Adva**n**ced **I**ndirectly Heated **Ca**rbonate Looping Process

Accelerating CCS Technologies
Project No. 299653, ANICA

<u>ACT2 Final Report</u>

ANICA

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1 Identification of the Project and Report

Project title	ANICA—Advanced Indirectly Heated Carbonate Looping Process
Project ID	299653
Coordinator	Dr.-Ing. Jochen Ströhle
Project website	*https:\\act-anica.eu*
Reporting period	Final

Project Partners

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2 Executive Summary

The ANICA —Advanced Indirectly Heated Carbonate Looping Process— project aimed at developing new concepts of the indirectly heated carbonate looping (IHCaL) process for CO_2 capture from lime and cement plants. The project consortium was composed by twelve partners from three different countries: Germany, United Kingdom (UK), and Greece.

The IHCaL process can use the raw materials for lime and cement production as sorbent, thus reducing energy requirements and costs. The heat for sorbent regeneration can be provided with waste-derived fuels (e.g. refuse-derived fuel, RDF), which has significant economic advantages. Furthermore, the use of heat pipes to provide heat indirectly avoids the necessity of generating pure oxygen for the combustion, thus reducing the specific energy requirements.

Within the ANICA project, the IHCaL process was developed with respect to integration strategies for its application in the lime and cement production. For this, different configurations were established and the corresponding mass and energy balances were computed. It was shown that using IHCaL technology with heat from waste-derived fuels enables carbon dioxide removal, i.e. net negative CO_2 emissions, in the lime production. Low values of specific primary energy consumption per CO_2 avoided (*SPECCA*) were obtained (< 2.5 $MJ_{LHV}/t_{CO2,av}$), which reveals the high potential of this application in terms of energy efficiency.

Two test campaigns in a 300 kW_{th} pilot test rig were performed, validating the utilization of the IHCaL process in the cement and lime production. In particular, the feasibility of burning RDF for sorbent regeneration and capturing CO_2 from this off-gas was demonstrated through the commissioning and first-time operation of the solid feeding system and flue gas tract into the carbonator. The results from spent sorbent analyses showed that the sorbent integration in the lime and the cement industry would be possible, but more experimental validation is required, especially regarding the use of cement raw meal in IHCaL facilities.

Key components of the IHCaL were designed using simulative and experimental methods. New process and reactor models were developed and validated with data from pilot tests. These included CFD models that were used for the interpretation of the test results, as well as for the design of full-scale facilities and a demonstration plant. Novel concepts of solid-solid heat exchangers for pre-heating of solids entering the calciner as well as a two-stage calciner were developed and evaluated with the aim of reducing the heat requirements for the calcination. Furthermore, advanced layouts and different configurations of the heat-pipe heat exchanger were assed to maximize the heat transfer to the calciner.

The IHCaL technology configurations for lime and cement applications were evaluated by qualitative (FMECA) and quantitative (Monte Carlo) risk assessment, techno-economic analysis, and life cycle assessment. The main risks were identified through the collaborative work from many partners from the academia and the industry, thus reducing uncertainties and facilitating the technology scale up. The IHCaL process has the potential to decarbonise lime and cement plants with low CO_2 avoidance costs (20–25 €/$t_{CO2,av}$, for optimized configurations) and reduced environmental impact enabling net negative CO_2 emissions. Finally the potential synergies between the IHCaL process and the Leilac Direct Separation process were assessed. The integration of these two technologies would enable CO_2 capture from lime and cement plants with very high capture rates (98%) and reduced costs (31 €/$t_{CO2,capt}$).

To pave the road for industrial application of the IHCaL technology, a 2-MW_{th} IHCaL plant demonstrator was designed, included concept design and basic engineering. A lime plant operated by one of the industrial partners was selected as the host site. The plant layout was established using 3D-CAD software and the investment and operation costs were estimated.

3 Role and Contribution of Each Project Partner

Technische Universität Darmstadt (TUDA)

The Institute of Energy Systems and Technology at the Technische Universität Darmstadt (TUDA) contributed with the process development of the IHCaL process for lime applications. Within this task, new models and process configurations were developed. TUDA also contributed with the pilot testing of the IHCaL concepts in their 300 kW_{th} test rig. For this, the testing facility was expanded with a flue gas recirculation tract and a feeding system for solid fuels, i.e. coal and refuse-derived fuel (RDF). The pilot tests demonstrated the technical feasibility of capturing the CO_2 from the combustion within the IHCaL facility. The results and samples from the pilot tests served as input for other work packages and validated the integration concepts of the IHCaL for lime and cement applications. TUDA was also involved in other tasks such as the project coordination (lead), the "development of a solid-solid heat exchanger" (lead), and the "dissemination and exploitation" of the ANICA project (lead).

Friedrich-Alexander Universität Erlangen-Nürnberg (FAU)

Utilization of coal and biomass is one of the main focuses of the Chair of Energy Process Engineering of FAU. Others are dealing with CCS-technologies, synthesis of natural gas by gasification and methanation, and combustion technologies for coal and biomass. The institute has many years of experience in the field of development and integration of high-temperature heat pipes in different processes.

Within the ANICA project, FAU has conducted experiments in a lab-scale unheated second-stage calciner and tested a compact heat-pipe heat exchanger concept. Furthermore, FAU worked on the development of advanced heat pipes.

