



# Investigation of the fuel influence on the carbonate looping process in 1 MW<sub>th</sub> scale



Jochen Hilz\*, Martin Helbig, Martin Haaf, Alexander Daikeler, Jochen Ströhle, Bernd Epple

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

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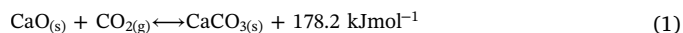
## ABSTRACT

The carbonate looping (CaL) process is an efficient post-combustion carbon capture technology to reduce the amount of CO<sub>2</sub> released by fossil fuelled power and industrial plants. The presented work discusses the results obtained in long-term pilot tests performed in a 1 MW<sub>th</sub> CaL pilot plant at Technische Universität Darmstadt. These long-term pilot tests aimed to improve the process and gain reliable experimental data to scale up the CaL process to industrial size. The type of fuel, sorbent, flue gas composition, reactor design, and operating conditions were varied to investigate long-term effects on the performance during pilot operation. Steady-state conditions were achieved while parameters were not changed during periods up to 60 h. The stability of the CaL process in semi-industrial size was proven by steady-state CO<sub>2</sub> absorption for more than 1200 h with interconnected circulating fluidized bed reactors with absorption rates in the carbonator higher than 90% and overall capture rates higher than 95%. A long-term pilot test is presented and the stability of hydrodynamics and temperature profiles in carbonator and calciner reactor are shown. The enrichment of inactive sorbent impurities like ash or calcium sulphate was observed and is dependent on the fuel type and fuel particle size in the calciner.

## 1. Introduction

The carbonate or calcium looping (CaL) technology is an efficient post-combustion carbon capture technology suited for retrofitting existing power and industrial plants to reduce the amount of CO<sub>2</sub> released by combustion of fossil fuels. The CaL technology was initially proposed by Shimizu et al. [1]. The principle process scheme is shown in Fig. 1.

The most developed CaL configuration is a system consisting of two interconnected circulating fluidized bed (CFB) reactors, a carbonator and a calciner. The process is based on the reversible carbonation-calcination reaction of limestone as described in Eq. (1).



In the carbonator, CO<sub>2</sub> contained in the flue gas from an emission source, e.g. an upstream power plant, reacts with the lime (CaO) and forms calcium carbonate (CaCO<sub>3</sub>) in an exothermic reaction at around 650 °C according to Eq. (1). The CO<sub>2</sub> absorption efficiency in the carbonator  $E_{\text{carb}}$  describes the flow of CO<sub>2</sub> absorbed related to the flow of CO<sub>2</sub> fed in the carbonator from an upstream emission source. The carbonator efficiency is a crucial process parameter and can be calculated as follows:

$$E_{\text{carb}} = \frac{F_{\text{CO}_2, \text{carb}, \text{in}} - F_{\text{CO}_2, \text{carb}, \text{out}}}{F_{\text{CO}_2, \text{carb}, \text{in}}} \quad (2)$$

The maximum carbonator efficiency for removing CO<sub>2</sub> from the gas flow entering the carbonator is limited by the minimum CO<sub>2</sub> volumetric concentration derived from the chemical equilibrium. It limits the actual carbonator efficiency at the current reactor temperature. The chemical equilibrium is a function of the reactor temperature and is given in Eq. (3) from Silcox et al. [2]:

$$\nu_{\text{CO}_2, \text{eq}} = 4.137 \times 10^7 e^{\frac{-20,474}{T}} \quad (3)$$

The CaCO<sub>3</sub> is transferred to the calciner where the temperature is increased up to around 900 °C to release the CO<sub>2</sub> bound in the solid phase in the endothermic backward reaction according to Eq. (1). The released CO<sub>2</sub> leaves the calciner in a highly concentrated gas stream and the CaO is recycled to the carbonator. The directly heated or oxy-fired CaL process is the basic concept where 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).

