Two novel approaches for hydrogen production; chemical-looping reforming and steam reforming with carbon dioxide capture by chemical-looping combustion

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ABSTRACT

Two novel approaches for H$_2$ production by reforming of natural gas which utilize chemical-looping are presented. Chemical-looping reforming is a method for partial oxidation of fossil fuel in which a solid oxygen carrier is used as a source of undiluted oxygen. Steam reforming with CO$_2$ capture by chemical-looping combustion resembles conventional steam reforming but the furnace containing the reformer tubes is replaced by chemical-looping combustion. The two concepts have been compared with a process study, in which CO$_2$ capture have been considered. It is found that both alternatives have potential to achieve efficiencies in the order of 80%, excluding heat losses but including capture and compression of CO$_2$. Additionally, the feasibility of chemical-looping reforming has been demonstrated both in a fixed-bed reactor and in a continuously operating reactor consisting of two interconnected fluidized beds.

KEYWORDS: Chemical-looping reforming, chemical-looping combustion, steam reforming, partial oxidation, carbon sequestration

1. INTRODUCTION

In later years, concerns that emissions of CO$_2$ from combustion of fossil fuels might lead to disastrous changes in the climate of the earth have been growing. A majority of the scientific community concludes that global CO$_2$ emissions would need to be reduced greatly in the near future. This is a huge task. Whether we like it or not our current dependence on fossil fuels can hardly be overestimated.

One way to reduce CO$_2$ emissions that is receiving steadily increasing interest is carbon sequestration, which is the capture of CO$_2$ in flue gases and storing it where it is prevented from reaching the atmosphere. Carbon sequestration has potential to greatly reduce CO$_2$ emissions from large point sources such as power plants and industries. In contrast, CO$_2$ capture applications for small mobile emission sources such as cars, trucks and airplanes seem implausible.

H$_2$ contains no carbon so it can be utilized without CO$_2$ emissions. It can be burnt in combustion engines or gas turbines as it is or in mixture with other gaseous fuels. H$_2$ is also the ideal fuel in most types of fuel cells, which are applications that promises much higher efficiency than conventional engines. At present, all commercially available methods for large scale production of H$_2$ involves reforming of fossil fuels, which result in CO$_2$ emissions to the atmosphere and thus is contributing to anthropogenic climate change. But since large facilities are used for H$_2$ production, CO$_2$ capture should be feasible. If H$_2$ is produced from fossil fuel with CO$_2$ capture, it could be used as a CO$_2$-free fuel for vehicles or other applications.

2. BACKGROUND

Here two novel processes for H$_2$ production by reforming of fossil fuels with CO$_2$ capture are described and evaluated, namely chemical-looping reforming and steam reforming with CO$_2$ capture by chemical-looping combustion. The basic characteristics of the concepts are discussed and examined in a process study. In addition to this, experiments verifying the viability of the chemical-looping reforming concept are briefly presented. The paper summarizes the work of Rydén et al [1, 2, 3, 4].
2.1 Production of H\textsubscript{2} from fossil fuels

The gaseous mixture of H\textsubscript{2} and CO is usually referred to as synthesis gas. It is an important product that has many uses, for example as feedstock for production of ammonia, methanol and synthetic fuels. Synthesis gas can be produced from all kinds of fossil fuels. Steam reforming, reaction (1), is suitable for reforming of light fuels such as natural gas. Partial oxidation, reaction (2), can be used for heavier fuels such as oil or coal. CO\textsubscript{2} reforming, reaction (3), can be used if synthesis gas with extra high CO content is wanted.

\[
\text{Steam reforming: } \quad C_nH_m + nH_2O \rightarrow nCO + (n+\frac{m}{2})H_2 \quad \text{(highly endothermic)} \quad (1)
\]

\[
\text{Partial oxidation: } \quad C_nH_m + (\frac{m}{2})O_2 \rightarrow nCO + (\frac{m}{2})H_2 \quad \text{(slightly exothermic)} \quad (2)
\]

\[
\text{CO}_2 \text{ reforming: } \quad C_nH_m + nCO_2 \rightarrow (2n)CO + (\frac{m}{2})H_2 \quad \text{(highly endothermic)} \quad (3)
\]

At present, the most important method for synthesis gas generation is catalytic steam reforming of natural gas. Steam reforming takes place in reactor tubes packed with catalyst. Generally, the outlet temperature of the reformer tubes is 700-950 °C and the pressure 15-40 bar. The tubes are located inside a furnace, and energy for the endothermic reaction (1) is provided by direct firing.