VDZ Technology gGbmH (VDZ)

VDZ Technology gGmbH (VDZ), with 135 years of experience in energy and resource-efficient cement production, is directly linked to more than 70 cement producers worldwide. A broad connection between VDZ and the cement industry facilitated the knowledge sharing and results dissemination among the end users. VDZ organized and hosted the final ANICA public workshop. VDZ was the leader of the work package "process development" for the development of novel process concepts for the integration of IHCaL carbon capture into lime and cement plants, and helped define the operating conditions to be used for pilot testing. This work package included kiln process simulations for fully integrated carbonate looping process into a cement plant. In addition, VDZ provided heat and mass balances of full-scale IHCaL integrated configurations for cement plants as a basis for techno-economic, environmental, societal, and risk assessments in the process assessment study.

thyssenkrupp Polysius GmbH (TKIS)

thyssenkrupp Polysius GmbH (TKIS) is one of the leading engineering companies for the cement industry. Within the ANICA project, TKIS lead the work package "Design of a fluidized bed demonstration plant".

TKIS defined a concept for integrating a fluidised bed IHCaL demonstration plant into the host site and elaborated the process flow diagrams, defined the major components and developed an instrumentation & control concept of the demonstration plant. Together with TUDA and FAU, the basic layout and dimensions of the IHCaL reactor system (carbonator, calciner, combustor, heat pipes, and solid-solid heat exchanger) were defined. TKIS designed the detailed plant layout using 3D CAD software, including specification of the main reactors. Finally, TKIS estimated the equipment costs of the major components, the costs of consumables, and the staff costs for operating the plant. Based on these data, the investment and operating costs of the demonstration plant were calculated.

Lhoist Germany Rheinkalk GmbH (LGE)

Lhoist Germany Rheinkalk GmbH is a subsidiary of the Belgian Lhoist group that employs around 6.000 permanent staff in 25 countries and is the world-leading supplier of high quality lime, dolime and minerals. The company is specialised in the exploration, mining and refinement of limestone, dolomite and related products. Thanks to its Corporate Business & Innovation Center (BIC) in Nivelles, Belgium, LGE has developed a vast experience in material characterization as well as the development of new products and processes. LGE also developed special test procedures to evaluate the hardness and the CO_2 uptake of various sorbents in the frame of carbonate looping.

LGE contributed by providing limestone as raw material for the pilot tests and by determining the chemical and physical properties of the “burnt material” from the IHCaL process with respect to its quality and potential utilization in different fields of lime applications. LGE also contributed in developing concepts to integrate the IHCaL demonstration plant into the reference lime plant.

Dyckerhoff GmbH (DYCK)

Dyckerhoff GmbH (DYCK) is an internationally operating producer of cement and concrete within the group of companies belonging to Buzzi Unicem (Italy). DYCK contributed with the pre-selection of raw meals for the experimental work within the ANICA project. Different raw meal samples from Göllheim and Geseke were provided to TUDA. The samples, which added up to 3–4 tonnes of raw meal, were used for fluidisation tests.

Furthermore, DYCK supported the spent sorbent characterization through the performance of chemical and mineralogical analysis of fluidised meals from FAU. For this, X-ray fluorescence and X-ray diffraction were used. Additionally, burning tests on raw meals at different temperatures were performed in the Wilhelm Dyckerhoff Institute (WDI). The tests took place at laboratory scale, including chemical and mineralogical analysis.

Finally, DYCK supported the partners with advice concerning different issues that arouse throughout the project. These included questions regarding clinker and cement chemistry, and properties of cement, such as hydration behaviour.

Prezero Stiftung & Co. KG (PREZ)

Prezero Stiftung & Co. KG —previously Suez Recycling Süd GmbH— operates waste treatment plants for the production of waste-derived fuels and provides nearly all kinds of waste management services, e.g. collection, mechanical treatment, incineration, and landfilling. The original task of PREZ was to deliver waste-derived fuels for the operation of the 300 kW_{th} pilot plant at TUDA. However, during the course of the project, it was detected that the fuels provided by PREZ are not suitable for this pilot plant.

ESTRA Energy Strategies Ltd. (ESTRA)

ESTRA Energy Strategies Ltd. (ESTRA) is a small engineering consulting company based in the United Kingdom. ESTRA was responsible for the qualitative and quantitative risk and reliability analysis. The qualitative risk analysis was performed with the Failure Mode, Effect and Criticality Analysis (FMECA) method. The quantitative risk analysis was based on the Monte Carlo and System Dynamics methods. Furthermore, ESTRA supported ULSTER with the techno-economic assessment of the IHCaL processes.

Ulster University (ULSTER)

Ulster University (ULSTER) is located in Northern Ireland, UK. The work in this project was conducted by the Centre for Sustainable Technology (CST). This department undertakes multidisciplinary research to design, create, develop, improve, demonstrate, and evaluate emerging, existing, and alternative sustainable energy and environmental technologies, producing many high-quality publications in clean coal technologies, CO_2 capture and storage technologies. ULSTER, leading the "process assessment", carried out comprehensive techno-economic assessment and life cycle analysis for the full-scale implementation of lime and cement plants integrated with IHCaL within ANICA, including energy requirements, efficiencies, break even costs of products for the cost of CO_2 avoided in comparison to other CO_2 capture solutions.

Calix Europe Limited (CALIX)

Calix Europe Limited's (CALIX) mission is to provide the most compelling solution for decarbonising unavoidable process emissions from the cement and lime sectors. With proven multi-tonne per hour CO_2 capture plants, CALIX can also use any energy source, including renewable electricity, to provide future-proof optionality for cement and lime producers as they transition to net zero. CALIX is also partnering with Heirloom, a direct air capture (DAC) company, to use their Leilac technology to decarbonise limestone, which allows Heirloom to use lime's absorbent qualities in a new and crucial way to remove atmospheric CO_2.

CALIX's contributions to the ANICA project focused on evaluating the coupling of Leilac and IHCaL technologies by providing process concepts, detailed process modelling, and a techno-economic assessment that informed ANICA's technology roadmap.

