

Full Length Article

Long-term pilot testing of the carbonate looping process in 1 MW_{th} scale

Jochen Hilz*, Martin Helbig, Martin Haaf, Alexander Daikeler, Jochen Ströhle, Bernd Eppele

Institute for Energy Systems and Technology, Technische Universität Darmstadt, Otto-Berndt-Str. 2, 64287 Darmstadt, Germany

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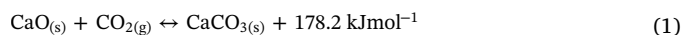
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ABSTRACT

The carbonate looping (CaL) process is an efficient post-combustion carbon capture technology to reduce the amount of CO₂ released by fossil fuelled power and industrial plants. This work presents the results gained in long-term pilot tests performed in a 1 MW_{th} CaL pilot plant at Technische Universität Darmstadt. The experimental data were obtained from continuous operation under realistic CaL process conditions, i.e. coal originated flue gas from a coal fired furnace to be decarbonized in the carbonator and oxy-fuel conditions in the calciner. Steady-state conditions, achieved while parameters were not changed during periods up to 60 h, delivered reliable experimental data to scale up the CaL process to industrial size. The feasibility of the CaL process in semi-industrial size was proven by steady-state CO₂ absorption for more than 1200 h with absorption rates in the carbonator higher than 90% and overall capture rates higher than 95% under a wide range of operating parameters, e.g. type of fuel, flue gas composition or reactor temperatures. Exemplary operation to achieve steady-state conditions is presented to show the evolution of the carbonator absorption efficiency and solid properties, such as conversion from CaO to CaCO₃ by carbonation and to CaSO₄ by sulphation. The carbonator performance was assessed by reasonably closing the carbon balances and applying the active space time as a key parameter to interpret the trends observed.

1. Introduction

Carbonate looping or calcium looping (CaL) is an efficient post-combustion CO₂ capture technology using limestone based sorbents, and which was initially proposed by Shimizu et al. [1]. CaL is particularly suited for retrofitting existing power plants. As shown in Fig. 1, the CO₂ contained in the flue gas from an emission source, e.g. an upstream power plant, is absorbed by calcium oxide (CaO) in the carbonator in an exothermic reaction at around 650 °C forming calcium carbonate (CaCO₃) according to Eq. (1).



The CaCO₃ is transferred to the calciner. By increasing the temperature up to around 900 °C, the CO₂, bound in the solid phase, is released in the endothermic backward reaction according to Eq. (1). A gas stream of highly concentrated CO₂ leaves the calciner, while the CaO is returned to the carbonator. In the basic concept, i.e. the directly heated or oxy-fired CaL process, the heat for the endothermic calcination reaction is provided by oxy-fuel combustion of fuel (e.g. coal) with oxygen from an air separation unit (ASU). Various concepts of optimized CaL processes have been proposed utilizing indirect heat transfer

from a combustor to the calciner [2–4]. A very promising approach for the indirectly heated CaL process (IHCaL) heat pipes has recently been tested [5].

The CaL technology promises substantial abatement in energy penalty [6–8] and costs [9–11] compared to other CO₂ capture technologies, e.g. amine scrubbing (post-combustion), integrated gasification combined cycle (pre-combustion) or oxy-fuel combustion. CaL is the only CO₂ post-combustion technology offering additional power generation from the additional heat input required for the sorbent regeneration since the calciner adds an oxy-fired new power plant system to the existing upstream plant. Both carbonation and calcination reactions run at very high temperatures allowing a highly efficient heat recovery from different sources offered by the CaL process.

The oxy-fired CaL process passed through a rapid development of the Technology Readiness Level (TRL) [12] from concept (TRL2) to demonstration at industrially relevant environment (TRL6) [13]. A system consisting of two interconnected circulating fluidized bed (CFB) reactors, a carbonator and a calciner, is the most developed CaL configuration. Synergies between CaL and existing CFB reactor systems at industrial scale promoted the rapid development of this technology during the last decade. Several research groups worldwide

Abbreviations: ASU, Air separation unit; CaL, Calcium/carbonate looping; CFB, Circulating fluidized bed; IHCaL, Indirectly heated carbonate looping process; LHV, Lower heating value; PSD, Particle size distribution; TGA, Thermogravimetric analysis; XRF, X-ray fluorescence

* Corresponding author.

