

CO₂ CAPTURE FROM LIME AND CEMENT PLANTS USING AN INDIRECTLY HEATED CARBONATE LOOPING PROCESS – THE ANICA PROJECT

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Abstract

The overall aim of the ANICA project is to develop concepts of the indirectly heated carbonate looping (IHCaL) process for CO₂ capture from lime and cement plants. CO₂ avoidance costs of the IHCaL process for lime and cement production plants are expected to be in a well below 25 €/t, which is close to current CO₂ prices and significantly lower than competing CO₂ capture solutions for lime/cement plants. The novel process concepts are developed with the aid of advanced process simulations. The technology is demonstrated in a 300 kW_{th} pilot plant at industrially relevant conditions using the same fuels, sorbents, and operating conditions as can be expected in large-scale commercial IHCaL plants for lime and cement applications. A detailed techno-economic assessment and a life-cycle-analysis is performed for both lime and cement applications. The basic design of a 20 MW_{th} IHCaL demonstration plant is developed using two different technologies, i.e. fluidized bed reactors and Direct Separation technology, and costs of these plants are estimated. The paper presents an overview of the project as well as first results related to process simulations and design of the pilot plant.

Keywords: CO₂ capture; Lime plant; Cement plant; Calcium looping; Pilot plant

1. Introduction

Several different CO₂ capture processes are currently being developed, but most of them have the consequence of high energy consumption leading to lower plant efficiencies and increased costs [1]. Pre-combustion capture is not suitable for lime and cement processes since it is not possible to capture CO₂ that is released by calcination of raw material within the industrial process. These so-called process emissions amount to around 65 % of total CO₂ emissions of a cement plant. Oxyfuel combustion has the potential to capture the CO₂ from both the process emissions and the combustion emissions. In that case, a complete redesign of the cement production process is required, as well as the need for oxygen production. In order to avoid oxygen production, the process CO₂ can be captured using indirect heating of the cement calciner in a process called Direct Separation. However, the CO₂ from combustion of fuels is not captured, so that this process is limited to about 65% capture. Post-combustion capture using amine based solvents is a rather mature technology that can be used to retrofit lime and cement plant, but is associated with rather high energy requirements for regeneration of the solvent leading to high CO₂ avoidance costs [2]. The carbonate looping process (also named calcium looping) is another post-combustion capture technology using lime-based materials such as natural limestone as a solid sorbent. It offers synergies with the lime and cement industry through the possibility to use purged CaO for lime or cement production. Based on techno-economic assessment, CO₂ avoidance costs were determined for MEA absorption (80 €/tCO₂), oxyfuel process (44 €/tCO₂), chilled-ammonia process (66 €/tCO₂), membrane-assisted

CO₂ liquefaction (84 €/tCO₂), and calcium looping (52-55 €/tCO₂) [2]. Although CO₂ avoidance costs are lowest for oxyfuel technology, the calcium looping process can more easily be retrofitted to existing cement plants, since it has a lower impact on the cement production process.

The carbonate looping (CaL) process has the potential to significantly reduce the efficiency penalty compared to solvent-based technologies, since the process operates at high temperatures, which allows the utilization of heat for power production in a highly efficient steam cycle [3-5]. The CO₂ contained in the flue gas of an industrial plant is absorbed by CaO in the carbonator at around 650 °C [3, 6]. The CaCO₃ formed hereby is transferred to the calciner, where the CO₂ is released by an increase of temperature to around 900 °C. The stream of highly concentrated CO₂ is ready for further processing and storage/usage, and the regenerated CaO is transferred back to the calciner closing the solid loop. As the calcination reaction is endothermic, the calciner needs to be supplied with heat. The most straightforward heat supply is the direct combustion of fuel with oxygen in the calciner. For power applications, the efficiency penalty related to the oxygen demand of the directly fired calciner is around 3 %-points [3].

The efficiency penalty of the process can be further decreased when the need for technical oxygen in the plant can be avoided. This can be achieved by indirect heating of the calciner, e.g. through metallic walls, by solids circulation [3], or via heat pipes [7, 8]. Heat pipes offer an excellent heat transfer performance based on evaporation and condensation of a liquid (i.e. sodium for temperatures >800 °C) inside a closed pipe and have been successfully applied to an indirectly heated

gasification process. For CO₂ capture from coal-fired power plants, the net efficiency of the IHCaL process is around 1.5 % points higher than that of the standard CaL, [7, 9]. The CO₂ avoidance costs have been calculated to 22.6 €/t_{CO2} excluding CO₂ storage [9].