Besides the CO<sub>2</sub> fed in the carbonator ( $F_{\text{CO}_2, \text{carb}, \text{in}}$ ), CO<sub>2</sub> formed by the oxy-fuel combustion ( $F_{\text{CO}_2, \text{coal}}$ ) and released by calcination of fresh

*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](mailto:jochen.hilz@est.tu-darmstadt.de) (J. Hilz).

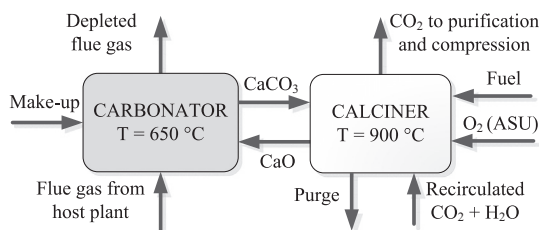
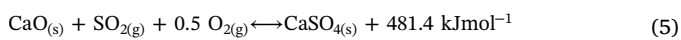


Fig. 1. Principle of the carbonate looping process.

limestone ( $F_{\text{CO}_2,0}$ ) have to be considered to describe the overall  $\text{CO}_2$  capture efficiency  $E_{\text{total}}$  of the CaL process. The total efficiency  $E_{\text{total}}$  is defined as the relationship between the captured  $\text{CO}_2$  at the calciner outlet ( $F_{\text{CO}_2,\text{calc},\text{out}}$ ) and total amount of  $\text{CO}_2$  introduced in the process as previously mentioned:

$$E_{\text{total}} = \frac{F_{\text{CO}_2,\text{calc},\text{out}}}{F_{\text{CO}_2,\text{carb},\text{in}} + F_{\text{CO}_2,0} + F_{\text{CO}_2,\text{coal}}} \quad (4)$$

A challenge of the CaL technology is a decreasing reactivity with increasing number of cycles of carbonation and calcination [3]. Therefore, a continuous feed of limestone as make-up is required in order to maintain a certain capacity of the sorbent to absorb  $\text{CO}_2$  [4]. In addition, different fractions of impurities, i.e. ash and calcium sulphate accumulate in the sorbent representing inactive material not reacting with  $\text{CO}_2$ . The ash enters the system with the coal and a fraction of the sorbent chemically deactivates by the reaction with sulphur ( $\text{SO}_2$ ) to calcium sulphate ( $\text{CaSO}_4$ ). The deactivated material cannot be regenerated below 1000 °C. The sulphur is mainly introduced in the process by the combustion of coal in the calciner and reacts under atmospheric conditions [5,6] as follows:



In comparison to other  $\text{CO}_2$  capture technologies, e.g. amine scrubbing (post-combustion), integrated gasification combined cycle (pre-combustion) or oxy-fuel combustion, CaL promises substantial advantages in terms of energy penalty [7–9] and costs [10–12]. Since both carbonation and calcination reactions run at very high temperatures, CaL offers additional power generation from the heat input required for the sorbent regeneration. The calciner adds an oxy-fired new power plant system to the existing upstream plant and the highly efficient heat recovery from different sources offered by the CaL process exhibit a great advantage to other  $\text{CO}_2$  post-combustion technologies.

The development of the oxy-fired CaL process is driven by the synergies between the most developed CaL process configuration of two interconnected CFB reactors and existing CFB reactor systems at industrial scale. The CaL technology has passed through a rapid development from concept to demonstration at industrially relevant environment (TRL6) during the last decade [13]. The feasibility of the CaL process has been demonstrated in various pilot plants, e.g. the 0.2  $\text{MW}_{\text{th}}$  pilot in Stuttgart [14], the 1  $\text{MW}_{\text{th}}$  pilot in Darmstadt [15], the 1.7  $\text{MW}_{\text{th}}$  pilot in La Pereda [16] or the 1.9  $\text{MW}_{\text{th}}$  pilot in Taiwan [17]. Besides the basic concept of the directly oxy-fired CaL process, various concepts of optimized CaL processes have been proposed utilizing indirect heat transfer from a combustor to the calciner [18–20]. The concept of the indirectly heated CaL process (IHCaL) based on heat pipes has recently been tested and proofed in 300  $\text{kW}_{\text{th}}$  scale [21].