Centre for Research & Technology HELLAS (CERTH)

The Centre for Research and Technology Hellas (CERTH) is one of the largest research centres in Greece. Within CERTH, the Chemical Process and Energy Resources Institute (CPERI) has extensive expertise in Computational Fluid Dynamics (CFD) and CO_2 capture process modelling.

In the context of ANICA, CERTH has built both a pure Eulerian, as well as an Eulerian-Lagrangian Dense Discrete Phase model (DDPM) to simulate the complex two-phase flow phenomena in the bubbling bed calciner. Additionally, CERTH developed a novel solid-solid heat exchanger design featuring two L-valves with concentric vertical tubes. Cold flow experiments were conducted at CERTH on a small scale, while the heat exchanger was assessed under real industrial conditions by means of proper heat transfer models. Furthermore, CERTH worked together with TUDA and developed an integrated model of the IHCaL process at a lime plant at full scale for both the tail-end and the fully integrated concepts.

CaO Hellas (CH)

CaO Hellas (CH) is the largest group of companies in Greece and the Balkans specialized in the production of lime and other chemical products based on lime. Within the ANICA project, CH assisted in the integration of the IHCaL concept into their lime plant in Thessaloniki. For this, they provided process data for simulations and technical information of the plant.

Additionally, CH performed analysis of spent sorbent samples from the pilot plant operation campaigns. The sample material was evaluated and compared with the top-grade commercial product CH supplies to the Greek Alumina production industry.

4 Short Description of Activities and Final Results

The work programme of the ANICA project was divided into eight work packages (WPs), as illustrated in Figure 1. Within this chapter, the activities and final results are presented in sections, each corresponding to one of the work packages (Sections 4.1–4.8). At the end of this chapter, the financial overview and the list of deliverables are included (Section 4.9 and Section 4.10, respectively).

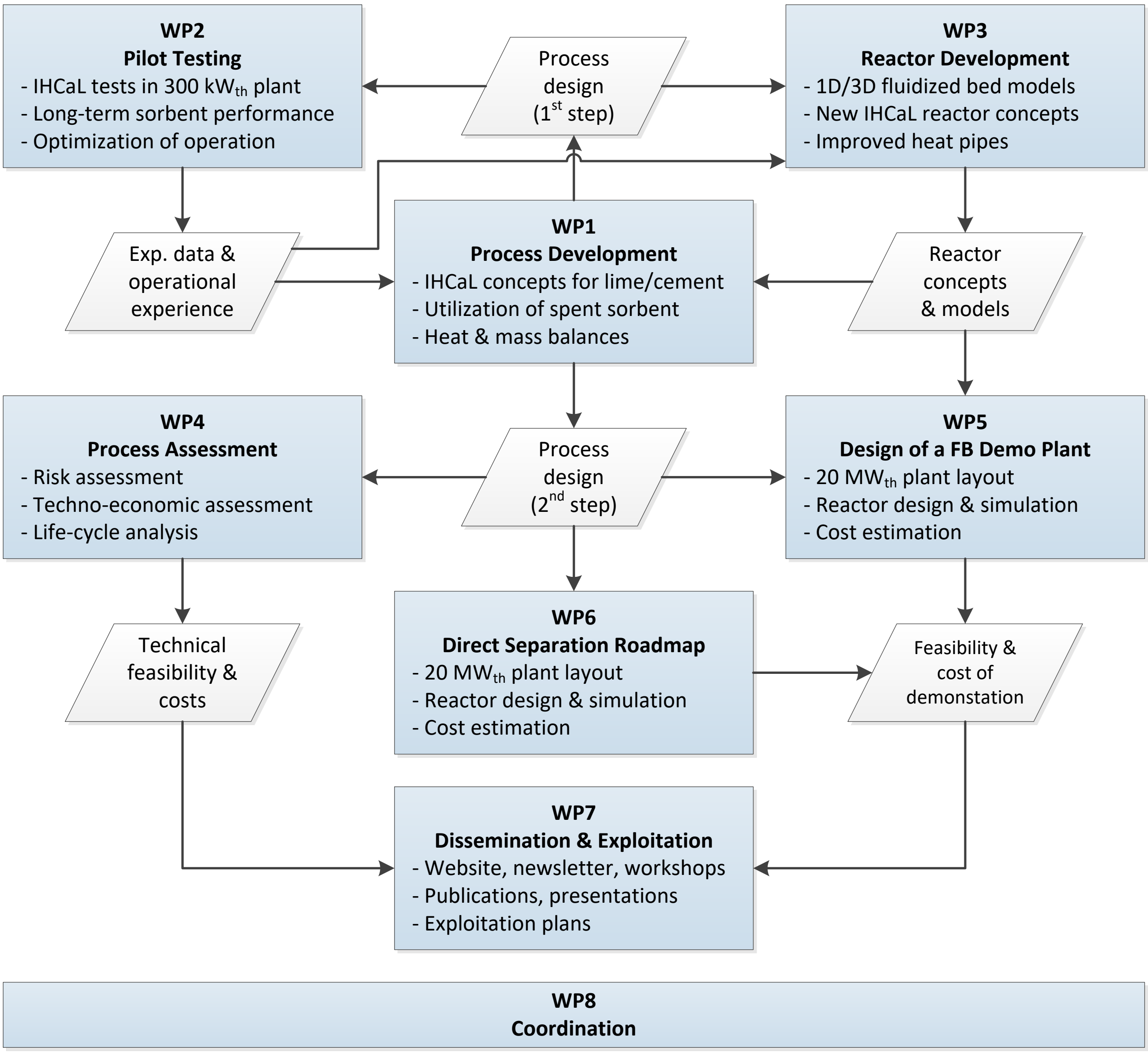

Figure 1. Project structure from the ANICA proposal [1].

