International Journal of Greenhouse Gas Control 64 (2017) 174–182

Contents lists available at ScienceDirect

International Journal of Greenhouse Gas Control

journal homepage: www.elsevier.com/locate/ijggc

Potential and limitations of power generation via chemical looping combustion of gaseous fuels

Florian Zerobin*, Tobias Pröll

Department of Material Sciences and Process Engineering, University of Natural Resources and Life Sciences, Peter Jordan Str. 82, A-1190 Vienna, Austria

ARTICLE INFO

Keywords:
Pressurized chemical looping combustion
Fluidized bed
Gas turbine combined cycle
CLC
GTCC

ABSTRACT

Pressurized chemical looping combustion (CLC) was studied with regard to its potential for power generation from gaseous fuels, such as natural gas. A process simulation model was set up for a simplified gas turbine combined cycle (GTCC) around a pressurized CLC reactor system and studied with respect to process parameters influencing electric efficiency. The process model is based on typical large scale GTCC arrangements with a gas turbine topping cycle and a heat recovery steam generator unit (HRSG). The results are compared to conventional GTCC process with similar arrangement and process parameters. It was found that the CLC process comes along with considerable technological limitations for the efficiency of the combined cycle: (i) turbine inlet temperature is limited by the oxygen carrier material, (ii) pressure drop of CLC AR path increases the required air compression work, and (iii) the requirement for low pressure steam for gas-sealing between air reactor and fuel reactor reduces the efficiency of the steam cycle. These effects limit the achievable net electric efficiency to values below 45%, which is similar to what could be reached with atmospheric pressure CLC in a conventional steam cycle power plant arrangement (e.g. Benson-type steam generator). The gas turbine inlet temperature (TIT) was identified as the greatest limitation to the process, the pressure ratio has to be reduced accordingly to maintain sufficient exhaust gas temperatures for the HRSG, which limits the efficiency potential of the gas turbine. As a conclusion, when it comes to power generation from gaseous fuels, these limitations will need to be resolved to make CLC technology competitive to conventional GTCC power plants combined with post combustion CO_2 capture technologies.

1. Introduction

Chemical looping combustion (CLC) was originally proposed by Ishida et al. (1987) to improve the efficiency of gas-turbine cycles. Such concepts had earlier been proposed by Knoche and Richter (1968) and were later studied by Ishida and Jin (1994) and by Anheden and Svedberg (1998) in the context of reducing the energy penalty for carbon capture from gas turbine combined cycle (GTCC) and integrated gasification combined cycle (IGCC) power plants. State of the art GTCC plants reach net electric efficiencies of up to 60% without CO_2 capture. According to Boot-Handford et al. (2014), post combustion capture units will impose an efficiency penalty of 8–10%-pts. on GTCC processes with about 60% net electric efficiency. If CLC is operated at atmospheric pressure and a state of the art steam cycle is used for power generation, the net electric efficiencies can be expected to reach about 45% without compression and purification of the CO_2 (Zerobin et al., 2016). For power generation from gaseous fuels, conventional steam generator cycles are hardly competitive to GTCC in combination with post combustion CO_2 capture technologies. Therefore, the technological step to pressurized operation seems necessary if CLC should be used for power production from natural gas. In a GTCC concept, CLC would need to be operated with high reactor exhaust gas temperatures up to 1200 °C or higher and at increased pressure up to 2 MPa. The high temperatures are a challenge for oxygen carrier materials, as well as the design of fluidized bed reactors, which are typically operated at temperatures below 1000 °C. Pressurized operation constitutes a critical challenge for the reactor system since the reaction intensity with respect to mass and surface of the solid oxygen carrier increases in proportion to pressure. Therefore and for other reasons, dual fluidized bed systems have only been operated at atmospheric pressure up to 950 °C so far. Alternating fixed bed vessels may be operated at increased pressure with the challenge of heat integration and limitations in the achievable temperature at turbine inlet. Early studies (Ishida et al., 1987; Knoche and Richter, 1968; Ishida and Jin, 1994; Anheden and Svedberg, 1998), as well as more recent works (Porrazzo et al., 2016; Naqvi et al., 2007) on the topic do not address such limitations in detail,

* Corresponding author.
E-mail address: florian.zerobin@boku.ac.at (F. Zerobin).

http://dx.doi.org/10.1016/j.ijggc.2017.07.011
Received 21 February 2017; Received in revised form 8 July 2017; Accepted 12 July 2017

Nomenclature

AR	Air reactor
CLC	Chemical looping combustion
CLC-CC	Chemical looping combustion combined cycle
FR	Fuel reactor
GTCC	Gas turbine combined cycle
HP	High steam pressure level
HRSG	Heat recovery steam generator
IP	Intermediate pressure level
LP	Low steam pressure level
TIT	Turbine inlet temperature
TOT	Turbine outlet temperature
$\dot{m}$	Mass flow (kg/s)
G^0	Gibbs energy (J/mol)
h	Specific enthalpy(kJ/kg)
h_S	Specific enthalpy after isentropic change of state (kJ/kg)
K_P	Equilibrium constant(–)
p	Pressure (Pa)
Δp	Fluidized bed reactor pressure drop (Pa)
P_{el}	Electric power output (W)
Q	Transferred heat (W)
Q_{fuel}	Fuel power (W)
R	Universal gas constant (J/mol K)
R_O	Oxygen transport capacity (kg/kg)
s	Specific entropy (kJ/kg K)
$\dot{S}$	Entropy flow (kJ/K s or W/s)
T	Temperature (K)
X_S	Oxygen carrier degree of oxidation (perovskite system) (–)
$X_{S,sim}$	Oxygen carrier degree of oxidation (simulated system)
Δ	Indicates difference (between AR and FR)
ν_i	Stoichiometric coefficient (–)
η_{el}	Electric efficiency (%)
η_S	Isentropic efficiency (%)