Although the feasibility of the CaL process has been confirmed and the technology underwent a great progress, a realistic assessment of sorbent performance and accumulation of deactivated as well as inactive material requires long-term pilot operation. Therefore, it is necessary to achieve steady conditions of the sorbent phase by a homogeneous mixing of existing inventory and continuous make-up feed in extensive long-term operation.

It is important to conduct the pilot tests under realistic operating conditions with respect to the oxy-fuel combustion in the calciner. The

carbonation-calcination reaction is dependent on the calcination conditions. This implies the significant influence of the presence of high  $\text{CO}_2$  concentrations and water vapour on the calcination of the sorbent that was pointed out in various investigations [22–24]. These conditions are achieved by recirculating flue gas instead of mixing artificial calciner primary gas consisting of  $\text{O}_2$  and  $\text{CO}_2$ .

The long-term operation under realistic process conditions, e.g. homogeneous mixing of inventory and make-up as well as oxy-calcination under flue gas recirculation, is required to overcome the uncertainties of previous pilot tests as a basis for scale-up scenarios and to gain confidence about the performance of the CaL technology. 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 approach of achieving steady-state operation and evaluating these results in terms of carbonator performance by a modelling approach have recently been shown by Helbig et al. [25] and Hilz et al. [26]. This data guarantees a solid ground for the scale-up, design and engineering of a 20  $\text{MW}_{\text{th}}$  pilot plant.

This paper presents the results obtained from long-term pilot testing in the CaL pilot plant at Technische Universität Darmstadt. In addition to the results published by Helbig et al. [25] and Hilz et al. [26], this publication presents information about long-term pilot tests focusing on the stability of hydrodynamic and temperature profiles in the CFB reactors as well as the influence of fuel burnt in the calciner on the sorbent composition and particle size distribution (PSD). Different types of fuels with varying particle size distributions were fired in the calciner and results from continuous pilot operation of two out of four test campaigns are taken into account.

## 2. Experimental setup

A semi-industrial scale CaL pilot plant consisting of two interconnected CFB reactors and a combustion chamber with a thermal capacity of 1  $\text{MW}_{\text{th}}$  each is located at Technische Universität Darmstadt. Previous CaL tests in 1  $\text{MW}_{\text{th}}$  scale [15,27] were focused on proof of operation decarbonizing synthetic flue gas, a mixture of air and  $\text{CO}_2$ , and operating the calciner with oxygen enriched air. To operate with coal originated flue gas and to address the challenge of oxy-calcination, the pilot plant was upgraded to operate with coal originated flue gas from the furnace and oxy-combustion in the calciner.

Fig. 2 shows the scheme of upgraded 1  $\text{MW}_{\text{th}}$  CaL pilot plant. The CaL reactor system 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. High efficiency cyclones at both reactors separate the particles from the decarbonized flue gas and  $\text{CO}_2$  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 components of industrial CFB systems, i.e. refractory lining, start-up burners fired by propane, two-pass heat exchangers to cool down the off-gases, bag filters to remove entrained solid particles, water-cooled screw conveyors to drain off solids. To extract the released heat from the exothermic carbonation reaction and to operate at various reactor temperatures, the carbonator is equipped with five axially arranged internal cooling tubes. Flue gas provided by the combustion chamber is fed to the carbonator with a fan. A continuous mass flow of make-up to the carbonator is provided by a gravimetric dosing system. Air and oxy-fuel combustion is possible to be operated in the calciner with different types of coal, i.e. hard coal or lignite, in various particle size fractions from pulverized to coarse, sieved up to 10 mm. The coal is continuously fed by a gravimetric coal dosing system either in the return leg or in the bottom of the calciner. The air combustion is used for start-up to heat the reactor whereas oxy-firing is applied in regular CaL operation. The oxygen required for oxy-combustion is provided by a liquefied oxygen

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