The feasibility of the IHCaL process using heat pipes was demonstrated in a 300 kW_{th} pilot plant during more than 300 hours of stable CO₂ capture at various operating points [10, 11]. Furthermore, a large-scale heat pipe with a length of 6 m and an outer diameter of 48.3 mm demonstrated the possibility of an up-scale during long-term performance tests for a total run-time of more than 1500 hours at University of Erlangen-Nürnberg.

Concepts of the IHCaL process for CO₂ capture from lime and cement plants are currently being investigated in the ACT project ANICA (Advanced Indirectly Heated Carbonate Looping Process). This paper will present the process concept, an overview of the ANICA project as well as first results related to process simulations and design of the pilot plant.

2. Process Concept

The main innovation of the IHCaL process is the use of an indirectly heated calciner for regeneration of the CaL sorbent. The heat for the calcination is produced by the combustion of fuel with air in an additional external combustion chamber and transferred to the calciner by means of heat pipes. The flue gas of the external combustion chamber is directed to the carbonator, where most of the CO₂ contained in this flue gas is absorbed by CaO. The main advantages of the IHCaL process compared to the standard CaL process are summarized as follows:

- No air separation unit is needed to produce technical pure oxygen, which leads to lower investment costs and to a lower energy consumption.
- Fewer impurities (sulphur, ash) from a supplementary firing are brought into the Ca-loop, so that spent sorbent will be of higher purity and therefore be better suited for further utilization.
- Lower CaO deactivation rates are expected due to “mild” calcination around the heat pipe surfaces compared to rather harsh conditions in an oxy-fired calciner, so that sorbent remains more reactive.
- Lower attrition rates are expected due to a low fluidization velocity in the calciner, which improves the operability of the fluidized bed system.
- An almost pure CO₂ stream leaves the calciner, which allows for technically easy and cost-effective CO₂ purification process for compression and storage/utilization of CO₂.

So far, the IHCaL process has solely been evaluated with respect to CO₂ capture from power plants. This section discusses novel technical concepts for integrating the IHCaL process into lime and cement production plants.

2.1 Integration of IHCaL into a lime plant

The integration of the IHCaL concept into the lime production process can be realized either as a tail-end solution in an existing lime plant (placed after the kiln and capturing the CO₂ of the flue gas) or as an integrated solution, as illustrated in Figure 1.

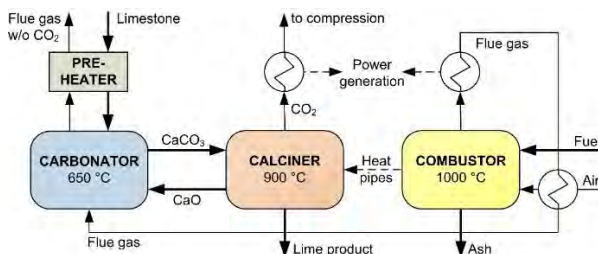


Figure 1: Concept of an IHCaL process integrated into a lime plant

Raw material (limestone) is used as sorbent for the IHCaL process, and the purge from the IHCaL process consists of CaO, which is the main product. The heat of the flue gas leaving the carbonator or calciner could be used to pre-heat the raw material. The heat of the remaining flue gases can be extracted in heat recovery steam generators for power generation using a highly efficient steam cycle as well as for pre-heating the combustion air. The produced electricity can be partially used on site for various consumers (such as fans, pumps, mills, electrolysis for CO₂ utilization) and partially sent to the grid, generating additional value/income for the plant operator. This concept results in a completely new lime production process that has various advantages compared to conventional lime kilns (such as shaft furnaces or rotary kilns):

- The fuel is no longer in direct contact with the lime, which improves the purity of the lime and allows the utilization of “dirty” but cheap fuels, such as waste-derived fuels.
- The temperature in the calciner is uniform and well-controlled, which enables to meet precise product specifications.
- A smaller particle size is used (0.1 – 0.5 mm) compared to shaft furnaces, which reduces the calcination time, intensifying production and minimizing inhomogeneities within the particle mixture.