Synthesis gas is also used for production of pure H\textsubscript{2}. Firstly, CO and H\textsubscript{2}O are converted into CO\textsubscript{2} and H\textsubscript{2} via water-gas shift, reaction (4).

\[
\text{Water-gas shift: } \quad CO + H_2O \rightarrow CO_2 + H_2 \quad \Delta H_{\text{fus}} = -38.9 \text{ kJ/mol} \quad (4)
\]

Water-gas shift takes place in a separate reactor vessel, usually operating at temperatures in the order of 300-500 °C. Additional steam can be added, but this is usually not needed for synthesis gas produced by steam reforming. If very low CO concentration is required, the first shift reactor is followed by a second reactor operating at lower temperature.

After the water-gas shift, a gas mixture consisting mainly of H\textsubscript{2} and CO\textsubscript{2} is obtained. In a steam reforming process there may be substantial amounts of unreformed CH\textsubscript{4} as well. H\textsubscript{2} is separated from impurities in one or more purification steps. The primary alternative for H\textsubscript{2} purification is pressure swing adsorption, where impurities are adsorbed in a bed of solid adsorbent at elevated pressure while H\textsubscript{2}, which is highly volatile and has low polarity, passes straight through. Another option for H\textsubscript{2} purification is absorption, which involves scrubbing the shifted synthesis gas with a solvent that interacts physically or chemically with impurities but not with H\textsubscript{2}. Amine solvents such as MEA or MDEA are well-suited for separation of H\textsubscript{2} and CO\textsubscript{2}. The resulting CO\textsubscript{2} rich solvent is pumped to a separate stripper column where CO\textsubscript{2} is released and the solvent is regenerated.

2.2 Chemical-looping combustion and chemical-looping reforming

Chemical-looping combustion is an innovative combustion technology that can be used for CO\textsubscript{2} capture in power generating processes. Direct contact between fuel and combustion air is avoided. Instead, a solid oxygen carrier performs the task of bringing oxygen from the air to the fuel. Chemical-looping combustion takes place in two separate reactors. In the fuel reactor the oxygen carrier is reduced by oxidizing the fuel. In the air reactor the oxygen carrier is oxidized with air. The sum of reactions is combustion of the fuel with oxygen. The combustion products are not diluted with N\textsubscript{2} so pure CO\textsubscript{2} can be recovered by condensation, see figure 1 below.

Suitable oxygen carriers include metal oxides such as Fe\textsubscript{2}O\textsubscript{3}, Mn\textsubscript{3}O\textsubscript{4}, CuO and NiO. An overview of the research dealing with oxygen-carriers for chemical-looping combustion can be found in the works of Cho [5], Johansson [6] and Adánez et al [7]. Information of various subjects regarding chemical-looping combustion such as design of experimental reactors, power production with CO\textsubscript{2} capture and more about oxygen-carriers can be found in the doctoral theses by Brandvoll [8], Johansson [9] and Wolf [10]. Although the oxygen carrier does not necessarily have to be a metal oxide, the abbreviation MeO is usually used to describe the oxidized form while Me is used for the reduced form.

Chemical-looping reforming is similar to chemical-looping combustion, but complete oxidation of the fuel is prevented by using low air to fuel ratio. Hence chemical-looping reforming can be described as a process for partial oxidation of hydrocarbon fuels that it utilizing chemical-looping as a source of undiluted oxygen. This is a considerable advantage compared to conventional technology since the need for expensive and power consuming air separation is eliminated, see figure 2.
Oxygen carrier particles for chemical-looping reforming have been examined by Zafar et al [11], who made tests in a fluidized-bed reactor with oxygen-carrier particles as fluidizing agent, and by Mattisson et al [12]. These two studies indicates high reaction rate and good selectivity towards H\(_2\) and CO for oxygen carriers with NiO as active phase, while oxygen carriers based on Fe\(_2\)O\(_3\), CuO and Mn\(_3\)O\(_4\) suffered from poor selectivity and produced CO\(_2\), H\(_2\)O and unreformed CH\(_4\) rather than CO and H\(_2\).