E-mail address: jochen.hilz@est.tu-darmstadt.de (J. Hilz).

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Nomenclature

d_{50}	median particle diameter (μm)
$(dX/dt)_{\text{carb}}$	carbonation reaction rate (1/s)
E_{carb}	carbonator absorption efficiency (%)
$E_{\text{carb,eq}}$	maximum carbonator absorption efficiency at equilibrium conditions (%)
F_0	molar make-up flow ($\text{mol}/\text{m}^2\text{s}$)
f_{active}	active particle fraction of carbonator inventory (–)
$F_{\text{CO}_2,\text{carb,in}}$	molar flow of CO_2 entering the carbonator ($\text{mol}/\text{m}^2\text{s}$)
$F_{\text{CO}_2,\text{carb,out}}$	molar flow of CO_2 leaving the carbonator ($\text{mol}/\text{m}^2\text{s}$)
G_s	solid circulation flow between reactors ($\text{kg}/\text{m}^2\text{s}$)
k_s	reaction rate constant (1/s)
LR	sorbent looping ratio ($\text{mol}_{\text{Ca}}/\text{mol}_{\text{CO}_2}$)
M_{Ca}	weighted molar weight of Ca particles (g/mol)
MR	make-up ratio ($\text{mol}_{\text{Ca}}/\text{mol}_{\text{CO}_2}$)
$n_{\text{Ca,carb}}$	Ca inventory in carbonator (mol/m^2)
$n_{\text{Ca,carb,active}}$	active Ca inventory in carbonator (mol/m^2)
t^*	time required to increase the carbonate content from X_{calc} to X_{ave} (s)
T_{calc}	average calciner temperature ($^{\circ}\text{C}$)
T_{carb}	average carbonator temperature ($^{\circ}\text{C}$)
T_{carb}	average carbonator temperature ($^{\circ}\text{C}$)

$W_{\text{s,carb}}$	total carbonator inventory (kg/m^2)
X_{ave}	average CO_2 carrying capacity ($\text{mol}_{\text{CaCO}_3}/\text{mol}_{\text{Ca}}$)
X_{calc}	molar carbonate content of sorbent leaving the calciner ($\text{mol}_{\text{CaCO}_3}/\text{mol}_{\text{Ca}}$)
X_{carb}	molar carbonate content of sorbent leaving the carbonator ($\text{mol}_{\text{CaCO}_3}/\text{mol}_{\text{Ca}}$)
X_{sulphur}	molar sulphur content of the sorbent ($\text{mol}_{\text{S}}/\text{mol}_{\text{Ca}}$)
y_{CaO}	CaO mass fraction of the sorbent (wt.%)
y_{CaCO_3}	CaCO_3 mass fraction of the sorbent (wt.%)
y_{CaSO_4}	CaSO_4 mass fraction of the sorbent (wt.%)
y_{ash}	ash mass fraction of the sorbent (wt.%)
$v_{\text{CO}_2,\text{calc,in}}$	inlet calciner volumetric CO_2 fraction (vol.%)
$\overline{v_{\text{CO}_2,\text{carb}}}$	average carbonator volumetric CO_2 fraction (vol.%)
$v_{\text{CO}_2,\text{carb,eq}}$	equilibrium carbonator volumetric CO_2 fraction (vol.%)
$v_{\text{CO}_2,\text{carb,in}}$	inlet carbonator volumetric CO_2 fraction (vol.%)
$v_{\text{CO}_2,\text{carb,out}}$	outlet carbonator volumetric CO_2 fraction (vol.%)
$v_{\text{CO}_2,\text{eq}}$	equilibrium volumetric CO_2 fraction (vol.%)
$v_{\text{H}_2\text{O,calc,in}}$	inlet calciner volumetric H_2O fraction (vol.%)
$v_{\text{O}_2,\text{calc,in}}$	inlet calciner volumetric O_2 fraction (vol.%)
τ_{active}	active space time (s)
φ	gas–solid contacting effectivity factor (–)
φk_s	apparent carbonation rate constant (1/s)

demonstrated the feasibility of the CaL process. Various pilot plants in sufficient scale and with different reactor configuration and capacities were built and operated, e.g. the 0.2 MW_{th} pilot in Stuttgart [14], the 1 MW_{th} pilot in Darmstadt [15], the 1.7 MW_{th} pilot in La Pereda [16] or the 1.9 MW_{th} pilot in Taiwan [17].

