulation and thereby avoids many technical and scale-up challenges related to the looping concept. First demonstration of the concept with carbon monoxide (CO) as reducing gas and Ni-based oxygen carrier showed that the reactor could operate autothermally to continuously convert cold feed gases into hot product gases.

In this paper, the GSC concept is further tested with syngas and methane as reducing gases. First testing of the reactor with carbon monoxide showed large heat losses, but operation could still be maintained. Experiments with syngas (62% CO and 38% H₂) and methane were however carried out with external heat supply to compensate for part heat losses and show the heat removal stage which is an important component of the GSC concept. Three target temperatures have been tested for both fuel gases.

Syngas showed complete conversion with no carbon deposition, leading thereby to high CO₂ purity (95%) and capture efficiency (95%). Slightly low CO₂ purity has been achieved (around 80%) when methane was used which was mainly caused by unconverted methane and existence of carbon monoxide in the gas stream. High CO₂ capture efficiency (95%) has however been achieved due to the absence of carbon deposition.

In the light of its excellent performance, syngas is suitable to fuel the GSC reactor. A syngas fuelled GSC reactor cluster would be deployed in a baseload integrated gasification combined cycle (IGCC) plant where its relative simplicity regarding design, scaling and pressurization could lead to a cost effective solution relative to alternative technologies.
Methane could also be potentially used as fuel for the GSC concept if the issue of the slow reactivity at the start of the reduction stage is solved. This could be achieved through the use of oxygen carriers with low catalytic activity with respect to the steam methane reforming reaction. Successful operation with methane could be important in a future electricity market where moderate penetrations of intermittent wind/solar power require flexible power production with integrated CO₂ capture.

1 Introduction

Carbon capture and storage (CCS) is increasingly seen as a short term sustainable way for fulfilling the world energy needs while minimizing the environmental impact of fossil fuel combustion [1, 2]. Chemical-looping combustion (CLC) has arisen as a promising technology to carry out power production with CO₂ capture at lower energy penalty compared to other CO₂ capture technologies, such as pre- and post-combustion technologies [3]. CLC is a combustion process which integrates power generation and CO₂ capture. It consists of two interconnected reactors, an air and a fuel reactor with a solid oxygen carrier circulating between them, thereby playing the role of oxygen transporter which transfers the oxygen required for fuel combustion from the air to the fuel. Consequently, any contact between the fuel and the air is inherently avoided in this process, and therefore a pure CO₂ stream, ready for compression and sequestration, after steam removal, is produced [4].

In addition to development and selection of suitable oxygen carrier materials [5, 6], research on CLC has so far been focused predominantly on the use of dual Circulating Fluidized Beds (CFBs) configuration for the recirculation of the oxygen carrier between the air and the fuel reactors [3, 6]. Although this configuration has been demonstrated experimentally at lab and pilot scales [7-11], the dual CFB-based CLC process is still facing many technical and operational complexities which arise mainly from the interconnected reactors configuration. Aside from the design and operational complexity created by the need to manage the solids exchange so that mass and heat balances within the closed loop are fulfilled, the exchange of solids itself brings additional costs and complexity. This circulation between the two reactors requires efficient and costly particles separation system such as a cyclone. This particle separation is particularly difficult due to the extremely harsh reactive and high temperature conditions. These challenges magnify under high pressure operation at which the CLC technology is more efficient compared with other capture technologies.

Attempts have followed in recent years to address these issues where reactor concepts with no external solid circulation have been proposed [12-14]. The Gas Switching Combustion (GSC) concept which is the subject of this paper has been proposed as an alternative to the interconnected fluidized bed reactors used in the standard CLC process, where it uses a single fluidized bed reactor with alternating air and gaseous fuel feeds into the reactor (Figure 1). External solids circulation is inherently avoided by the use of a switching mechanism which allows periodic feeding of fuel and air streams to the single reactor where the oxygen carrier material undergoes redox reactions. The first experimental demonstration of this concept has been carried out using carbon monoxide (CO) as fuel [15]. High CO₂ capture efficiency and CO₂ purity have been achieved by this process while maintaining simple operation and
control of the process. This concept will bring other interesting benefits such as the ability of operation under high pressure conditions and ease of scale up; pressurized bubbling fluidized bed is indeed a mature technology and has been available at industrial scale for decades.