The central work package (WP1) consisted in the development of the process (Section 4.1). In a first step, previous models were used to develop integration concepts and to define the operating conditions for the pilot testing (WP2, Section 4.2). The experimental data from WP2 was used to validate 1D and 3D models of the IHCaL reactors developed in WP3 (Section 4.3). WP3 also included the development of novel concepts to optimize the IHCaL reactor system. The results from WP3 were used to optimize the process concepts in a second step within WP1 (Section 4.1). Using the updated heat and mass balances from WP1, the integration concepts were assessed with respect to risks, techno-economics, and environmental impact (WP4, Section 4.4). The results from previous work packages were used for the design of a 2 MW_{th} demonstrator (Section 4.5) and for the integration of the IHCaL with the Leilac Direct Separation technology (Section 4.6). The project results were disseminated and exploited within WP7 (Section 4.7). The final work package (WP8) consisted in the management of the project (Section 4.8).

4.1 Process Development

4.1.1 Integration into the Lime Process

This task included the development of integration concepts and the detailed analysis of the process integrated into the lime production. This work built upon other investigations within the ANICA project, such as the pilot testing of the IHCaL in lime conditions (Section 4.2), and the detailed modelling of the main components of the IHCaL (Section 4.3). Based on the project findings, a rigorous process model was developed and used to analyse CO_2 capture concepts for two real lime plants —lime plant Hönnetal (Germany) and lime plant Thessaloniki (Greece)— using IHCaL technology. One integration concept consisted in the tail-end retrofitting of an existing lime plant (see Figure 2). The other concept involved a full integration of the IHCaL process for production of lime and capture of the CO_2 generated in the combustor.

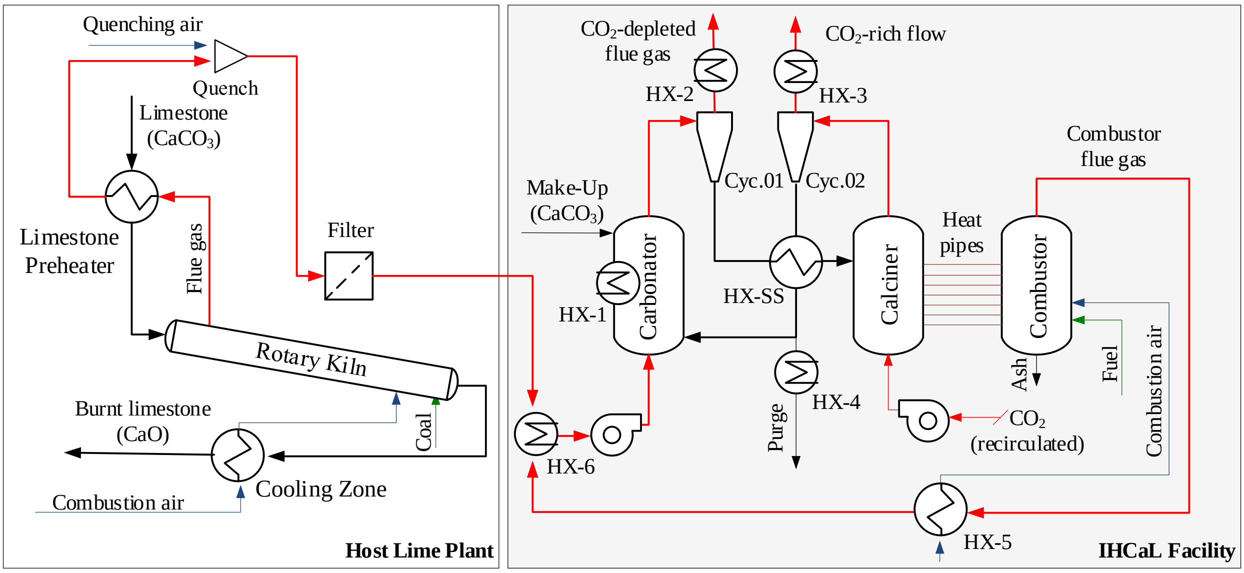

Figure 2. Tail-end IHCaL configuration for lime plant Hönnetal (LGE) in Germany from [2].

Firstly, simple models were used to obtain preliminary results [3]. Afterwards, using the experience gained throughout the ANICA project, more exact calculations were performed. The results show that the original estimations are valid to obtain qualitative information on the sensitivity of the process to the variation of key operating parameters.

Considering the scientific advances achieved throughout the ANICA project, the potential of the technology is now better understood. Very low values of specific primary energy consumption per CO_2 avoided (*SPECCA*) were obtained. The results range between 0 and 2.5 $MJ_{LHV}/t_{CO2,av}$, which reveals a very high energy efficiency compared to other processes for the same application, e.g., 7 $MJ_{LHV}/t_{CO2,av}$ for MEA scrubbing and around 3 $MJ_{LHV}/t_{CO2,av}$ for oxy-fired carbonate looping [4]. Furthermore, the technology has the potential to enable net negative CO_2 emissions [2].

Different optimization routes were analysed. Among them, the fully integrated configuration with low circulation rates and the firing of waste-derived fuels are key features to achieve efficient CO_2 capture in the lime sector with IHCaL technology. These approaches still require further research in order to enable the commercial application of the technology.

4.1.2 Characterization of Spent Sorbent Regarding Utilization in the Lime Process

Solid samples were taken after the calciner from the loop seal during pilot test (see Section 4.2.2 and 4.2.3) in order to assess their quality to be used in lime production. The analysis were performed at laboratories of LGE and CH in terms of chemical and physical properties of the used sorbent from pilot tests with respect to utilization in their lime applications. Spent sorbents were characterized by reactivity test and wet slaking behaviour according to EN 459-2 (determination of physicochemical parameters and neutralization capability of Milk of Lime). The results were assessed in terms of the potential to be used in the specific market.

