# DESCRIPTION OF THE WORK AND PRELIMINARY RESULTS OF THE AC2OCEM PROJECT IN FACILITATING CARBON CAPTURE TECHNOLOGY IN THE CEMENT INDUSTRY USING OXYFUEL COMBUSTION

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## Abstract

The Paris agreement compels $CO_2$ intensive industries, such as the cement industry to significantly decrease the $CO_2$ emissions. The source of the $CO_2$ emissions from the cement industry is related to the calcination and the combustion process, which is why innovative measures have to be implied to reach the $CO_2$ reduction goal. According to previous studies, the oxyfuel technology to facilitate the carbon capture and storage technology is the most economically feasible potential to solve the issue. In frame of the AC²OCem project, a European consortium has come together to reduce the knowledge gap and the time to market of the oxyfuel technology in the cement industry. Experimental and analytical work on the oxyfuel technology for retrofitting existing cement plants will be focused on optimizing the calciner and the oxyfuel burner with 100 % alternative fuels. In addition to the experimental and analytical tests regarding the burner technology in pilot-scale facilities, design and optimization of retrofitting existing plants is performed, taking into consideration the boundary condition of two demonstration plants located in Slite, Sweden and Lägerdorf, Germany. For the retrofit design, the heat integration in oxyfuel condition and the effect of false air ingress for both plants is studied and it is found that a heat exchanger system would adequately supply the energy to the drying unit. A realistic value for false air has been set at 6 % as an initial design parameter of the CPU. Within the scope of the AC²OCem project, the kiln and calciner oxyfuel burners will be promoted to higher technology readiness levels for newly-build and up-to 100 % oxygen combustion cement plants. A techno-economic analysis and life cycle assessment for retrofitted existing and new-build cement plants will also be performed.

***Keywords:*** *Alternative fuel, CCS, Cement, Oxyfuel, SRF*

## 1. Introduction

The ambitious target set by the Paris agreement, 2 °C scenario by 2050, necessitates overall around 2,800 Mt $CO_2$ emissions reduction. The International Energy Agency (IEA) has emphasized that 40 % of the decrease in emissions must be made within the European industry, specifically those that are $CO_2$ intensive, such as the cement and steel industries [1]. European countries taking part in the Paris agreement have come up with several approaches to reduce the $CO_2$ emissions by 1,700 Mt. Further measures must be taken to account for the remaining 1,100 Mt of $CO_2$. For this purpose, industry and research institutions are working together to come up with technically and economically feasible methods to reduce and ultimately eliminate these emissions [2]. Both the IEA and the International Renewable Energy Agency (IRENA) have confirmed that carbon capture usage and storage (CCUS) is important to achieve the goal set by the Paris agreement [3,4].

### *1.1 Cement Industry*

The cement industry in one of the highest energy and $CO_2$ intensive industries, where $CO_2$ is not only emitted due to the combustion process but also as the main side product of the calcination process. The process related emissions are directly proportional to the amount of clinker used, the main raw material in the cement industry. The important $CO_2$ reduction approaches in the cement industry include switching to low-carbon or carbon-free fuels, reducing the clinker to cement ratio and advancing the process technology [3,5,6].

The IEA has shown that these approaches can potentially achieve 44 % of the target $CO_2$ emission reduction [6]. In order to achieve the decarbonization of the sector and reach the 2 °C scenario goal, further measures have to be taken into account. In frame of the European Cement Research Academy (ECRA) project, the potential of carbon capture and storage (CCS) is studied and the advantages and disadvantages of the different carbon capture methods in the cement industry, pre-combustion, post-combustion, oxyfuel technology and carbonate looping were assessed. It was concluded that the oxyfuel technology and post combustion capture showed high potential [7]. In the CEMCAP project, the oxyfuel technology, chilled ammonia-based absorption, membrane-assisted liquefaction and calcium looping were assessed and compared with MEA-based absorption. Among these technologies, the oxyfuel technology was found to have the lowest cost of $CO_2$ avoided, but at the same time a high risk related to the close integration with the clinker burning process was highlighted for this technology [8,9].

