Chemical Engineering Journal 294 (2016) 478–494

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Pre-combustion packed bed chemical looping (PCCL) technology for efficient H_2-rich gas production processes

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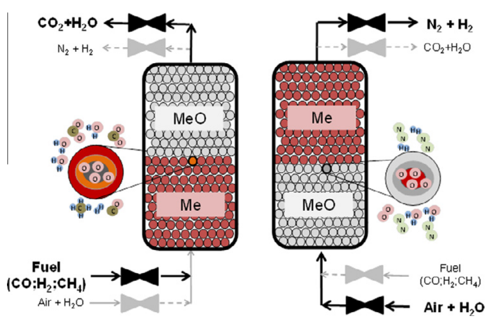
HIGHLIGHTS

- A novel process based on chemical looping for H_2/N_2 syngas production has been presented.
- The experimental proof of principle has been carried out in a 2 kW packed bed reactor.
- A Fe-based OC has been tested in the range 600–800 °C.
- The PCCL system has an electrical efficiency up to 51.5%.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 25 August 2015
Received in revised form 15 January 2016
Accepted 2 March 2016
Available online 8 March 2016

Keywords:
CLC
Pre-combustion
CCS
Ammonia production
Combined cycle
Packed bed reactor

ABSTRACT

A novel reactor system is presented and investigated for the production of a hydrogen rich gas stream for power or ammonia production, based on pre-combustion chemical looping (PCCL) technology using dynamically operated packed bed reactors. In this process, the oxygen carrier (OC) is alternately oxidized with a gas mixture of air and steam to produce a H_2/N_2 product gas stream combining oxidation by air and water-splitting, and subsequently reduced with syngas producing a concentrated CO_2 stream. The process is carried out at elevated pressure, but at intermediate temperature (in the range of 600–900 °C), which allows circumventing the extremely high temperatures required in chemical-looping combustion. In addition, the N_2/H_2 gas stream can be produced at the required composition for ammonia production, rendering this process also competitive with the conventional ammonia production. A preliminary experimental study has been carried out in a 2 kW_{th} packed bed reactor using an iron-based oxygen carrier. The influence of the operating temperature and the initial solid composition during the oxidation cycle on the H_2-rich gas yield has been investigated. The complete reduction to pure iron reduces the reactivity of the material due to sintering, whereas a controlled reduction to wüstite (FeO) allows to maintain a higher stability of the material, although the oxygen capacity is decreased.

A preliminary thermodynamic assessment of the integrated PCCL plant for power production with natural gas has been carried out reaching an electrical efficiency of 49–51.5% (depending on the plant arrangement) with a carbon capture rate above 95%. The main parameter affecting the plant performance was found to be the steam requirement during the oxidation cycle. The comparison with benchmark technologies has confirmed the potential of the PCCL system.

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http://dx.doi.org/10.1016/j.cej.2016.03.011
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Nomenclature

AGR	acid gas removal
AR	air reactor
BOP	balance of plant
CCS	carbon capture and storage
CGE	cold gas efficiency, $Q_{LHV,syngas}/Q_{LHV,coal}$
CLC	chemical looping combustion
CPU	CO_2 process unit
EDX	energy dispersive X-ray analysis
FR	fuel reactor
GT	gas turbine
HP-IP-LP	high-intermediate-low pressure
HR	heat removal phase
HRSC	heat recovery steam cycle
HRSG	heat recovery steam generator
HT-IT-LT	high-intermediate-low temperature
IC	Inter-cooled
IGCC	integrated gasification combined cycle
LHV	lower heating value, MJ/kg
MEA	mono-ethanol-ammine
NG	natural gas
NGCC	natural gas combined cycle
OC	oxygen carrier
Ox	oxidation phase
PBR	packed bed reactor
PCCL	pre-combustion chemical looping
PSA	pressure swing adsorption
Red	reduction phase
SC	steam cycle
SEM	scanning electron microscopic analysis
SH//RH	superheated/reheated
SPECCA	specific primary energy consumptions for CO_2 avoided, MJ_{LHV}/kg_{CO2}
ST	steam turbine
TGA	thermogravimetric analysis
TIT	turbine inlet temperature
TOT	turbine outlet temperature
WGS	water gas shift