2.2 Integration of IHCaL into a cement plant

The integration of IHCaL into the cement process can be realized either as a tail-end solution or as an integrated solution using the raw meal as sorbent. A tail-end solution is placed at the back-end of the cement plant capturing CO₂ from the fuel gas before sending it to the stack. This option could easily be retrofitted to existing cement plants. However, a drawback of the tail-end solution is that the process CO₂ is released twice, and the solids mass flows in the IHCaL reactor exceed that of the cement plant, leading to rather huge plant sizes. Although the energy penalty is low since the additional fuel introduced into the combustor is efficiently converted to power, the economical performance is questionable due

to the rather high investment and high additional fuel consumption considering the strongly varying revenues of the electricity market.

The integrated solution could offer significant benefits compared to the tail-end variant, as calcium carbonate is a main constituent of cement clinker raw materials, and the kiln system already includes a calciner. One possibility for such an integrated solution is illustrated in Figure 2. Raw materials for clinker production (mainly limestone) are used as sorbent for the IHCaL process, and the purge from the IHCaL process (mainly CaO and other oxides from silica, alumina, and iron) is fed to the cement kiln. Hence, the calciner of the IHCaL unit replaces the pre-calciner of a conventional cement plant. The flue gas leaving the carbonator (and/or calciner) is used to pre-heat the raw materials, potentially using existing cyclone arrangement of the cement plant. Excess heat of the calciner/combustor is extracted in heat recovery steam generators for power generation in a highly efficient steam cycle (potentially utilizing parts of an already existing power plant on the cement production site) and for pre-heating the combustion air. The produced electricity can be partially used on site for various consumers (such as fans, pumps, mills, electrolysis for CO₂ utilization) and partially sent to the grid, generating additional value/income for the plant operator.

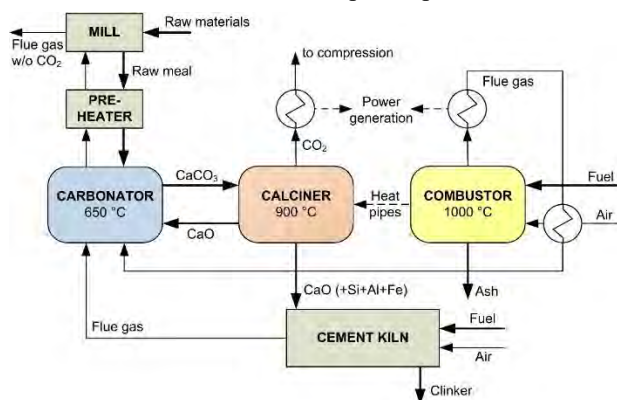


Figure 2: Concept of an IHCaL process integrated into a cement plant

However, there are some technical challenges related to the integrated solution. The use of cement raw meal instead of limestone as sorbent may cause difficulties, as these contain other components like silica, alumina and iron to form the mineral clinker phases, which may have a negative impact on the activity of the sorbent for CO₂ absorption. Furthermore, the rather small particle size and stickiness of the raw meal may impose challenges regarding fluidization characteristics (Geldart A/C particles) and fouling of heat pipes in a bubbling fluidized bed. The risks related to fluidization and fouling could be reduced by various means. One option is to replace the bubbling bed calciner by an entrained flow calciner. However, an entrained flow reactor is much larger than a fluidized bed, and heat transfer rates to heat pipes are much lower due to the low solids loading.

Fluidized beds are generally well suited for conversion of fuels that are difficult to exploit as an energy source and complicated in terms of handling and logistics, such as

solid waste. Although fluidized beds are used in IHCaL technology, using such fuels has never been tested with immersed heat pipes. The ANICA project will address this issue by pilot tests using waste derived fuels to provide proof of concept prior to commercial application.