### *1.2 AC²OCem Project*

Eleven partners from five European countries, Norway, Germany, Switzerland, France and Greece have formed a consortium in scope of the ACT program to evaluate the feasibility of oxyfuel combustion in the cement industry to accelerate the carbon capture and storage technology. The AC²OCem project brings together the expertise of four industrial end-users, two technology providers and five research institutions. In scope of AC²OCem, oxyfuel technology will be evaluated for retrofitting existing cement plants and for new-build plants. This paper will provide a comprehensive overview of the project goals and objectives, in addition to a brief summary of achieved results and an outlook on planned work.

The main goal of the AC²OCem project is to accelerate the deployment of CCS by closing the knowledge gaps and advancing the technology development for a successful operation of the first oxyfuel demonstration cement plants. The project will focus on reducing the $CO_2$ avoidance cost, and increasing the plant efficiency.

The project consists of six work packages, WP1 is for management and dissemination. Within WP2, advanced oxyfuel burners, pilot-scale experiments as well as CFD simulations will be performed to evaluate the oxyfuel kiln burner in retrofitted and new-build plants with up to 100 % alternative fuels. WP3, optimization of oxyfuel calciner, is focused on evaluating the impact of the flue gas composition on the calcination process under oxyfuel conditions, and demonstrating pilot-scale calcination tests in oxyfuel conditions using up to 100 % alternative fuels. In WP4, integration of 1st generation oxyfuel technology by retrofit to existing cement plants, Heidelberg Cement and LafargeHolcim have selected two demonstration plants in Slite, Sweden and Lägerdorf, Germany respectively to design and optimize the oxyfuel technology, transferring the technology from TRL6 to TRL8. Different flue gas recirculation ratios and raw meal qualities will be simulated to optimize the overall process and a techno-economic analysis will be performed for retrofitted cement plants (considering the two demonstration plants). In WP5, oxyfuel technology of 2nd generation for new-build cement plants, the design and optimization of the new-build oxyfuel cement plants will be evaluated, promoting the technology from TRL2 to TRL6 and a techno-economic feasibility study will be performed. In WP6, life cycle assessment, the environmental sustainability aspects of oxyfuel technologies for retrofitted and new-build cement plants will be evaluated.

## 2. Oxyfuel burner technologies

The focus of this work package is on the experimental and analytical investigation of an oxyfuel burner. The first part of the work package is dedicated to the design and evaluation, experimental and analytical, of an oxyfuel burner for 100 % alternative fuel in oxyfuel atmosphere. The second part of the work package is dedicated to the design and evaluation of an advanced oxyfuel burner for the combustion of alternative fuels with up to 100 % oxygen. The experiments will take place in the 500 kW pilot-scale combustion chamber at the University of Stuttgart that has been adapted to simulate a cement kiln. The experimental data obtained will also be used to validate the CFD simulation models of the retrofitted and new-build cases performed by CERTH and SINTEF respectively.

### *2.1 Description of the 500 kW combustion facility at the University of Stuttgart*

The 500 kW pilot-scale down-fired combustion chamber at the University of Stuttgart (Figure 1) has been optimized to simulate the atmosphere inside a cement kiln by an advantageously designed burner muffle that electrically pre-heats the secondary inlet gas up to 800 °C to account for the recirculated combustion gases from the clinker cooler [10]. The combustion chamber is 7 m in length and 0.8 m in inner diameter and measurement ports are distributed along the length of the chamber to allow axial and radial in-flame measurements.

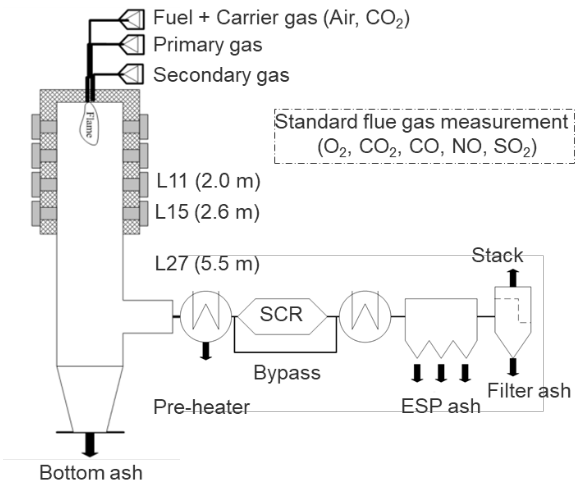


Figure 1: Sketch of 500 kW combustion chamber at IFK, University of Stuttgart

### *2.2 100 % SRF combustion experiments*

Three different cases were performed, the first is a reference case with air and 100 % SRF (refer to Table 1 for the SRF properties, taking into account that SRF is an inhomogeneous fuel and the results are bound to have high uncertainties). In the second case, AIR28, the carrier, primary and secondary gases were air, yet an additional oxygen stream was fed into the primary gas inlet along with air, the total oxygen concentration in the inlet gas was 28 vol. %. The third case, OXY31, is an oxyfuel case with 100 % SRF, 31 vol. % total oxygen in the inlet gases and $CO_2$ for the rest of the combustion gases.