Greek market (CH): All four samples from the first campaign generated similar results, giving a > 1mm sieving residue in the 5.5–6% range. A material of this granulometry in practice is suited only for the supply of the local Alumina Industry in Greece. Nevertheless, it exhibited favourable performance compared to the established commercial product CH supplies to the Greek Alumina Industry. The samples from the second campaign are unsuitable for utilization as product because they failed the reactivity tests and presented high amounts of carbonate. Additionally, a substantial concentration of sand particles was noticed.

German market (LGE): Four samples from the pilot testing (see Section 4.2.2) fulfilled the ‘CL90’ specifications[1]. These samples could therefore be used in existing applications for such quicklime types. However, more detailed information on the share of these samples on the total mass flow in the IHCaL process are necessary to verify the potential production capacity. Compared to the original limestone, all samples showed higher SiO_2 values (sometimes > 10 wt-%), indicating a potential contamination of the sorbent, most probably by bed material from the combustor (SiO_2-rich sand). Reactivity tests of samples with rather high SiO_2 but low CO_2 values showed higher t_{60}-times and lower maximum temperatures because of the “dilution effect” of the SiO_2. This indicates that the quicklime fraction of these samples has a “soft burnt” reactivity.

4.1.3 Integration into the Cement Process

An integration concept for the IHCaL process in a cement plant was developed. As part of this task, the existing VDZ process model for the clinker burning process was enlarged to represent the fully integrated IHCaL process. The used reference plant is based on the BAT (Best Available Technique) standard as defined in the European BREF (Best Available Technique Reference) document for the production of cement [5]. The concept should allow an efficient and safe operation. Sophisticated control systems are crucial due to the interdependent nature of the circulating loops, requiring precise management to minimize fluctuations [6].

In a series of simulations, the process was optimized to reduce fuel demand in the combustor and kiln while enhancing CO_2 capture efficiency considering specific constraints. These constraints included maintaining tertiary air temperature below 450 °C due to requirements of the combustor, achieved by increasing cooler air volume. Similarly, the gas temperature before the calciner needed to be below 450 °C, which was achieved by waste heat recovery units and water injections. Water vapour was added to lower the CO_2 concentration and enhance heat transfer. A gas mixture of 30 % water vapour and 70 % carbon dioxide ensures optimal conditions, enabling calcination at 880–900 °C with 97 % degree of calcination achieved in the hot meal entering the kiln.

Both plants produce 125 t/h clinker and process 200 t/h raw meal. In the IHCaL plant, 380 t/h of sorbent is recirculated between the calciner and carbonator, which is nearly double the amount used as raw meal. Since calcination takes place at a higher CO_2 atmosphere and higher temperature, the raw meal enters the rotary kiln with higher enthalpy and clinker formation. Consequently, 30% less fuel is required in the rotary kiln. In the combustor, on the other hand, 3.3 times the amount of fuel is required compared to the reference calciner, as a larger amount of material (raw meal + sorbent) needs to be heated to 880 °C.

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[1] The European building lime standard EN 459 defines different classes of quicklimes for the use in building applications; the class with the highest purity (‘CL 90’) needs to have, among other parameters, a minimum of 90 wt.-% ‘CaO + MgO’ and a maximum of 4 wt.-% CO_2 (EN 459-1).

The fully integrated IHCaL process plant needs 2.3 times more thermal energy than the reference plant and avoids 57.4 % of the reference CO_2 emissions. To achieve this capture rate, almost two times more mass is needed in the loop than fresh raw meal for clinker production. Due to the interdependency of CO_2 generation in the calciner and the carbonator reaction as well as the sophisticated heat integration system, the operation of the process is very complex and smaller process fluctuations could cause major instabilities in other process components. To transport this amount of material, mechanical energy is needed, which was not considered in the present study.

4.1.4 Characterization of Spent Sorbent Regarding Utilization in the Cement Process

DYCK carried out several research activities to investigate the feasibility of using purged material from the IHCaL process as basic raw material for the production of Ordinary Portland Cement (OPC) cements. Therefore, purges from pilot testing (Section 4.2.2 and Section 4.2.3) were investigated with the aim to be reused in the clinker burning process. VDZ assisted the characterization based on their expertise in the field of cement and clinker production.

From the analytical results, no clear statement to the usage of deactivated sorbent in cement clinker can be derived, due to the limited amount of samples from pilot tests. The results of the performed XRD-analysis of the phase formation (for samples collected during the first test campaign) revealed that the quality of this materials might be sufficient. A reliable characterization of sorbent requires the sampling of significant uncontaminated amount of material from the process. Since no pilot tests with cement raw meal could be performed, no purged meal was available for the used sorbent assessment.

4.1.5 Experiments on Cement Raw Meal

During the initial commissioning trials of the batch calciner at FAU, attempts were made to assess the calcination of raw meal pellets. However, unforeseen challenges emerged, including agglomeration issues and the inability to carry out carbonation. The phase formation of the cement raw meal was identified as a probable cause for these problems. Hence, the consortium decided to perform additional work (which was not part of the original work plan) in order to address these challenges. This was done by FAU in collaboration with DYCK and VDZ.