2.3 Novel concepts of the IHCaL reactor system

One major challenge of the IHCaL process is the high heat duty of the calciner since the flue gases from the external air-fired combustor are additionally fed to the carbonator. Furthermore, the large number of required heat pipes may lead to a rather large size of the calciner. The following options are investigated in the ANICA project to counterbalance these effects in terms of CAPEX and OPEX:

- An improved heat pipe performance reducing the number and/or size of the heat pipes
- A solid/solid heat exchanger transferring heat from the “hot” solids leaving the calciner to the “cold” solids coming from the carbonator, so that less heat is required in the calciner to heat up the incoming solids to 900 °C
- A two-stage calciner that lowers the temperature of the solids exiting the calciner by a dilution with steam in the 2nd stage, aiming at reduced heat demand of the calciner.

3. The ANICA project

The ANICA (Advanced Indirectly Heated Carbonate Looping Process) project is a collaboration between various academic and industrial partners from Germany, the United Kingdom, and Greece, funded by national authorities within the funding program ACT (Accelerating CCS Technologies). It started in October 2019 and lasts for three years. This section gives a brief overview of the objectives and the work program of the ANICA project.

3.1 Objectives

The overall aim of the project is to develop a novel technology with very low energy penalty and costs using lime-based sorbents – namely the indirectly heated carbonate looping (IHCaL) process – for CO₂ capture from lime and cement plants. The specific objectives of the project are:

- 1) To decrease the costs for CO₂ capture from lime and cement plants below 25 €/t_{CO2} by developing novel IHCaL process concepts with > 90 % CO₂ capture efficiency, > 95 % CO₂ purity, and > 45 % net electrical efficiency for heat utilization.
- 2) To aim at net negative CO₂ emissions by utilizing waste derived fuels with a high biogenic fraction.
- 3) To achieve more than 90 % utilization of sorbent as raw material for lime and cement production employing a high level of integration of heat and material streams.

- 4) To reduce energy requirements and equipment costs by 30 % compared with current design through development of new reactor concepts.
- 5) To demonstrate IHCaL technology in relevant environment (i.e. TRL 6) by long-term pilot tests at 300 kW_{th} scale under realistic conditions (i.e. fuels, sorbents, and operating conditions) for lime and cement applications, thereby proving the long-term performance/stability of sorbent and heat pipes.
- 6) To enable fast and reliable scale-up of the technology by developing accurate 1D and 3D models of the dual fluidized bed reactor system with an uncertainty of less than 10 %.
- 7) To provide a basis for comparing the IHCaL process with competitive CO₂ capture solutions (amine wash, oxyfuel combustion) for lime and cement plants by evaluating risks, economic performance, and environmental impact of the full-scale IHCaL process.
- 8) To accelerate the deployment of IHCaL technology by providing the basic design, plant layout and a cost estimation of a semi-industrial IHCaL demonstration plant at 20 MW_{th} scale on a cement production site, which could bring the technology to the next level of maturity (i.e. TRL 7) in a follow-up project.

3.2 Work program

This section briefly summarizes the work program of the ANICA project. Concepts for integrating IHCaL into lime and cement plants (see Sections 2.1 and 2.2) are developed in two steps. In a 1st step, these process concepts are based on existing knowledge/models and are used to define the operating conditions for pilot tests. The experimental data of these pilot tests are used for validation of 1D and 3D models for the IHCaL reactors. The validated models and novel concepts for optimising the IHCaL reactor systems (see Section 2.3) are used to further update the process concepts for integrating IHCaL in a 2nd step. The updated heat & mass balances are used for the assessment of these process concepts with respect to risks, techno-economics, environmental impact, and societal readiness. The process and reactor concepts are further used for the design of a 20 MW_{th} demonstration plant using two different technologies, i.e. fluidized bed technology and Direct Separation Technology.

4. Results and Discussion

Around one year after the start of the ANICA project, most of the tasks are still on-going or have not started yet. This section presents selected preliminary results related to simulations of integrating the IHCaL process into lime plants and preparations for pilot testing.

4.1 Process simulations

Two concepts for integrating the IHCaL process into an existing lime plant in Germany, which uses a preheated rotary kiln (PRK) to burn limestone, have been

investigated by means of process simulations. With the PRK, more than 600 tons of lime are produced per day in normal operating conditions. One concept is a tail-end solution in an existing lime plant placed after the kiln and capturing the CO₂ of the flue gas. The other is an integrated solution in which the lime production and the carbon capture are realized within the IHCaL facility.