As a clean combustion technology for power production, the ultimate purpose of the GSC is its integration in a natural gas fired power plant (NGCC) or in an Integrated Gasification Combined Cycle power plant (IGCC). A recent process simulation study [16] confirmed the GSC-IGCC concept as a promising concept for high-efficiency baseload power-production with integrated CO₂ separation. As a first step towards the demonstration of the GSC concept, however, its behavior with syngas derived from solids fuel gasification and methane should be understood. This paper will therefore investigate the GSC concept flexibility to the fuel type. Specifically, methane and syngas are evaluated based on performance parameters such as CO₂ capture efficiency, CO₂ purity, carbon deposition and temperature rise. Ultimately, the most suitable gaseous fuel for this concept will be identified.

![Figure 1: Simple illustration of a GSC reactor. Air and fuel streams enter at room temperature and hot process gases are delivered to the downstream power cycle and CO₂ drying and compression unit.](image)

2 Experiments and methods

2.1 Experimental set up

The experimental setup (Figure 2) consisted of a pseudo-2D vertical column with a height of 1.5 m, a width of 0.3 m and a depth of 0.015 m. The pseudo-2D configuration was selected for the purpose of extracting local experimental data for detailed CFD model validation and was based on a cold flow unit with similar dimensions [17]. The reactor column was made from Inconnel 600 in order to withstand the harsh conditions of high temperatures (up to 1000 °C) and reacting gas-solid flows. A ceramic porous plate with 40 micron pores and 7 mm
thickness was used as the gas distributor. A freeboard was added to the top to prevent particles losses. The setup was equipped with four electrical heaters (Thermcraft Inc.) installed on the back of the reactor in order to heat up the reactor to the target temperature (referred also as the operating temperature). Each of these heaters is controlled separately allowing for full control of the heat supply to the desired region in the reactor. Blanket insulation (25 cm thickness) was used to maintain the heat in the system.

Three mass flow controllers (Bronkhors BV) were used for feeding gases to the reactor. The three gas lines gather at a three-way electrical valve installed before reaching the distributor. This three-way electrical valve is used to switch between feed gases. A cooler was positioned at the outlet of the reactor to cool down the stream of hot gases before it is sent to the vent.

The gas composition was measured using a Mass Spectrometer from MKS Instruments Inc. and the temperature was measured in each port along the entire front of the reactor using type K thermocouples. All the measurement instruments, flow controlling devices were controlled through a Labview application. The Labview application was also used for data acquisition and storage. Further details about the experimental setup can be found in [15, 18].

![Figure 2: Schematic representative of the experimental set up.](image)
2.2 Operation

All GSC demonstration experiments reported in this paper were completed with an amount of oxygen carrier material corresponding to a 0.3 m static bed height in the reactor. Nickel oxide particles supported on Al₂O₃ (manufactured by VITO) were used as the oxygen carrier. The NiO/Al₂O₃ ratio is 65/35 and the particle size cut-offs D₁₀, D₅₀ and D₉₀ were determined to be 117.4, 161.7 and 231.3 µm respectively. About 37% of the final particle consists of free NiO which is available for reaction. The powder has a loosely packed density of 1950 kg/m³ and a tapped density of 2166 kg/m³. This oxygen carrier was selected because it has been successfully used in previous studies [19, 20].

In autothermal operation, the reactor is first heat up externally to the target temperature before starting gas switching. When heaters are switched off a pure gaseous fuel is fed for a fixed amount of time called as the "fuel time" to reduce the oxygen carrier. A feed with pure air follows to oxidize the reduced oxygen carrier and remove the heat which builds up from the highly exothermic oxidation reaction. Five seconds purging with pure N₂ is applied between the air and the fuel stages to avoid direct contact between fuel and air and thereby eliminate the risk of explosion.