If the fuel is CH\(_4\), the oxygen carrier is NiO and the reactor temperature is 1200 K, reaction (5) occurs in the air reactor.

\[
\text{Regeneration:} \quad \text{Ni} + \frac{1}{2}\text{O}_2 \rightarrow \text{NiO} \quad \Delta H_{1200} = -234 \text{ kJ/mol} \tag{5}
\]

In the fuel reactor, reactions (6-9) may occur, depending on the air ratio. Steam or CO\(_2\) could be added to the fuel to enhance the relative importance of reaction (8) or reaction (9) respectively. This could be used to adjust the H\(_2)/CO\) ratio in produced synthesis gas or to suppress formation of solid carbon in the fuel reactor. For chemical-looping combustion as much fuel as possible should be completely oxidized according to reaction (6).

\[
\begin{align*}
\text{Oxidation:} & \quad \text{CH}_4 + 4\text{NiO} \rightarrow \text{CO}_2 + 2\text{H}_2\text{O} + 4\text{Ni} \quad \Delta H_{1200} = 136 \text{ kJ/mol} \tag{6} \\
\text{Partial oxidation:} & \quad \text{CH}_4 + \text{NiO} \rightarrow \text{CO} + 2\text{H}_2 + \text{Ni} \quad \Delta H_{1200} = 211 \text{ kJ/mol} \tag{7} \\
\text{Steam reforming:} & \quad \text{CH}_4 + \text{H}_2\text{O} \rightarrow \text{CO} + 3\text{H}_2 \quad \Delta H_{1200} = 226 \text{ kJ/mol} \tag{8} \\
\text{CO}_2 \text{ reforming:} & \quad \text{CH}_4 + \text{CO}_2 \rightarrow 2\text{CO} + 2\text{H}_2 \quad \Delta H_{1200} = 259 \text{ kJ/mol} \tag{9}
\end{align*}
\]

Chemical-looping processes could be designed in several ways but circulating fluidized beds are likely to have an advantage over other alternatives since this design provides good contact between gas and solids and allows a smooth flow of oxygen-carrier particles between the reactors. Continuous chemical-looping combustion in circulating fluidized beds has been demonstrated by Lyngfelt et al [13], Ryu et al [14], Johansson [9] and Abad et al [15].

### 3. DEMONSTRATION OF THE FEASIBILITY OF CHEMICAL-LOOPING REFORMING

In order to demonstrate the feasibility of chemical-looping reforming, tests were made in a continuously operating laboratory reactor that was built up from two interconnected fluidized beds. 350 g of particles consisting of 60 wt% NiO and 40 wt% MgAl\(_2\)O\(_4\) were used as bed material, oxygen carrier and reformer catalyst. The particles were prepared by freeze granulation and sintered at 1300 °C for 6 hours before they were sieved to a size of 90-212 µm. Natural gas was used as fuel, in some of the experiments with 25 vol% steam added. There was a continuous circulation of oxygen-carrier particles between the reactors. The reactor was operated at atmospheric pressure and a temperature of 830-920 °C. The air ratio was varied between the experiments.

In the fuel reactor, the particles were reduced by the fuel which in turn was partially oxidized to H\(_2\), CO, CO\(_2\) and H\(_2\)O according to reactions (6-9). There was complete conversion of CH\(_4\) and higher hydrocarbons and the actual composition depended on the air ratio. Low air ratio resulted in high concentrations of CO and H\(_2\). In the air reactor the reduced oxygen carrier was reoxidized with air according to reaction (5). For most cases, the amount of oxygen reacted with the fuel was 0.4-0.5 compared to what was required for complete...
combustion. 33-44% of the available NiO on the oxygen-carrier particles was reduced to Ni during operation. In total, 24 hours of reforming with dry natural gas and 17 hours with 75 vol% natural gas and 25 vol% steam were recorded.

The oxygen-carrier particles did not agglomerate, but soft lumps were found in the air reactor after the experiments had been finished. A likely explanation for this is the low gas velocity used, 0.1-0.5 m/s. Carbon formation was apparent for some of the experiments with dry natural gas since small amounts of carbon were accumulating in the fuel reactor, likely in the pressure measurement taps. The precise level of carbon formation could not be calculated since the reforming gas was analysed after that H$_2$O had been removed by condensation. There were also gas leakages between the reactor halves which complicated evaluation of measured data. With 25 vol% steam added to the fuel there was no or very small accumulation of carbon in the reactors and the composition of the reformer gas was close to thermodynamic equilibrium for the gas mixture, if it was assumed that there was no carbon formation. This indicates that the carbon formation was low when steam was added to the fuel.

The study verified the feasibility of chemical-looping reforming. Details can be found in the work of Rydén et al [4].