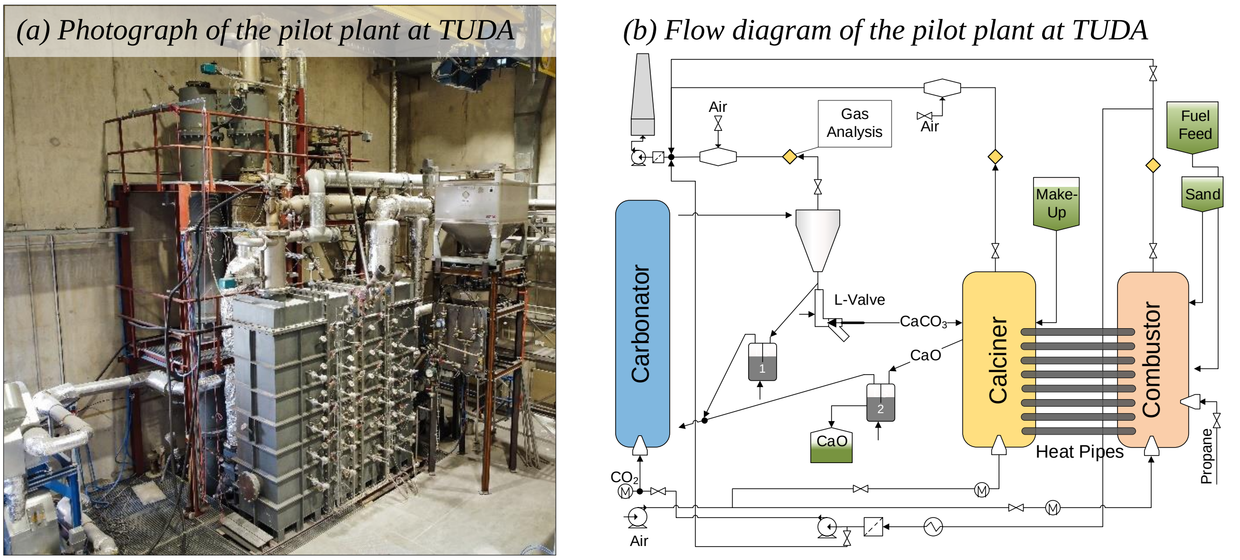
Utilizing thermogravimetric analysis (TGA), suitable parameters for calcination were determined to facilitate subsequent carbonisation. At 900 °C, it was observed that the calcination time was inadequate, and a significant amount of free lime only manifested above 930 °C. Beyond this temperature, a residence time of one minute proved effective. Fluidisation tests revealed that neither the cement raw meal nor the pellets could be fluidised without a substantial material discharge. Furthermore, agglomeration tests demonstrated that cement raw meal did not exhibit agglomeration, whereas agglomeration occurred with the cement pellets.

FactSage equilibrium simulations were conducted, indicating that the formation of belite is inevitable for the presence of free lime. Phase analyses revealed that the free lime content is constrained by phase formation compared to limestone, with belite identified as a significant phase. Additionally, spurrite was detected as a "free-lime-taking" phase. The study showed that cement raw meal, when used in carbonate looping with a 75% admixture of untreated cement raw meal, complies with cement industry standards, ensuring sufficient product quality.

As part of this task, preliminary insights were gathered regarding the behaviour of cement raw meal in the carbonate looping process. Altogether, a suitable operating point for calcination was identified, ensuring that the cement raw meal provides enough free lime for subsequent carbonation. Although cement raw meal can be fluidised, it exhibits high entrainment rates, necessitating the development of solutions for particle recirculation.

4.2 Pilot Testing

The aim of this work package was to push forward the IHCaL technology to the next level of maturity by demonstrating the technology in a real environment for cement and lime applications. This was achieved with tests in an upgraded 300 kW_{th} pilot plant with real flue gas (see Figure 3).



(a) Photograph of the pilot plant at TUDA

(b) Flow diagram of the pilot plant at TUDA

Figure 3. 300 kW_{th} IHCaL pilot plant at TUDA: photograph (a) and flow diagram (b).

4.2.1 Design of Pilot Plant Upgrades

Within this task, a flue gas path (ducts, heat exchanger, filter, and fan) from combustor to carbonator and a solid-fuel feeding system for the combustor were designed and installed. The feeding system enabled IHCaL operation fuelling lignite and refuse-derived fuel (RDF) in the pilot plant. Furthermore, additional sampling and measuring equipment (pressure sensors, thermocouples, and gas analysers) to improve the operability and data collection were installed. This task suffered from the difficulties caused by the COVID-19 pandemic and the shortage in the global supply chains leading to a delay in the whole project of around 12 months.

4.2.2 Pilot Testing at Lime Plant Conditions

During the first pilot test, the IHCaL test facility at TUDA was operated with the new plant arrangement for 10 days. The new flue gas path was commissioned successfully and real flue gas enriched with CO_2 was introduced into the carbonator. The operating conditions were defined according to a highly integrated solution, based on results from the process development [7]. Natural limestone from LGE with a mean particle diameter of $d_{p,m}$ = 180 µm was used as sorbent.

Throughout this operational period, CO_2 capture rates in the carbonator (E_{carb}) of up to 90 % were achieved and around 60 solid samples were collected. Six of the samples were selected for further analysis by LGE with respect to particle size distribution, using laser granulometry; chemical composition (CaO, $CaCO_3$, $CaSO_4$, ash), using X-ray fluorescence and diffraction; specific surface area, using the BET method; and porosity, using BJH method. Furthermore, sampled material was sent to LGE and CH for further characterization concerning its utilization in the lime production process (see Section 4.1.2). Over 10 tonnes of sorbent were purged from the system. In the first half of the test campaign, high entrainment rates were observed. After incrementing the make-up feeding rate, the hydrodynamics of the coupled fluidised bed reactors were improved. Around 48 hours of stable operation were achieved.

After the campaign, TUDA and FAU jointly inspected the heat pipes with respect to deformation or damage. Considerable deformation was detected for a few heat pipes, which could have been caused by short periods (of up to 2 min) of elevated temperatures in the combustor (> 1000 °C).

4.2.3 Pilot Testing of Tail-End Integration into a Cement Plant

The IHCaL test facility at TUDA was operated with a new plant configuration for 9 days, aiming at industrial conditions for CO_2 capture from typical cement plants. Limestone from LGE with a mean particle diameter $d_{p,m} = 450\ \mu m$ was used as sorbent. Using this coarser material improved the hydrodynamics significantly and a more stable solid circulation was achieved.