Experiments were performed with three gaseous fuels with 4 minutes fuel time; CO was used for first testing of the reactor, then a detailed study with syngas and methane has followed. Constant gas feed of 45 Nl/min was applied for both reduction and oxidation stages. Various dependent variables were extracted from each experiment: the CO₂ purity, the CO₂ capture efficiency, the amount of carbon deposition, the maximum temperature variation and the oxidation/reduction time ratio (henceforth called the Oxi/Red time ratio). All dependent variables are averaged over the three complete redox cycles.

Throughout the study, all temperature measurements are taken at 0.3 m in the central axis of the reactor. Temperature variations at this point are followed throughout the experiment and used to determine the start and end of the redox cycles. Gas species measurements are taken at a height of 0.7 m which is a point in the freeboard well above the expanded bed. These measurements are used to determine the CO₂ purity and capture efficiency as well as the carbon deposition.

The pressure along the height of the reactor is measured regularly in order to quantify the bed weight and to inspect any unexpected loss of particles caused by elutriation of particles due to gas expansion when the reactor reaches higher temperatures.

2.3 Reactor performance

The performance of the GSC concept is assessed based on the quantification of the following dependent variables:

- CO₂ capture efficiency. This was calculated by determining the percentage of the total produced CO₂ which exited in the outlet gas over the entire OXI stage (OXI will refer to oxidation and heat removal stage to avoid any confusion). The CO₂ capture efficiency was then taken as 100% minus the percentage of CO₂ that escaped to the atmosphere with the depleted air. Naturally, a high degree of CO₂ capture is desired in order to keep greenhouse gas emissions minimal.
• CO₂ purity. This was calculated by determining the amount of depleted air and unconverted gaseous fuel present in the outlet stream over the entire reduction stage. The CO₂ purity was then taken as 100% minus the percentage of impurities (depleted air and gaseous fuel) in the reduction stage outlet stream which would be sent to compression and storage in a real plant. Naturally, a high CO₂ purity is desired so that the volume of gas to be compressed, transported and stored is kept to a minimum.

• Carbon deposition. Carbon deposition reactions (2CO → CO₂ + C or CH₄→ C + 2H₂) can occur on the Ni-based oxygen carrier (employed in this study) in the reduction stage to subsequently be combusted in the OXI stage and released to the atmosphere as CO₂. This is an undesired phenomenon and it was quantified as the molar ratio of the total amount of CO₂ released during the OXI stage and the total amount of CO/CH₄ fed to the reactor during the fuel stage. Carbon deposition was measured in a separate cycle where a long purging stage with nitrogen was applied after the reduction stage. This eliminated CO₂ leakage and ensured that all CO₂ measured during the subsequent OXI stage could be safely attributed to combustion of the deposited carbon.

• Temperature rise. Due to the transient cycling operation of the reactor, the reactor temperature will vary throughout the cycle (Figure 3). It is important that this variation is not so large that lower temperatures cause incomplete fuel conversion or higher temperatures damage the reactor or downstream equipment. The difference between the maximum and minimum temperatures in the redox cycle will therefore be recorded in each experiment.

• Oxi/Red time ratio. With minor heat losses the OXI stage will be much longer than the fuel stage if constant flow rates are used. This is partially due to the molar density of oxygen in air which is lower than the molar density of the pure gaseous fuel, but primarily due to the large amount of heat must be removed from the system by the cold air after the oxygen carrier has been fully oxidized. The ratio between the OXI stage time and the reduction stage time will therefore be recorded for each experiment.

3 Results

Results will be presented and discussed as follows: first operation of the reactor has been carried out with CO as has been done previously in [15] to evaluate the reactor performance deviations from that found in [15] due to lower efficiency of the insulation material. Subsequently, a detailed study with syngas and methane at three target temperatures will be presented. Note that a mixture of hydrogen and carbon monoxide was used to simulate syngas (38% H₂ and 62% CO).