Table 1**:** SRF properties including the proximate and ultimate raw fuel analysis

| | Unit | a.r. |
|---|---|---|
| Moisture | wt. % | 2.03 |
| Volatile matter | wt. % | 79.4 |
| Ash | wt. % | 11.0 |
| Fixed Carbon | wt. % | 7.62 |
| Carbon (C) | wt. % | 56.8 |
| Hydrogen total (H) | wt. % | 8.19 |
| Hydrogen organic (H) | wt. % | 7.96 |
| Nitrogen (N) | wt. % | 0.653 |
| Sulfur (S) | wt. % | 0.276 |
| Net calorific value $H_u$ | J/g | 24818 |

The gas composition in the primary, secondary and carrier gas can be controlled independently of each other. In the AIR28 case, pure oxygen is fed directly into the primary air inlet stream at the burner resulting in 60 vol. % oxygen in the primary gas. The secondary and carrier gases are air. In the OXY31 case, the SRF carrier gas is $CO_2$, the primary and secondary gases contain 70 vol. % and 22 vol. % oxygen respectively. The AIR28 and OXY31 case are both evaluated based on the reference air case. In the oxyfuel case the gas radiation and heat capacity are the properties with the highest effect on the combustion process. At 1123 °C, the molar heat capacity ratio of $CO_2$ to $N_2$ is 1.7, which directly influences the flame temperature and stability [11]. Figure 2 depicts images of the SRF flame at 18, 33 and 48 cm from the burner, it is observed that at higher oxygen concentrations, in the primary gas, the flame is more compact near the burner. To achieve a stable flame and maintain the flame temperature, for coal, the total oxygen concentration must be between 25 and 42 vol. % [11]. Other researchers have shown that the type of fuel influences the total oxygen concentration necessary to maintain a similar temperature profile during oxyfuel combustion with $CO_2$ in comparison to the air reference case [12,13].

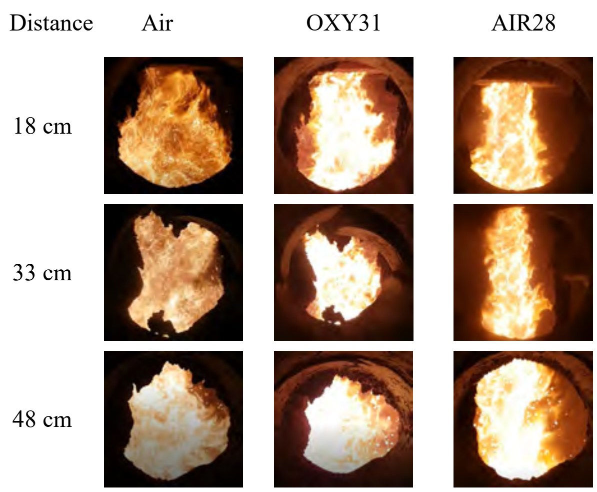


Figure 2: 100 % SRF flame captured in the Air, OXY31 and AIR28 cases at different distances from the burner

Figure 3 depicts the temperature and oxygen concentration profiles measured from the burner along the centerline of the furnace. The temperature profiles up to 48 cm are similar for the three cases despite the fluctuation of the oxygen consumption in the region. It is observed that the rate at which the temperature increases from the burner (0 cm) to the maximum measured flame temperature (measured at 116 cm from the burner for the Air and OXY31 cases and at 99 cm for the AIR28 case) is higher in the AIR28 case compared to the air and OXY31 cases. The temperature profile in the OXY31 case is on average 40 °C less than in the reference case, yet the oxygen consumption, especially in the main flame zone (between 63 and 167 cm), reacts similarly in the two cases. Higher fluctuations in the oxygen consumption is also noticed in the OXY31 case. The experiments have proven a stable SRF flame, yet one possibility to increase the flame temperature above 1200 °C would be a mixture of different alternative fuels, for example one with less volatile matter.