A steady state model in ASPEN PLUS™ has been developed for the process simulations. The rotary kiln is modelled using three reactor blocks. To model coal combustion, the fuel is first inserted into a yield reactor that decomposes it into the elementary molecules and heat. Afterwards, the resulting stream is burned with the combustion air in a Gibbs reactor, which minimizes the Gibbs' free energy in order to calculate the heat production and the yield of products. For all combustion processes, an air-fuel equivalence ratio (λ) of 1.2 is considered according to the reference plant. The temperature of the IHCaL combustor is set to 1000 °C to allow for 100 K temperature difference to the calciner. The calcination and carbonation reactions in the IHCaL reactors are modelled with conversion reactor blocks, where the reactions take place at specified conversion rates according to previous models of the research group. The solid-gas separation in the cyclones is considered ideal. The fuel and raw material composition is defined according to the reference plant. The same lignite (LHV=21,500 kJ/kg) used to fire the PRK is implemented as fuel for the combustor in the IHCaL. Similarly, the limestone's composition from the reference plant (98.3 wt% CaCO₃) is used for all the limestone inputs in the model.

In order to evaluate the solutions proposed, the following key performance indicators are considered. The CO₂ capture efficiency, defined as the ratio of the captured CO₂ to the generated CO₂, is kept constant at 90 %. The product ratio, *PR*, considers the production of the entire process including the IHCaL unit in relation to the production in the reference plant. The heat ratio, *HR*, is used as the indicator of the heat requirement for CO₂ capture and lime production. It is calculated considering the lime produced, the heat requirement in the original process as well as in the entire process including CO₂ capture. For the power generation from the high temperature heat of the IHCaL, a heat-to-power efficiency of 45 % is assumed in this work, according to values of thermal power plants. For the CO₂ balance, the direct CO₂ emissions are calculated by the amount of CO₂ directly emitted into the atmosphere from the complete process per unit of lime produced.

With respect to the boundary conditions, the operating temperature of the calciner is set at 900 °C to enable a full calcination at nearly pure CO₂ atmosphere. The operation temperature of the carbonator is set to 650°C in order to achieve a maximum capture efficiency of around 90 %. The make-up flow, which is needed to avoid the build-up of inert species and to maintain the proper activity of the sorbent, is characterized with the make-up ratio, *A*, i.e. the ratio of the molar flow rate of make-up calcium species to the total molar flow rate of CO₂. For the tail-end concept, the make-up ratio was set to 0.2. For

the fully integrated solution, the make-up flow is determined by the raw material input that is considered the same as for the reference plant to have the same production. The specific sorbent circulation rate considering the molar flow rate of calcium species that are transferred from the calciner to the carbonator was set

the integrated concept is very similar to the reference plant because the raw material input is the same for both.

The direct fuel consumption of the reference PRK is 5090 MJ/t_{CaO}. The tail-end solution leads to a 154 % increase of the direct fuel consumption and a 70.5 % reduction of the direct CO₂ emissions with respect to the