As indicated before, the development of the CaL technology achieved a great progress confirming the feasibility of the technology. Nevertheless, a challenge of the CaL technology is a decreasing reactivity with increasing number of cycles of carbonation and calcination [18]. To diminish this effect, a continuous feed of limestone as make-up is required in order to maintain a certain capacity of the sorbent to absorb CO_2 [19]. Consequently, a realistic assessment of sorbent performance can only be provided by a homogeneous mixing of existing and continuous make-up feed. Therefore, extensive long-term operating is required to achieve steady conditions of the sorbent phase.

In addition, the carbonation/calcination reaction is dependent on the CO_2 partial pressure. The oxy-fuel combustion in the calciner implies calcination conditions under high CO_2 concentrations and the presence of water vapour. Various investigations [20–22] pointed out that CO_2 partial pressure and water vapour significantly influence the calcination of the sorbent. Realistic operating conditions can only be achieved by recirculating flue gas instead of mixing artificial calciner primary gas consisting of O_2 and CO_2 .

To overcome the uncertainties of previous pilot tests as a basis for scale-up scenarios, the long-term operation under realistic process conditions, e.g. homogeneous mixing of inventory and make as well as oxy-calcination under flue gas recirculation, is required to obtain the confidence about the performance of the CaL technology. Therefore,

extensive pilot tests in an upgraded pilot plant at Technische Universität Darmstadt were performed to take the next step towards maturity of the technology addressing the challenges mentioned before. The achievement of steady-state operation has recently been shown by Helbig et al. [23]. This approach guarantees a solid ground for the scale-up of a 20 MW_{th} pilot plant designed and engineered applying the experimental data from long-term pilot tests.

This paper contributes to the progress in the development of the CaL post-combustion CO_2 capture technology. The results obtained from long-term pilot testing in the CaL pilot plant at Technische Universität Darmstadt are presented focusing on the closure of the carbon balance during continuous pilot operation taking into account two out of four test campaigns. Thereby, different types of fuels as well as fuel with various particle size distributions were fired in the calciner. A modelling approach successfully applied in small scale experiments [24–26] is used to interpret the experimental results.

2. Experimental

2.1. Pilot plant setup

A semi-industrial scale CaL pilot plant consisting of two interconnected CFB reactors and a combustion chamber with a thermal capacity of 1 MW_{th} each is located at Technische Universität Darmstadt. In previous tests [15,27], the main focus was proof of operation in 1 MW_{th} scale. Therefore, CaL tests were accomplished decarbonizing synthetic flue gas, a mixture of air and CO_2 . The calciner was operated with oxygen enriched air in these tests not representing realistic operating conditions in terms of calcination. As a consequence from these results, the pilot plant was upgraded to operate with coal originated flue gas from the furnace and oxy-combustion in the calciner.

The scheme of upgraded 1 MW_{th} CaL pilot plant is shown in Fig. 2. The CaL reactor systems consists of a carbonator with an inner diameter of 0.6 m and a height of 8.6 m. The dimensions of the calciner are 0.4 m and 11 m, respectively. Both reactor loops include high efficiency cyclones separating the particles from the decarbonized flue gas and CO_2 rich stream leaving the carbonator and the calciner, respectively. The separated solids fall down in loop seals where a part is internally recirculated or transferred to the other reactor (from carbonator to calciner and vice versa). Both reactors are equipped with all conventional

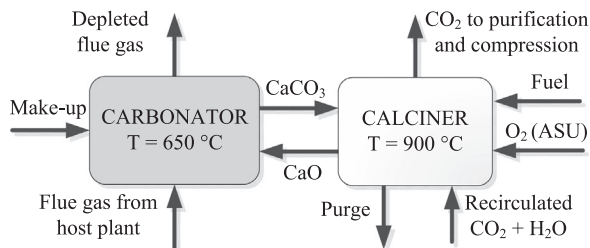


Fig. 1. Principle of the carbonate looping process.

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