promising low penalties and a high efficiency potential for pressurized CLC in GTCC arrangements. The aim of the present work is to evaluate the efficiency potential of CLC-CC using a recent oxygen carrier material and taking characteristic shortcomings of fluidized bed CLC into account.

2. Material and methods

2.1. Process evaluation with the IPSEpro software package

Commercial flow sheet simulation software usually offers substance property libraries, fast equation solvers, and options for designing suitable reactor models. However, most tools do not offer a possibility for treatment of solid streams or of gas-solid reactions. Therefore, for the present work, the commercial equation-solver IPSEpro was chosen in combination with a custom-made model library designed for chemical looping processes (Bolhàr-Nordenkampf et al., 2009). The main advantages of IPSEpro compared to other software is its transparent structure with respect to the model equations and prior experience in describing a dual fluidized bed processes using this software. All process units strictly fulfill mass and energy conservation requirements. For description of relevant substances in CLC (Table 1), thermodynamic properties such as enthalpy, entropy or heat capacity can be calculated via equations of state. The IAPWS-IF97 formulation is used to calculate the properties of water and steam (Wagner and Kruse, 1998), The NASA polynomials after Burcat and McBride (1997) are used to calculate ideal gas properties and inorganic solids are calculated according to Barin (1995). Chemical reaction equilibrium of gas-solid and K_p gas-phase reactions is formulated using the equilibrium constant

$$\sum_i (\nu_i \cdot A_i) = 0 \tag{1}$$

$$K_p(T) := \prod_i p_i^{\nu_i} \tag{2}$$

The equilibrium constant can be calculated by minimization of the Gibbs free enthalpy ΔG_R^0.

$$\Delta G_R^0 = -R \cdot T \cdot ln(K_p) \tag{3}$$

2.2. Model of the gas-turbine combined cycle reference process

Fig. 1 shows the considered process model of the benchmark GTCC plant. As a basis for comparison, a commercially available gas turbine was chosen, namely the Siemens SGT5-4000F with a nominal electric output of the gas turbine of 307 MW and a gas turbine single-cycle efficiency of 40.0% (Siemens, 2016). A single pressure level design was chosen for the heat recovery steam generator unit in order to keep the complexity of the process model within reasonable limits – especially in view of the comparison with the more complex CLC scheme. Compression and expansion processes such as the compressor of a gas turbine unit, as well as turbines or turbine stages are modelled as adiabatic state change using the isentropic efficiency concept for description of thermodynamic non-ideality.

$$\eta_{S,\ Compressor} = \frac{h_{2s} - h_1}{h_2 - h_1} \tag{4}$$

Typical net efficiency values of up to 60% are achieved in modern GTCC power stations which are equipped with highly efficient steam processes with multiple pressure levels. The more simple single-pressure steam cycle with single reheat, which was chosen for the present comparison, means a slightly reduced steam cycle efficiency both in the benchmark case and in the CLC case. CO_2 capture from the exhaust gas stream is achieved via an MEA scrubbing process (Jordal et al., 2012) and the CO_2 stream is finally compressed to 110 bar in a three-stage compression part with intercooling.

2.3. Model of the chemical looping combustion (CLC) reactor

Descriptions of the principle and basic concept of CLC can be found in literature (Adanez et al., 2012; Pröll, 2015). Fig. 2 shows the chemical looping combustion core process in the modelling environment. The loop seal fluidization, which is typically done with low pressure steam, is not shown in Fig. 2. The respective amounts of steam can be added to the feed gas streams into the reactors to account for this requirement. The solids circulate between the two reactors, where composition and temperature change. Fresh oxygen carrier can be added continuously for bed material makeup or be set to zero if no bed material is considered lost as dust load on the gas streams. For means of completeness, a continuous removal of oxygen carrier is modelled.

Table 1
Oxygen carrier systems implemented in IPSEpro (appended from (Bolhàr-Nordenkampf et al., 2009)).

Cu	Fe	Mn	Ni	Co	CaS
Cu/CuO^*	Fe/FeO	Mn/MnO	Ni/NiO^*	Co/CoO^*	$CaS/CaSO_4^*$
Cu/Cu_2O	FeO/Fe_3O_4	MnO/$Mn_3O_4^*$	*) system practically considered for CLC		
Cu_2O/CuO^*	FeO/$Fe_2O_3^*$	MnO//$Mn_2O_3^*$	Support materials implemented: Al_2O_3, CaO, MgO, $CaAl_2O_4$, $MgAl_2O_4$, $CuAl_2O_4$, $NiAl_2O_4$, SiO_2, TiO_2, ZrO_2		
	$Fe_3O_4/Fe_2O_3^*$	Mn_3O_4/Mn_2O_3			
		Mn_2O_3/MnO_2			