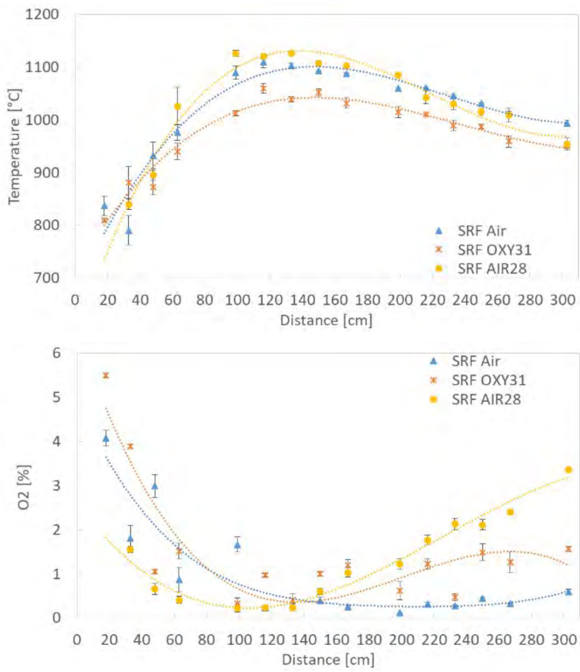


Figure 3: Oxygen concentration and temperature profiles measured from the burner along the centerline of the furnace

Figure 4 depicts the peak temperature measured in the three cases and their relative calculated adiabatic flame temperatures. Although the adiabatic temperature is over 1000 °C above the measured temperature, the trend is clear and confirms the results obtained. The adiabatic temperature and measured temperature of the OXY31 case were lower than that of the reference air case by 49 and 33 °C respectively. This result complies with the results of other researchers and suggests that the total inlet oxygen must be slightly increased to become comparable with the air reference case.

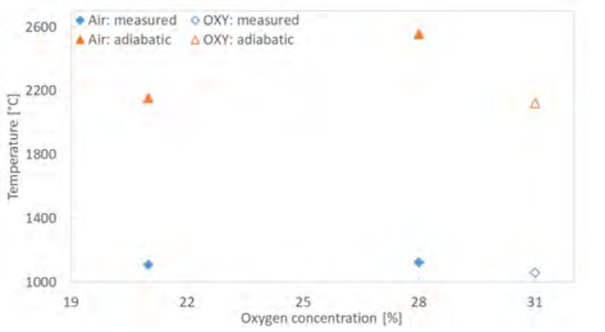


Figure 4: Adiabatic temperature calculation and maximum measured temperatures for 100 % SRF combustion with air, AIR28 and OXY31

These and future measurements will be used to verify CFD simulation performed by CERTH and SINTEF. Further measurements with higher oxygen concentration are also planned in the frame of the AC[2]OCem project.

# 3. Impact of oxyfuel technology on the calciner

The focus of work package 3 is on experimental investigations of calcination under oxyfuel conditions.

The studies will be conducted in three experimental facilities at the University of Stuttgart, thyssenkrupp Industrial Solutions AG and Air Liquide.

These experiments have been partially started and will be continued in the next months.

Several objectives are being investigated in each facility including the role of flue gas moisture level in the calciner to reduce and control the calcination temperature and minimize the risk of blockages under oxyfuel conditions as well as investigations on different oxyfuel calciner regimes. Theoretical and experimental calcination tests will be performed up to pilot-scale in a process relevant environment (TRL 6). To correlate the calcination performance in the oxyfuel environment to a reference scenario, the tests will be conducted under both air and oxyfuel conditions.

The impact of process conditions and flue gas impurities like sulfur and chlorines on calcination reaction will be evaluated at technical- and pilot-scales. Based on the results a moisture injection concept and process control strategy for retrofitted and new-build cement plants will be developed.

The experiments at the technical scale facility in the University of Stuttgart will be performed in an electrically heated entrained flow reactor (up to 50 kW), refer to Figure 5. The facility is capable of emulating co-current oxyfuel calciner conditions, which allows promising investigations under the relevant environment.