3.1 Carbon monoxide

In the ideal GSC process a target temperature is used as a threshold to switch from the OXI (oxidation and heat removal stages) to the reduction stage when the reactor temperature drops below the targeted temperature. A long heat removal stage using pure air follows the oxidation stage to remove heat built in the reactor due to the highly exothermic oxidation reaction. This mode of operation has been demonstrated in [15] where a good efficiency insulation system has been used.
In this study, experiments with CO as fuel have been carried at a target temperature of 700 °C. The external heaters were switched off after the reactor temperature reached 700 °C then periodic feeding with CO and air has followed for a number of redox cycles. Autothermal operation of the reactor has been achieved where the reactor temperature oscillated between minimal values reached at the start of the oxidation stage and maximal values reached at the end of the oxidation stage (Figure 3).

![Figure 3: Transient temperature at reactor heights of 0.02, 0.05 and 0.3 m over three redox cycles. CO was used as fuel at a target temperature of 700°C. The blue region is the reduction stage and the pink region is the OXI stage.](image)

In contrast to the previous study [15], no heat removal stage existed due to the substantial heat losses in the reactor. In order to maintain autothermal operation of the reactor, the switch to the reduction stage had to be made immediately when the temperature reaches the maximum. This switch occurred close to full oxidation of the oxygen carrier as can be observed from the transient oxygen mole fraction in the outlet gas stream approaching that of pure air as shown on Figure 4. Consequently a very low Oxi/Red time ratio has been found compared to that found in [15] despite the larger fuel feed.
Figure 4: Transient species mole fraction collected at a reactor height of 0.7 m over three redox cycles. CO was used as fuel at a target temperature of 700°C. The blue region is the reduction stage and the pink region is the OX1 stage.

3.2 Fuel sensitivity of the GSC concept

In industrial practice, natural gas and syngas (derived from gasification of coal or biomass) are the most important gases to consider for power production with the GSC concept via integration into an NGCC or IGCC power cycle. The following sections will therefore investigate and discuss the use of syngas and methane to fuel the GSC reactor.

In order to compensate for the large heat losses shown in the previous section, part of the heat losses will be compensated for via the external heaters. This will enable having longer redox cycle time, reaching the full oxidized state of the oxygen carrier and showing the existence of the heat removal stage which is an important component of the GSC concept. Yet, even with the external heat supply the cycle time is still far away from the ideal one as will be shown in the mass and energy balance calculations presented in the next section. Experiments have been carried out for target temperatures of 600, 700 and 800 °C. The reactor performance with each gaseous fuel was evaluated based on the dependent variables as specified above (section 2.3).

3.2.1 Mass and energy balance

Mass and energy balance calculations are made first to predict the ideal performance of the GSC concept with syngas and methane at a target temperature of 700 °C. The ideal cycle time $T_c$ can be calculated as specified by Eq.1, where $\Delta H_c$ is the total heat of combustion, $c_p$ is the mass specific heat capacity of the gas, $\Delta T$ is the gas temperature rise to reach the reactor temperature (note that the gas is fed at room temperature) and $m_g$ is the gas mass flow rate. For the case of syngas, for 240 s feed at 45 Nl/min with the assumption of full conversion, a
total heat of 1521.6 kJ is released in the reactor. Under the assumption of adiabatic operation, a total of 2.05 kg of gas is required to remove the heat released (the involved gases in the cycle are assumed to have the same specific heat capacity as air; 1.1 kJ/kg.K). In standard conditions of pressure and temperature, 45 Nl/min gas feed corresponds to 0.000859 kg/s. Therefore the time required for complete heat removal (the cycle time) is 3280.115 s. The expected ideal Oxi/Red ratio is then 12.66.

In the case of methane, the feed rate is 1/4 of that used for syngas (this is because 1 mole of CH₄ reacts with 4 moles of NiO). The heat released for 240 s feed with methane is 1636.08 kJ. This will require lower amount of air for heat removal than in the case of syngas and therefore shorter time cycle. After all calculation made the ideal Oxi/Red time ratio for methane is 9.69.

As shown above, however, such a high ratio of the oxidation and reduction stage times cannot be achieved in the lab-scale reactor employed in this study because of substantial heat losses.

\[ T_e = \frac{\Delta H_c}{c_p \Delta T m_g} \quad Eq. 1 \]

3.2.2 GSC with syngas

Complete syngas conversion has been achieved for the three operating temperatures. Consequently, high CO₂ purity values (95%) were achieved as shown in Table 1. When a complete fuel conversion is achieved, the only source of impurities in the reduction stage gas stream which could affect the CO₂ purity is the depleted air and nitrogen remaining in the reactor when switching to the reduction stage. No carbon deposition has been detected for the three operating temperatures, thereby leaving the CO₂ remaining in the reactor after the switch to air as the only source of impurities. This explains the high CO₂ capture efficiency reported in Table 1.