Symbols and units

cp	gas/solid heat capacity J/kg-K
m	mass flow rate, kg/s
Q_{LHV}	thermal power, MW_{TH}
N	molar flow rate, kmol/s
p	pressure, bar
T	temperature, °C
E_{CO2}	CO_2 specific emissions, kg_{CO2}/MWh_E
ΔT	temperature difference, °C
η_{el}	net electric efficiency

1. Introduction

Reduction of greenhouse gas emissions is one of the most important challenges that the power industry will face in the next decades [1]. Carbon capture and storage (CCS) technologies could cut by at least one order of magnitude the CO_2 emissions from fossil fuel-fired power plants. CO_2 capture technologies can be applied according to three main configurations: (i) post-combustion capture for flue gases, (ii) pre-combustion capture producing carbon-free H_2-based fuel for a power cycle and (iii) oxy-combustion, generating a highly concentrated CO_2 stream after water condensation. All these concepts lead to energy efficiency penalties in the order of 10–15 percentage points when implemented with the current commercially available technologies [2].

Pre-combustion CO_2 capture technologies are based on different conversion processes that occur in series. Starting from the pretreatment of the primary fuel, the production of the syngas is typically carried out by steam reforming or coal gasification. The syngas, rich in H_2 and CO, is then mixed with steam and fed to the water gas shift (WGS) reactors to convert the CO into H_2 and CO_2. This process is normally carried out at two temperature levels to improve both the thermal efficiency, which is favored by operating a high temperature WGS stage at around 320–500 °C allowing to recover the heat of reaction by high pressure steam production, and the CO conversion, which is favored at lower temperature (around 185–300 °C) due to the chemical equilibrium constraint. The shifted fuel is then sent to the cold acid gas removal unit where CO_2 and sulfur species are removed by absorption. The CO_2 desorption is carried out by thermal regeneration using low pressure (LP) steam in the reboiler of the desorber column or by pressure reduction in flash chambers where the CO_2 is released and sent to the compressors train. Pre-combustion separation processes are well known in the industry and widely adopted for the production of H_2 and chemicals (i.e. ammonia); a great research effort is nowadays devoted to demonstrate the feasibility of this system also for clean power production by using a H_2-fuelled gas turbine combined with a steam cycle for an efficient thermal integration. Natural gas power plants integrated with pre-combustion CO_2 capture systems have been presented in recent years reaching an energy efficiency penalty of 9–12 percentage points with respect to the reference natural gas combined cycle [3–5], while in case of IGCC, the CO_2 capture integration costs about 7–11 percentage points [6–10]. Also ammonia plants use fossil fuels as primary feedstock that is decarbonized in order to produce a gas mixture of H_2/N_2 that is fed to the ammonia synthesis plant operated at high pressure (150–300 bar) and temperature in the range of 350–550 °C [11]. About 30% of the energy losses are determined by the several conversion and separation steps required [12,13] in case of natural gas. Moreover, the high capital cost of an ammonia plant is mostly associated to the costly H_2 production unit rather than the ammonia synthesis plant.

Among the different technologies that have been discussed and presented for CCS, chemical looping combustion (CLC) represents one of the most viable solution to achieve a very high CO_2 reduction with a reduced energy penalty [14]. CLC uses a metal oxide (called oxygen carrier, OC), which is oxidized in contact with air and afterward reduced (releasing the oxygen) in the presence of a reducing agent, such as a fuel: the CO_2 contained in the exhaust gases from the reduction reactions is nitrogen-free and thus easily separated at high purity after water condensation. Different oxygen carrier materials have been proposed for the CLC operation [15,16]. From an energy point of view, the oxidation reaction is always exothermic and the O_2-depleted air is produced at high temperature while the reduction reaction can be exothermic or endothermic depending both on the materials used as OC and the fuel type/composition.

For the application of CLC several different studies have been published in recent years mainly based on a pressurized circulating fluidized bed reactor (CFBR) operated with natural gas [17–19] or sulfur-free syngas from a coal gasification unit as fuel for the CLC loop operated at high pressure [20,21] (Fig. 1a). The performance of the plants studied varied from 43 to 52% for a hundreds-MW

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