The solid-fuel feeding system was successfully commissioned and 48 hours of operation with solid fuels were achieved. However, it was not possible to attain high temperatures (> 900°C) in the combustor while fuelling lignite or RDF, due to incomplete combustion of solids. The operation revealed that it is necessary to optimize the combustor, e.g. improve the fuel-feeding point, deploy air staging, and regulate the internal circulation of solids. Throughout this operational period, CO_2 capture efficiencies (E_{carb}) of up to 85 % were accomplished and over 200 samples were collected. From them, 20 samples were selected for further analysis by LGE, CH, and DYCK with respect to chemical composition and usage for the cement and lime production.

Leakage problems of gases and solid material between calciner and combustor occurred especially during the first half of the pilot tests. A stable operation (especially at the beginning of the test campaign) was only possible with high make-up rates ($\Lambda > 0.2\ mol_{CaCO3}/mol_{CO2}$). The presence of sand contamination decreased over the time, probably due to the closing of cracks caused by thermal expansion. The inspections after the campaign revealed that significant cracks were present in the middle wall. These cracks were too severe to operate the reactors without serious reconstruction work. Such reconstruction work was not possible within the ANICA project due to temporal and financial constraints. The replacement of calciner, heat pipes and combustor would be necessary for further pilot tests at the 300 kW_{th} pilot plant.

The thermal load in the combustor had to be increased to 300–380 kW_{th} in order to achieve the target temperature in the calciner (> 800°C), which represents an increment of up to 100 kW_{th} with respect to previous test campaigns in the same facility. The high heat requirements were probably caused by the leakages of solids and gases between the reactors, which increased the sorbent loss through entrainment, making it necessary to add more make-up as normally required. Other factors that may have contributed to this problem are the incomplete combustion of solid fuels in the combustor and the reduced heat transfer efficiency of the heat-pipe heat exchanger (e.g. due to low bed height).

All the analysed sorbent samples have high concentrations of $CaCO_3$ ($> 0.5\ mol_{CaCO3}/mol_{Ca}$), which correspond to relatively fresh sorbent with less than five calcination-carbonation cycles, on average. Some explanations for this are the large amount of make-up that was added discontinuously, and the low operating temperature of the calciner, which compromises the calcination efficiency.

After the campaign, TUDA inspected the heat pipes with respect to deformation or damage. Single heat pipes (fabricated with 1.4841) showed strong deformations. In addition, a yellowish and greenish coating bonded to the surface of the heat pipes was visible on the combustor side, indicating a reaction of the high-temperature steel of the heat pipes with combustion gases of waste-derived fuel. Future tests are necessary to improve the understanding on the influence of combustion of waste-derived fuels on the long-term heat pipe performance.

4.2.4 Pilot Testing of High Integration into a Cement Plant

It was originally foreseen to operate the IHCaL pilot plant using cement raw meal from the DYCK cement plant Göllheim as sorbent. However, this was not possible due to several reasons. Firstly, it was not possible to feed cement raw meal into the 300 kW_{th} pilot plant due to flowability issues. Secondly, the malfunction of the middle wall did not allow any further tests in the pilot plant (see Section 4.2.3). Hence, alternative experimental investigations were proposed to the funding agency, which gave their approval. The experimental work for the assessment of the characteristics and operability of cement raw meal in IHCaL fluidised bed consisted in:

- hydrodynamics investigations using cement raw meal in a bubbling bed cold flow model; and
- reactivity investigations of cement raw meal using thermogravimetric analysis (TGA) and an electrically heated laboratory fluidised bed.

Two different types of cement raw meal were considered. The samples, provided by the project partner DYCK, were obtained from the cement plants in Göllheim and Geseke. The materials were assessed in terms of particle size distribution (PSD), attrition behaviour, minimum fluidisation velocity, and angle of repose (see Figure 4).

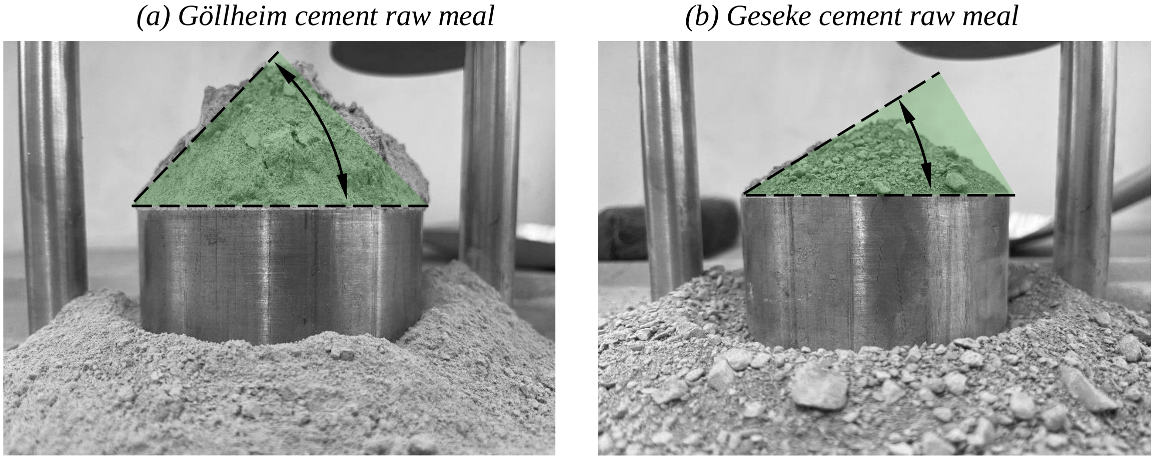

(a) Göllheim cement raw meal *(b) Geseke cement raw meal*

Figure 4. Angle of repose of raw meal samples from the DYCK cement plants in (a) Göllheim and (b) Geseke.