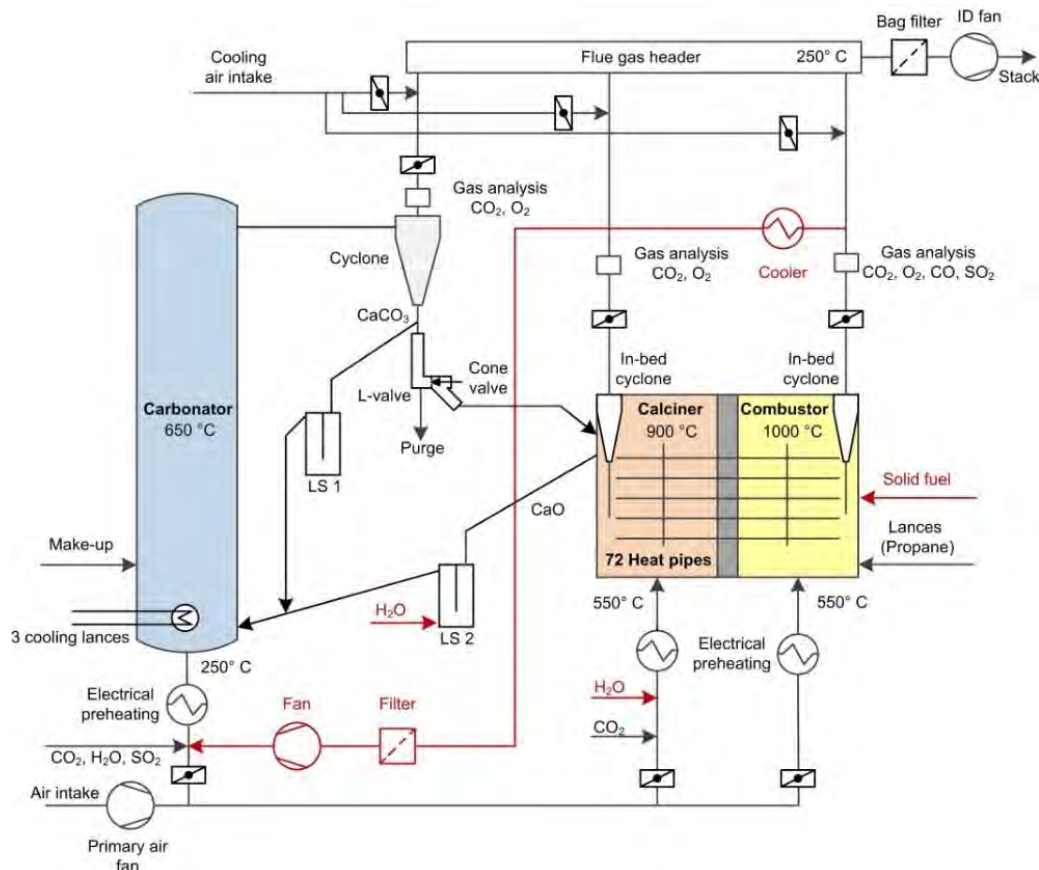


Figure 3: Process scheme of the 300 kW_{th} pilot plant with adaptions marked in red color.

to 6.

The main results of the base case process simulations are summarized in Table 1. The specific make-up ratio controls the production from the IHCaL facility in the case of the tail-end solution. While it may be more reasonable to keep this production low, it is necessary to maintain a certain make-up. Due to the high value of A for the integration solution, a highly reactive solid inventory can be expected, which could allow for a reduction of the solid circulation rate and/or solid inventory in the carbonator.

Table 1: Results of base case simulations

	Tail-end concept	Integrated concept
Specific make-up ratio (A)	0.20	0.47
Product ratio (PR)	2.26	1.01
Heat ratio (HR)	2.54	1.63

The product ratio is as high as 2.26 in the tail-end concept, which means that more lime is being produced in the IHCaL facility than in the PRK. The production of

reference plant. For the fully-integrated case, the increase in fuel consumption is only 63 %, and the reduction in direct CO₂ emissions is 87.4 %. For this case, the electricity generated through heat recovery amounts to 29.6 % of the total thermal energy input. This implies a further reduction in the net CO₂ emissions, considering the avoidance of the CO₂ from the grid's power generation. Furthermore, the combustor can be adapted to burn waste-derived fuels with a high biogenic content, which would allow for negative CO₂ emissions.

A more detailed assessment of the results and a sensitivity study is included in a publication of Greco-Coppi et al. [12].

4.2 Pilot plant design

One aim of the ANICA project is the demonstration of the IHCaL process for integration into lime and cement plants by pilot tests in a 300 kW_{th} pilot plant under real conditions, with the same fuels, sorbents, and operating conditions as specified in the process concepts for these plants. Coal and solid recovered fuel (SRF) are used as fuels, while limestone and cement raw meal are used as sorbents. This section describes the existing pilot plant and the adaptions that are currently being installed.

The design and experimental results of the existing 300 kW_{th} pilot plant (see Figure 3) were published by Reitz et al. [11]. The plant consists of the heat-pipe heat exchanger connecting the calciner and the combustor, a carbonator with a cyclone separator, loop seals, and a combination of a L-valve and a cone-valve for the coupling of the carbonator with the calciner. The pilot plant contains 72 heat pipes with an outer diameter of 33.7 mm and a length of 2.18 m. The carbonator operates as a circulating-fluidized bed (CFB) reactor. The combustor and calciner are bubbling fluidized bed (BFB) reactors. The dimensions of the three reactors are listed in Table 2.