The pilot-scale facility of thyssenkrupp Industrial Solutions AG, Figure 6, consists of a calciner and a 4-stage cyclone preheater with the feed rate of around 40 kg/h. The influence of moisture content on the calcination efficiency and the calcination temperature will be fully investigated in oxyfuel environments.

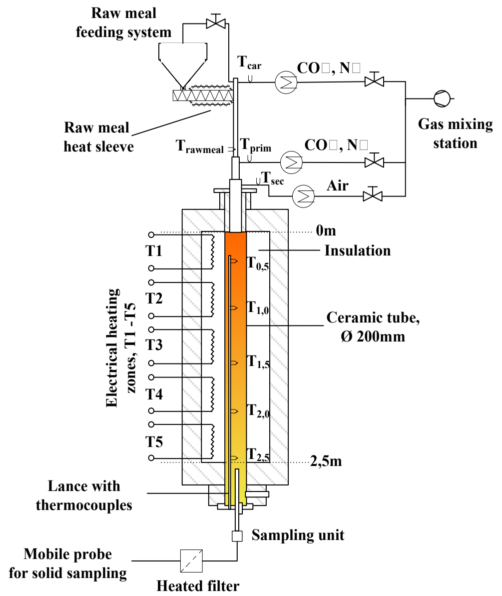


Figure 5: Sketch of the 50 kW technical-scale facility at the University of Stuttgart, Germany

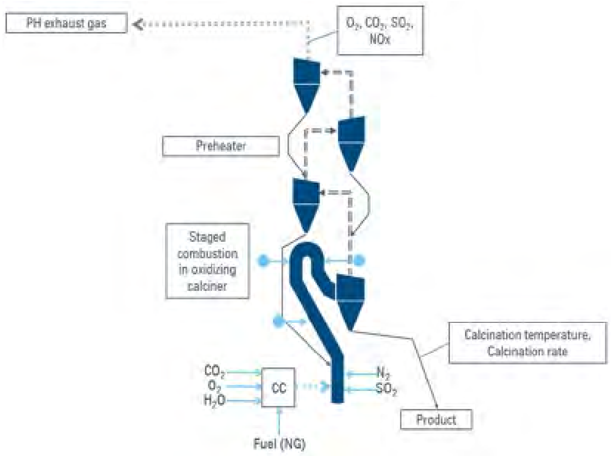


Figure 6: Sketch of the pilot-scale facility of thyssenkrupp Industrial Solutions AG, Germany

Evaluation of oxyfuel calcination process with up to 100 % alternative fuel combustion will be conducted in a 1 MW oxyfuel calciner facility of Air Liquid, Figure 7. The facility is constructed to emulate the conditions of calciners under oxyfuel conditions. The calcination of limestone is simulated thanks to the injection of cold $CO_2$ and fireclay in the combustion chamber.

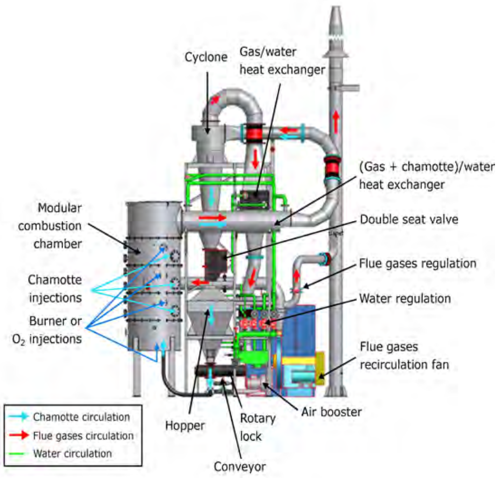


Figure 7: Sketch of the 1 MW calciner facility at Air Liquide, France

## 4. Integration of oxyfuel technology by retrofitting existing cement plants

### 4.1 Cement plants for the retrofit examples

The ability for retrofitting 1st generation oxyfuel technology to existing cement plants is evaluated within WP4 of the AC$^2$OCem project. The project partners, HeidelbergCement and LafargeHolcim, have selected their Slite plant in Sweden and the Lägerdorf plant in Germany respectively to serve as design examples. Both plants are operated with a relatively high alternative fuel rate (70-80 %) typical for Northern Europe and therefore represent a realistic case even for the near future. The major difference of the plant is the production process influenced by the given boundary conditions of the plant location. Where the process in Slite is similar to the reference case (as described in the CEMCAP project framework), the Lägerdorf plant is operated in a so-called semi-wet process. The natural raw material from the Lägerdorf quarry shows a high moisture content of about 20 %, which influences the pre-treatment of the material prior to being fed to the burning process.