4. PROCESS EVALUATION

There are two possible ways to utilize chemical-looping technology for H$_2$ production. Firstly, it would be possible to use chemical-looping reforming to produce synthesis gas for production of H$_2$. This option is briefly described in section 4.1 below. Secondly, it would be possible to replace the reformer furnace in a conventional steam reforming process with chemical-looping combustion. This option would provide inherent CO$_2$ capture and is briefly described in section 4.2 below. A comparative process study of H$_2$ production with CO$_2$ capture can be found in section 4.3 below.

4.1 H$_2$ production with CO$_2$ capture by chemical-looping reforming (CLR)

A schematic description of H$_2$ production with CO$_2$ capture by chemical-looping reforming is shown in figure 3.

Air is compressed (AC) before entering the air reactor (AR). The fuel is mixed with some steam before the fuel reactor (FR) to suppress carbon formation. The outlet from the fuel reactor is cooled and extra steam is added before high-temperature shift (HTS) and low-temperature shift (LTS). H$_2$O is removed from the shifted gas in a condenser (COND) before CO$_2$ is captured by absorption with MDEA solvent (MDEA). CO$_2$ for sequestration is obtained by regenerating the MDEA solvent in a stripper column operating at low pressure. If desired, some of the H$_2$ produced could be burned in a separate combustor (COMB) to increase the power output of the gas turbine (GT).
4.2 Steam reforming with CO₂ capture by chemical-looping combustion (SR/CLC)

A schematic description of steam reforming with CO₂ capture by chemical-looping combustion is shown in figure 4.

![Figure 4. Schematic description H₂ production by steam reforming with CO₂ capture by chemical-looping combustion. Heat exchangers have been left out.](image)

This concept resembles conventional steam reforming, but the furnace is replaced with chemical-looping combustion. The air reactor (AR) and fuel reactor (FR) is operating at atmospheric pressure. Steam reforming (SR) takes place at elevated pressure in reformer tubes packed with catalyst located inside the fuel reactor. Heat transfer conditions are improved considerably compared to conventional steam reforming due to the fluidized environment. The reformer gas is treated in a single water-gas shift reactor (HTS). Water is removed from the process stream by cooling in a condenser (COND) before it enters a pressure swing adsorption unit (PSA). The H₂ produced is delivered at elevated pressure, but a fraction is needed for purging and regeneration of the adsorbers. The resulting PSA offgas consist of CO₂, purge H₂, unconverted CH₄ and small amounts of CO. The offgas is delivered at low pressure and is used as fuel and fluidizing gas in the chemical-looping fuel reactor. Hence pure CO₂ for sequestration is obtained by chemical-looping combustion.

4.3 Comparative process study: H₂ production with CO₂ capture

In order to examine the potential efficiency of the process concepts described above, computer models have been made that calculates stream compositions, temperatures, power consumption, heat transfer and other characteristics. Both alternatives have been considered as stand-alone plants optimized for H₂ production. Integration with other nearby facilities might change the picture somewhat but have not been considered. Process data for two basic cases can be found in table 1 below. Conservative process parameters have been used, so it may be possible to increase preheating and reactor temperature slightly.

In addition to the chemical-looping processes, data for a steam reforming process (SR/MDEA) have been included as reference. Here steam reforming with high H₂O/CH₄ ratio for high conversion of CH₄ is considered. Reducing the H₂O/CH₄ ratio would increase efficiency at the expense of CO₂ capture. CO₂ is captured by absorption with MDEA after water-gas shift and cooling in a condenser. H₂ purification is by pressure swing adsorption, where the H₂ recovery rate is adjusted so that the reformer furnace can be powered exclusively on H₂ rich pressure swing adsorption offgas. Details about this and other possible processes for H₂ production by reforming of natural gas with CO₂ capture can be found in the work of Rydén et al [1]. The following assumptions have been used for the examples presented in table 1:

- The fuel is CH₄ which is delivered to the plant at suitable pressure and a temperature of 20 °C.
- The product is H₂ at a pressure of 20 bar. When amine absorption is used for CO₂ capture, the H₂ produced will also contain impurities such as CH₄, CO and CO₂ so further purification would be needed for many applications.
• The outlets from the reactors are assumed to be at thermodynamic equilibrium.
• No heat losses have been considered.
• Preheating of fuel, steam and air proceeds to 600 °C.
• Power consumption in pumps for feed-water and amine solvent has been neglected.
• CO₂ for sequestration is compressed to 100 bar. Transport and storage of CO₂ have not been considered.
• Multi-stage compression with intercooling is considered. CO₂ compression takes place in three steps while air and H₂ compression takes place in two steps.
• The oxygen-carrier is NiO/Al₂O₃ and the particle circulation is adjusted to give a temperature difference between the air reactor and the fuel reactor of 50 K.
• ηiso = 0.85 and ηmec = 0.99 for fans, pumps, compressors and turbines.
• Tambient = 20 °C and pambient = 1 bar.
• ΔTmin = 20 K for all heat exchangers.
• Δpdrop = 1.0 bar for reformer tubes, 0.5 bar for water-gas shift, adsorption and pressure swing absorption and 0.1 bar for fluidized-bed reactors.
• For CLR, steam is added after the fuel reactor so a (H₂O/CO) ratio of 2.5 is obtained prior the water-gas shift.
• For SR/CLC, PSA is assumed to separate 90% of the H₂ in the process steam as product while 10% is used for purging the adsorbers.
• 10% excess air is used for combustion and chemical-looping combustion.
• MDEA absorption is assumed to capture 95% of the CO₂ available after the water-gas shift. Regeneration takes place in a stripper column at 130 °C and atmospheric pressure. Heat consumption for the regeneration is set to 60 kJ/mol CO₂.

Table 1. Key data for the examined processes.

<table>
<thead>
<tr>
<th></th>
<th>CLR</th>
<th>SR/CLC</th>
<th>SR/MDEA</th>
</tr>
</thead>
<tbody>
<tr>
<td>T reformer tubes, outlet (°C)</td>
<td>-</td>
<td>810</td>
<td>850</td>
</tr>
<tr>
<td>T air reactor (°C)</td>
<td>1050</td>
<td>960</td>
<td>-</td>
</tr>
<tr>
<td>T fuel reactor (°C)</td>
<td>1000</td>
<td>910</td>
<td>-</td>
</tr>
<tr>
<td>T reformer furnace, outlet (°C)</td>
<td>-</td>
<td>-</td>
<td>1100</td>
</tr>
<tr>
<td>T preheating (°C)</td>
<td>600</td>
<td>600</td>
<td>600</td>
</tr>
<tr>
<td>(H₂O/CH₄) ratio in fuel</td>
<td>0.5</td>
<td>3.5</td>
<td>5.0</td>
</tr>
<tr>
<td>(O₂/CH₄) ratio for CLR</td>
<td>0.6</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>P reforming (bar)</td>
<td>10.0</td>
<td>21.0</td>
<td>21.5</td>
</tr>
<tr>
<td>T high temperature shift, outlet (°C)</td>
<td>380</td>
<td>400</td>
<td>310</td>
</tr>
<tr>
<td>T low temperature shift, outlet (°C)</td>
<td>230</td>
<td>-</td>
<td>250</td>
</tr>
<tr>
<td>Recovery rate in PSA (%)</td>
<td>-</td>
<td>90.0</td>
<td>68.2</td>
</tr>
<tr>
<td>CO₂ capture efficiency for MDEA (%)</td>
<td>95.0</td>
<td>-</td>
<td>95.0</td>
</tr>
<tr>
<td>Egas turbine/HH₄,in (%)</td>
<td>4.60</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Ecompression air/HH₄,in (%)</td>
<td>2.87</td>
<td>0.11</td>
<td>-</td>
</tr>
<tr>
<td>Ecompression H₂/HH₄,in (%)</td>
<td>0.87</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Ecompression CO₂/HH₄,in (%)</td>
<td>1.78</td>
<td>1.93</td>
<td>1.69</td>
</tr>
<tr>
<td>E net demand/HH₄,in (%)</td>
<td>0.93</td>
<td>2.04</td>
<td>1.69</td>
</tr>
<tr>
<td>(nH₂,out/nH₄,in)</td>
<td>2.74</td>
<td>2.77</td>
<td>2.53</td>
</tr>
<tr>
<td>HH₂,out/HH₄,in (%)</td>
<td>82.7</td>
<td>83.4</td>
<td>76.2</td>
</tr>
<tr>
<td>Purity of produced H₂ (%)</td>
<td>97.4</td>
<td>≈100</td>
<td>≈100</td>
</tr>
<tr>
<td>CO₂ capture efficiency (%)</td>
<td>92.6</td>
<td>≈100</td>
<td>87.6</td>
</tr>
<tr>
<td>Weighed reformer efficiency (%)</td>
<td>81.1</td>
<td>79.8</td>
<td>73.3</td>
</tr>
</tbody>
</table>

The weighted reformer efficiency has been calculated according to expression (10).