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>CO₂ purity</th>
<th>CO₂ capture efficiency</th>
<th>Oxi/Red time ratio</th>
<th>Temperature rise</th>
</tr>
</thead>
<tbody>
<tr>
<td>600</td>
<td>95.71677</td>
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<td>53.87</td>
</tr>
</tbody>
</table>

Figure 5 shows the gas composition data collected at a reactor height of 0.7 m over three redox cycles. It can be clearly seen from the oxygen mole fraction that the oxygen carrier reaches a fully oxidized state and a heat removal stage follows before switching to the reduction stage. Oxi/Red time ratios around 4 were found. This is still only 30 % of the expected ideal value predicted by the mass and energy balance despite the part of the heat losses supplied externally. The observed decrease in the Oxi/Red time ratio with the operating temperature results from the larger heat losses and heat removed by the gas feeds with further
increase in the temperature. The same reasons are behind the temperature rise decreases with the increase in the operating temperature.

![Figure 5: Transient species mole fraction collected at a reactor height of 0.7 m over three redox cycles. Syngas was used as fuel at a target temperature of 700°C. The blue region is the reduction stage and the pink region is the OXI stage.](image)

### 3.2.3 Methane

Operation with methane is found to be sensitive to the operating temperature. Incomplete methane conversion has been found for the three operating temperatures as can be seen in Table 2 although the conversion improves with higher temperatures. The transient methane mole fraction for a target temperature of 600 °C (Figure 7) shows higher amount of unconverted methane at the start of the reduction stage. Methane conversion then improves toward full conversion at the end of the reduction stage. A corresponding high percentage of carbon monoxide was also found at the start of the reduction stage and decreases following the same trend as methane toward full conversion at the end of the reduction stage.

The methane and carbon monoxide mole fraction trends can be explained as follows based on an understanding of the system of reactions between methane and Ni/NiO (a good overview can be found in [21]): At the start of the reduction stage methane conversion is slow because it splits first to carbon monoxide and hydrogen through the steam methane reforming (SMR) reaction (Eq.2). SMR is catalyzed by pure nickel and is therefore slow at the start of the reduction stage when the oxygen carrier is highly oxidized, leading to incomplete methane conversion. The highly reactive H₂ produced from the SMR reaction then reacts to completion with the abundant NiO available, but the less reactive CO shows some degree of fuel slip as part of it would be produced close to the bed surface via the SMR reaction. However, as more NiO is reduced to Ni, both the SMR and water-gas shift (WGS, Eq.3) reactions become faster. Increases in the SMR reaction improve methane conversion, while increases in the WGS reaction improve CO conversion to more reactive H₂. The continuous combustion of H₂ via
the heterogeneous reaction with NiO continuously drives both these equilibrium reactions to the right, thereby producing more H₂ and leading to improved overall conversion. When the oxygen carrier reaches a highly reduced state, hydrogen mole fraction in the gas stream increases due to the lack of oxygen (from NiO) leading to some H₂ slip. Higher operating temperatures however prolong the complete conversion of the produced hydrogen which leads to further conversion of carbon monoxide and methane. This explains the complete conversion of hydrogen observed for 700 and 800 °C (Table 2), over the whole reduction stage.

\[ \text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2 \quad \text{Eq. 2} \]

\[ \text{CO} + \text{H}_2\text{O} \leftrightarrow \text{CO}_2 + \text{H}_2 \quad \text{Eq. 3} \]

The incomplete conversion of methane and existence of no-combusted carbon monoxide reduces greatly the CO₂ purity when methane is used as a reducing gas. CO₂ purity in the range of 80% was found for 600 °C, with slight improvements when increasing the operating temperature to 800 °C. Operation under real power plant temperatures (1000 °C) would improve further the CO₂ purity. Otherwise, other oxygen carriers which favor direct methane combustion should be used.