### 4.2 Heat integration

All available waste heat from the process has to be supplied to the drying unit (hammer mill flash-dryer), making the heat integration in oxyfuel mode a challenge. Based on the plant specific boundary conditions and taking into account the mass and energy balances performed by combined modelling approaches of SINTEF and VDZ, an optimized oxyfuel design layout has been developed for each plant with the help of the equipment supplier thyssenkrupp. In this development process, the optimal performance in terms of energy use and minimum $CO_2$ generation, as well as the technological risk mainly due to false air ingress, has been assessed. Usually process waste heat from the flue gas is directly used in the drying unit, which is difficult to seal especially in case of retrofit. As shown in Figure 8 the energy demand of the Lägerdorf plant for drying is significantly higher than of Slite plant (or the CEMCAP reference with 6 % raw material moisture).

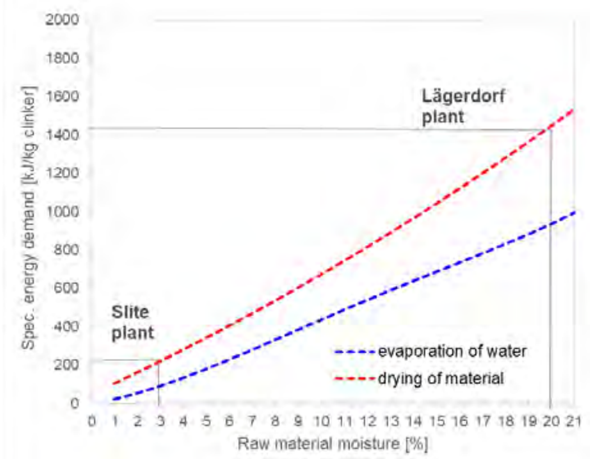


Figure 8: Energy demand for raw material drying for the Slite and Lägerdorf plants