Table 2: Dimensions of the pilot plant

	Unit	Height	Length x Depth
Carbonator (CFB)	m	8	0.25 (diameter)
Calciner (BFB)	m	2.3	1.05 x 0.3
Combustor (BFB)	m	2.3	1.05 x 0.3

The plant has previously been operated using an artificial flue gas (i.e. air mixed with CO₂, water seam, SO₂) entering the carbonator and propane as fuel in the combustor. Hence, the existing pilot plant has to be upgraded to enable realistic conditions for lime and cement applications. This includes the design and installation of a solid fuel feeding system for the combustor as well as a flue gas path (ducts, heat exchanger, filter, fan, and flow control) from combustor to carbonator. A flow sheet of the process is shown in the Figure 3, with the main upgrades depicted in red.

In order to feed the combustor with solid fuel such as SRF or coal, the pilot plant is expanded by a complete solid feeding and dosing system. The solid fuel is filled into a receiver tank. A gas-sealed flap ensures that no hot gases are leaving the system. Another container is connected to a loss-in-weight feeder to control the mass flow. A rotary valve has the function of a pressure seal. The solids are fed into the dense bed of the combustor by a screw feeder. For ash discharge, a discontinuous sluice system at the bottom of the combustor will be installed.

The flue gas from the combustor needs to be cooled and dedusted before entering the carbonator. After leaving the cyclone of the combustor, the flue gas passes an existing control valve, aiming to control the pressure in the combustor. The gas passes a sensor that analyses the gas composition in terms of CO, O₂, CO₂, SO₂, H₂O, and HCl. Afterwards, the flue gas is divided into a portion leaving the reactor system via the air quench through the chimney and a portion transferred to the carbonator. This flow is controlled by an additional control valve. Then, the flue gas is cooled down in the flue gas heat exchanger to around 200 °C, located vertically in the flue gas path behind the combustor. After the cooler, a bag filter is installed in order to separate the fine dust particles from the flue gas flow and protect the downstream equipment. Finally, a fan is required to compensate the pressure difference between combustor outlet and carbonator inlet as well as the pressure losses in the flue gas path.

Besides the above-mentioned major upgrades, some minor modifications are integrated in order to improve

the operability of the test rig. The existing make-up dosing is modified in order to feed a higher amount of solid material into the carbonator. A solid sampling system consisting of a conveyor is installed in the loop seal between carbonator and calciner in order to deliver enough solid samples for the investigation related to usage in cement or lime plants. All adaptations of the pilot plant are expected to be finalized in summer 2021, so that the pilot tests can be performed in the same year.

The pilot tests will be used to evaluate various key performance indicators of the IHCaL process. The CO₂ capture efficiency will be determined by measurements of the CO₂ concentrations and volume flows leaving the reactor system. The CO₂ product quality will be assessed by measuring the gas composition of the gas leaving the calciner. Furthermore, the purged sorbent will be analysed to evaluate the quality of the solid with respect to its usage in the clinker burning process for cement production or as lime product. Process models will be validated by the pilot data and then scaled to industrial size in order to calculate the key performance indicators of a real industrial process.

5. Conclusions

In the ANICA project, novel concepts for integrating the IHCaL process into lime and cement plants for CO₂ capture are being developed. These concepts have a high potential for reducing the energy penalty and costs compared to competing CO₂ capture technologies. Preliminary results of process simulations show that a highly integrated IHCaL process for lime production has significantly less heat requirements compared to a tail-end solution. The feasibility of the IHCaL process for lime and cement applications will be tested in a 300 kW_{th} pilot plant that has been redesigned to enable realistic conditions close to commercial applications. The experimental results of these pilot tests will be used to validate models that will then be applied to design demonstration and industrial plants. Based on the heat and mass balances of the industrial plants, a techno-economic evaluation will be performed to determine the lime/cement production and CO₂ avoidance costs of this technology. Assuming that the construction of a demonstrator will start after this project in 2023, the first commercial plant could go into operation around the year 2028.

Acknowledgements

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