The CO₂ capture efficiency is however maintained at a high level due to the absence of carbon deposition (Table 2). As mentioned above this parameter is only affected by the CO₂ leakage when switching to the oxidation phase.

Operation with methane exhibits higher temperature rise than syngas. This is attributed to the endothermic reaction of methane with nickel oxide oxygen carrier while syngas reaction with nickel oxide is slightly exothermic. Methane also results in lower decrease rate in the temperature rise than syngas. The improvement in methane conversion with the increase in the operating temperature results in further heat release in the reactor and compensates for the increased heat losses and heat removal by the cold gas feed. As for the Oxi/Red time ratio, values between 3.5 and 4 were found. This is still less than 40% the expected ideal values despite the external assistance with the heaters.
Figure 6: Transient species mole fraction collected at a reactor height of 0.7 m over one redox cycle. Methane was used as fuel at a target temperature of 600°C. The blue region is the reduction stage and the pink region is the OXI stage.

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>CH₄ mole fraction</th>
<th>H₂ mole fraction</th>
<th>CO mole fraction</th>
<th>CO₂ purity</th>
<th>CO₂ capture efficiency</th>
<th>Oxi/Red time ratio</th>
<th>Temperature rise</th>
</tr>
</thead>
<tbody>
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</tbody>
</table>

4 Conclusion

Further experimental demonstration of the Gas Switching Combustion (GSC) concept has been carried out in this paper. This concept has been proposed as an alternative of the conventional chemical looping combustion (CLC) process for power production with integrated CO₂ capture which uses interconnected fluidized bed reactors. In the GSC concept, a single fluidized bed reactor is used with alternating air and fuel gas feeds into a bed of oxygen carrier in the same reactor. This completely avoids any need for oxygen carrier circulation and thereby avoids many technical and scale-up challenges related to the looping concept.

After a previous successful demonstration of the GSC concept where it was showed that the reactor could operate autothermally to continuously convert cold feed gases into hot product gases, the GSC concept was further tested in this paper with other fuel gases of more commercial interest for power generation (syngas and methane). First testing of the reactor with carbon monoxide showed large heat losses in the reactor which imposes switching to the reduction stage just after the reactor temperature reaches the maximum (close to the end of the oxidation stage) in order to maintain autothermal operation. This results in unrealistically short cycles with low CO₂ capture efficiency and purity in addition to skipping the heat removal stage which is an important component of the GSC concept. Experiments with syngas (62% CO and 38% H₂) and methane were however carried out with partial external heat supply to compensate for part of heat losses and show the heat removal stage with these fuel gases. Three target temperatures were tested for both fuel gases.

Syngas showed complete conversion with no carbon deposition, leading thereby to high CO₂ purity (95%) and capture efficiency (95%). Incomplete methane conversion was however found together with existence of carbon monoxide in the gas stream. The transient gas composition measured in the freeboard showed a higher percentage of unconverted methane at the start of the reduction stage which improves toward full conversion at the end of the reduction stage. Gas composition data also showed the existence of large amount of carbon monoxide at the start of the reduction stage and which follows the same trend as methane where it reaches close to complete conversion at the end of the reduction stage. The existence of unconverted methane and carbon monoxide greatly reduces the CO₂ purity of the GSC
concept where values around 80 % were found. High CO₂ capture efficiency (95%) has however been achieved due to the absence of carbon deposition.

In the light of its excellent performance, syngas is suitable to fuel the GSC reactor. Its combination with a cheap oxygen carrier such as ilmenite would make the GSC concept more attractive as CO₂ capture technology to be integrated into an IGCC power plant. Methane could also be potentially used as fuel for the GSC concept if the issue of the slow reactivity at the start of the reduction stage is solved. This could be achieved through the use of oxygen carriers with low catalytic activity for the steam methane reforming reaction.

5 Acknowledgements

The authors would like to acknowledge the funding received from the Research Council of Norway which enabled the completion of this work. Joost Kors is acknowledged for constructing and maintaining the experimental setup.

6 References