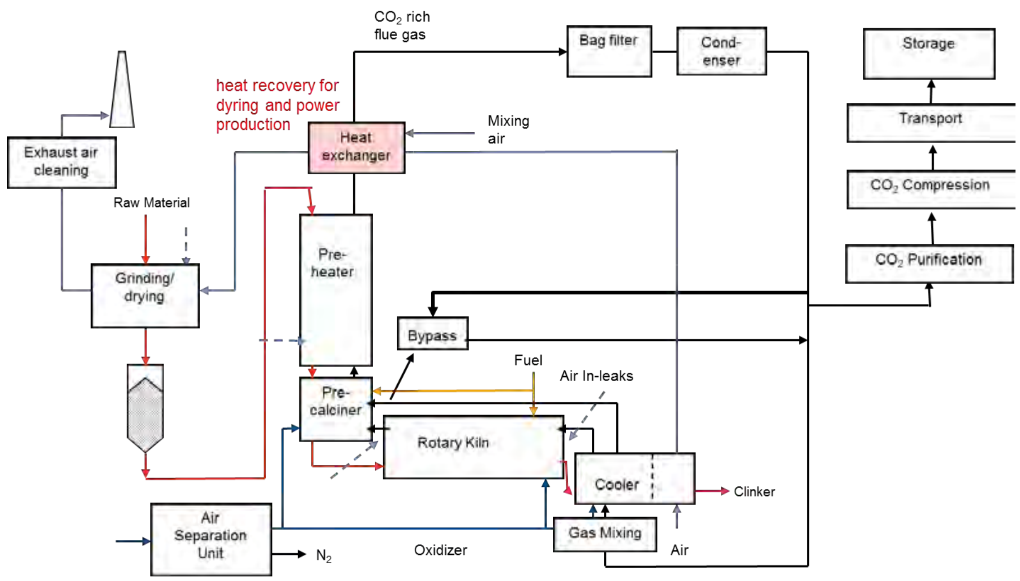


Figure 9: Oxyfuel 1st generation layout for Slite and Lägerdorf plant

The optimal location of the hammer mill dryer in the process layout played an important role. Although the direct integration is energetically reasonable, the operational risk by false air intrusion is high. In order to avoid dilution of the $CO_2$ and its negative impact on energy demand of the CPU, this layout has not been further developed. The overall energetic evaluation proved that a sophisticated heat exchanger system could provide enough energy (also in terms of gas volume and temperature) to the drying unit. For this reason the following layout has been chosen for both plants (Figure 9). The main technological risk for the Lägerdorf plant arises from the gas/gas heat exchanger installed in the flue gas path under hot (~500 °C) and dust loaded (~100 g/Nm$^3$) conditions. In case of the Slite plant, the high SOx emissions from the raw material makes an additional scrubber necessary in conventional mode. In which extend this scrubber has to be integrated into the oxyfuel layout is discussed with the supplier of CPU.

### *4.3 False air ingress*

For each oxyfuel process false air intrusion is the major aggressor due to influence on the target of enriching $CO_2$ to a reasonable level for operating an energy efficient CPU. 6 % false air (related to the total flue gas volume) has been determined as an optimistic and realistic value with regard to sealing the clinker burning process. But it is likely that within the year of kiln operation false air is increased due to wear at the sealings. In case of doubling the false air ingress the $CO_2$ concentration is reduced from initially 81 vol. % to 71-73 vol. % on dry basis (Figure 10). As a rule of thumb, gas supplier AirLiquide named a change of +/- 2 % spec. energy demand for each -/+ 1 percentage point $CO_2$ dry base concentration for design basis of a CPU. As a CPU is designed for a dedicated operational point (e.g. for a pessimistic $CO_2$ concentration), false air influences OPEX and CAPEX of the CPU.

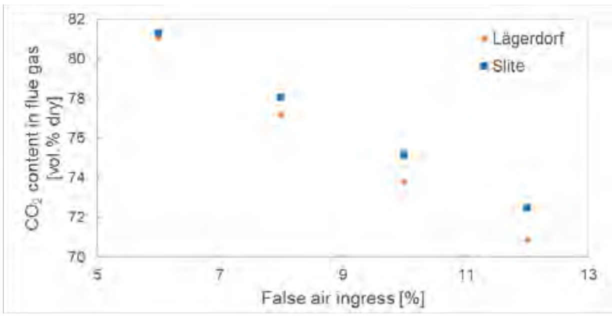


Figure 10: $CO_2$ concentration in flue gas depending on false air ingress

This again clearly shows the need for sophisticated long-living sealings and a good management for false air detection. Against this background the project partners have developed a guideline document for this purpose, which describes apart from the definitions, typical values, the economic effect and examples for improvement, the methods and techniques to detect false air. The resulting detection strategy (Figure 11) includes the continuous online measurement of $O_2$ and $CO_2$ (common UV, IR or paramagnetic measuring) as direct nitrogen measurement is complex and expensive. After having traced back the intrusion to a certain unit, false air can be further localized by ultrasonic detectors, thermal cameras or absolute pressure measuring devices. After the location of the leak is identified, the maintenance department will need to decide if it can be repaired during operation or if it may require a temporary fix until a kiln stop can be scheduled.

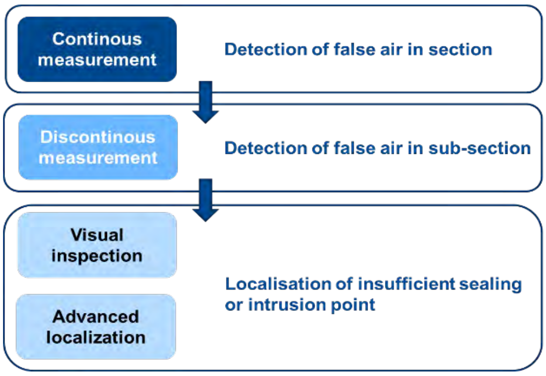


Figure 11: False air detection strategy

### *4.3 Techno-economic evaluation*

A techno-economic evaluation will be performed for integration of oxyfuel technology in the Slite and Lägerdorf plants, where the main performance indicator will be the cost of $CO_2$ avoided. A bottom-up approach will be used for estimation of CAPEX related to the modifications that have to be done to the plants based on equipment cost estimated with Aspen Process Economic Analyzer® for standard process equipment and estimates from the project's industrial partners and data available in the literature for non-standard equipment. Variable operating costs, which includes fuel and raw material costs, utilities, and other consumables, will be estimated based on the process models developed in the project, while fixed operating costs will be estimated using a factor approach. Based on the analysis of the two plants and the retrofitability study performed in the CEMCAP project [9], a guideline for techno-economic decision-making will be made for retrofitting oxyfuel cement plants.