Weighted reformer efficiency = \{ \left( \frac{(n_{H₂, out}/n_{CH₄, in}) + E_{net}}{(H_{H₂}/\eta_{el} \times H_{H₂})} \right) \times H_{H₂} \} / H_{CH₄}

(10)

\( (n_{H₂, out}/n_{CH₄, in}) \) = Moles H₂ produced per mole CH₄
\( E_{net} \) = Power surplus/demand (kJ/mol CH₄)
\( \eta_{el} \) = Reference efficiency for power generation with H₂ (0.58)
\( H_{CH₄} \) = Lower heating value for CH₄ (802.3 kJ/mol)
\( H_{H₂} \) = Lower heating value for H₂ (241.8 kJ/mol)
5. DISCUSSION

5.1 Chemical-looping reforming

Pressurized chemical-looping reforming potentially has very high efficiency, but the concept has some drawbacks as well. Fuel conversion is thermodynamically hampered by pressure so fuel reactor temperatures of 1000 °C or higher will be required to obtain sufficient conversion of the fuel. Integration with a gas turbine is indispensable in order to obtain high efficiency. Otherwise there would be a large efficiency penalty for air compression. It should also be noted that pressurized circulating fluidized beds are not conventional technology. Hence substantial development efforts would be needed to realize pressurized chemical-looping reforming.

Hydrogen production by chemical-looping reforming at atmospheric pressure is possible but would result in lower efficiency. Reforming involves large volumetric increase so the power consumption for compression of hydrogen to a suitable product pressure would be large. Additional fan power would be needed to overcome pressure drop in the water-gas shift reactors. If CO$_2$ capture is required an atmospheric process would need either a strong absorbing solvent such as MEA due to the low CO$_2$ partial pressure in produced synthesis gas, or gas compression prior to the CO$_2$ capture. Both these options would increase the energy penalty for CO$_2$ capture compared to the pressurized process described in figure 3. See the work of Rydén et al [1, 2, 4] for further discussion about pressurized and atmospheric chemical-looping reforming.

Due to the high overall efficiency, chemical-looping reforming is an interesting option for H$_2$ production. It is also interesting for generation of synthesis gas with low H$_2$/CO ratio. Such synthesis gas can not be produced directly by steam reforming and is needed for production of methanol, for Fischer-Tropsch synthesis and for other applications.

5.2 Steam reforming with CO$_2$ capture by chemical-looping combustion

The steam reforming concept has about the same efficiency as pressurized chemical-looping reforming and some advantages. There is no need for gas turbine, air compression or amine absorption. Additionally, the circulating fluidized beds can be operated at atmospheric pressure and moderate temperature while the reforming reactions still takes place at elevated pressure. This should make design and operation of the chemical-looping system comparably easy. Integration with pressure swing adsorption for production of H$_2$ with very high purity would be straightforward. CO$_2$ is captured by chemical-looping combustion so there should be almost 100% capture efficiency. Hence steam reforming integrated with chemical-looping combustion seems like an appealing option for H$_2$ production with CO$_2$ capture. It should also be pointed out that there is no efficiency penalty compared to conventional steam reforming, except for CO$_2$ compression. It would even be possible to increase H$_2$ production at the expense of export steam due to the favorable heat transfer from fluidized bed to reformer tube surface, see Rydén et al for information about this topic [2].

Possible drawbacks include erosion and corrosion of the reformer tubes. It is also clear that the fuel reactor will become rather large since it must be able to contain the reformer tubes, so the volume of oxygen-carrier particles that is needed will be large. A tentative reactor design can be found in the work of Rydén et al [2].

6. CONCLUSIONS

It can be concluded that chemical-looping reforming is a promising method for production of synthesis gas and H$_2$. The concept has been successfully demonstrated in a continuously operating reactor. If chemical-looping reforming is pressurized and integrated with a gas turbine, there is potential to obtain an overall efficiency in the order of 81% for H$_2$ production with CO$_2$ sequestration. Chemical-looping reforming also seems like an attractive method for synthesis gas generation.

Steam reforming with CO$_2$ capture by chemical-looping combustion also seems well suited for large scale production of high purity H$_2$ with CO$_2$ capture. This concept utilizes conventional technology and moderate temperature and pressure, and may be easier to put into practice than chemical-looping reforming. An overall efficiency in the order of 80% seems possible.

7. ACKNOWLEDGEMENTS

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