## 5. Oxyfuel technology for new-build cement plants

The idea of the 2nd generation oxyfuel layout developed by thyssenkrupp is to avoid the effort for flue gas recirculation with the aim to reduce CAPEX and OPEX costs. A simulation study of VDZ and SINTEF based on a reference plant (described in CEMCAP framework) will evaluate the operational performance of this layout as basis for a comparative techno-economic study to 1st generation oxyfuel. First modelling results using reference conditions showed the significant need of heat integration when using this kind of process to increase the overall use of energy. Potential to improve the SPECCA is feasible by either a change in gas ducting or the integration of a power generation unit. Further calculations will be made with regard to the use of alternative fuels (implying the integration of a bypass system) and its impact on energy efficiency.

## 6. Life cycle assessment

Life cycle assessment (LCA) is a widely used tool to evaluate the sustainability of a product considering all the production and use stages, e.g. from raw materials extraction to disposal. This method has been largely applied to address the carbon emissions of several cement production technologies, including pre- and post-combustion carbon capture technologies [14]. An unprecedented LCA of oxyfuel retrofitted cement plants will be provided based on realistic operational data. It will be compared with the analysis of a 2$^{nd}$ generation new-build cement plant, considering different production scales. The assessment will also consider the operation of oxyfuel cement plant with high shares of alternative fuels (with substantial shares of biogenic carbon), testing the possibility of reaching negative emissions in the cement production process. The LCA will include not only climate change impacts, but also other categories like non-renewable energy use, human toxicity, ecotoxicity, acidification, particulates and ozone formation. A structural path analysis will be used to identify hotspot stages with increased environmental pressure. Options to improve these pressures, including technology options and pollutants emission minimization and heat recovery measures under consideration in this project, will also be explored in scenario analyses. The quantification of net carbon benefits from decarbonization of the cement industry in Europe from a large-scale deployment of the oxyfuel technology on reducing $CO_2$ emissions and other environmental impacts will also be performed.

## 7. Conclusion

The aim of the AC²OCem project is to decarbonize the cement sector, which is a $CO_2$ intensive industry due to the $CO_2$ emissions from the calcination and the combustion process. The project focuses on designing and optimizing the oxyfuel combustion technology to promote CCS in existing and new-build plants. The boundary layer information from the Heidelberg Cement plant in Slite and the LafargeHolcim plant in Lägerdorf will allow the project partners to design and optimize layout with oxyfuel technology. The different energy demands for the raw material drying are taken into account and the effect of the false air ingress on the CPU is evaluated. Pilot-scale experiments and CFD simulations are also performed to optimize the combustion of alternative fuels under oxyfuel conditions. Additionally the effect of the oxyfuel technology on the calcination process is studied and the process is optimized. The project also focuses on new-build plants, where the burner technology will be evaluated in pilot-scale experimental set-up and evaluated analytically. An optimized process layout will be designed aiming at reducing the CAPEX and OPEX costs. A techno-economic assessment and life cycle assessment of retrofitted and new-build processes will also be evaluated. In scope of the project, the partners will publish the finding in journals and in the scope of various conferences to increase the public acceptance and knowledge regarding the oxyfuel and CCUS technology.

## 8. Nomenclature

| Symbol | Description |
|---|---|
| AC²OCem | Accelerating Carbon Capture using Oxyfuel technology in Cement production |
| ACT | Accelerating CCS Technologies |
| CAPEX | Capital costs |
| CCS | Carbon Capture and Storage |
| CCUS | Carbon Capture Utilization and Storage |
| CEMCAP | $CO_2$ capture from cement production |
| CFD | Computational Fluid Dynamics |
| CPU | $CO_2$ purification unit |
| ECRA | European Cement Research Academy |
| IEA | International Energy Agency |
| IRENA | European Cement Research Academy |
| LCA | Life Cycle Assessment |
| MEA | Monoethanolamine |
| OPEX | Operating costs |
| SRF | Solid Recovered Fuel |
| TRL | Technology readiness level |
| WP | Work package |

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