The experimental results are useful to characterize the sorbents regarding utilization in an IHCaL CO_2 capture facility. Cement raw meal from Göllheim is not suitable for bubbling bed operation: no stable fluidisation could be achieved, the high angle of repose (see Figure 4.a) indicates too low flowability, high attrition rates leads to very high material loss through entrainment.

The Geseke material is more promising. If the coarser particles are continuously removed, e.g. through purges from loop seals, the material can be fluidised properly. The attrition rates calculated with the experimental data were much lower than for the Göllheim raw meal, which confirms the higher mechanical stability of Geseke samples. Furthermore, the samples have high flowability (see Figure 4.b), comparable to that of limestone. Finally, preliminary results from reactivity tests [8] suggest that Geseke raw meal has a high carbonation activity, which would make it especially suitable for CO_2 capture in a carbonate looping process.

In further research work, cement raw meal types similar to the Geseke material (low amount of clay) should be investigated regarding utilisation in IHCaL CO_2 capture. Furthermore, a market analysis of existing cement plants using this kind of cement raw meal should be performed.

4.3 Reactor Development

4.3.1 Process Model Development

1-D fluidised bed models for the calciner, combustor, and carbonator of an IHCaL system, utilizing limestone and raw meal as sorbent for the capture, were developed. The simulations were validated with available experimental results from pilot tests using limestone as sorbent. The models of the full cement integration could not be fully validated because of insufficient experimental data (see Section 4.2.4).

The heat transfer model assumes bubbling bed hydrodynamics in both the calciner and the combustor. Many semi-empirical correlations were tested, and the best-fitting one was selected as the appropriate for the simulations. The model is useful to scale-up the IHCaL process, as it provides a calculation tool to estimate the necessary amount of heat pipes —a critical component from the economic perspective of the investment costs— for a good carbon capture performance.

The calciner model considers the operation of the calciner as a bubbling bed, modelled as a continuous-stirred tank reactor with solid particle flow and an exponential distribution for CO_2 partial pressure. TGA analyses were performed to determine the exact kinetic behaviour of the used lime. It was found that the type of lime has a strong influence in terms of temperature and concentration conditions to achieve calcination within the process residence times. The results show good agreement with the experimental data from the pilot tests and are useful to optimize the IHCaL process in terms of calciner operating temperature and steam fluidisation requirements.

The carbonator model considers sorbent deactivation, circulating fluidised bed hydrodynamics in the reactor, the carbonation reaction kinetics, and the limitation of the equilibrium conditions. For the validation, the aging of the sorbent throughout the experimental campaigns was estimated. The solid sample analysis and the energy and mass balance of the plant were used as inputs for the simulation. The vast majority of the validation operating points could be accurately simulated with less than 20% relative error.

4.3.2 CFD Model Development

An Eulerian-Eulerian, two fluid model (TFM), and an Eulerian-Lagrangian Dense Discrete Phase model (DDPM) were built for the calciner reactor of the 300 kW_{th} pilot plant located at the premises of TUDA (see Figure 5). Regarding the TFM model, numerical simulations were conducted within ANSYS® Fluent (v19.2) [9] commercial platform. The drag force was modelled with the advanced sub-grid Energy Minimization Multi Scale (EMMS) approach, which considers flow heterogeneity aspects. Regarding heat transfer, the models considered were both convection and radiation. As for the results, there was a good match with the experiments retrieved from the previous CARINA project [10].

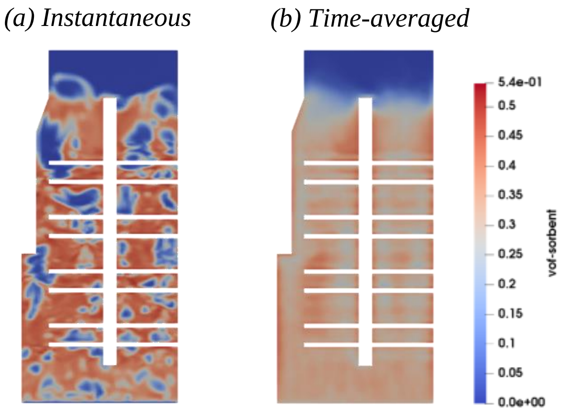

(a) Instantaneous *(b) Time-averaged*

Figure 5. Instantaneous ($t = 15$ s) (a) and time-averaged (b) volume fraction of solid particles (*vof*) of the IHCaL calciner using an Eulerian-Eulerian CFD model. Adapted from [8].

As a follow-up, the CFD model combined with several empirical heat transfer correlations was used to parametrically investigate the effect of fluidisation velocity on the heat transfer coefficient of the heat-pipe heat exchanger. In this way, important conclusions were drawn on the role of each heat transfer mechanism and their dependence on the hydrodynamics. It was shown that changing the fluidisation velocity has a mild effect on the total heat transfer, due to the counterbalancing effect of fluidisation velocity on radiative and convective components. Finally, this model was used to parametrically investigate the arrangement of the heat pipes, concluding that a staggered arrangement with a horizontal pitch of 1.5–2 pipe diameters allows for optimum heat transfer. More information on the TFM model can be found in [11].

The development of the DDPM model was proven to be quite demanding. Being still at an early stage of development in the commercial ANSYS Fluent platform, DDPM required several advancements to be applicable in the dense, high inventory bubbling bed flow considered here. The inter-particle forces were modelled using custom user-defined functions, incorporating both normal and tangential components. In addition, the Lagrangian equation of particle motion was reformulated. It was shown that this was the reason for total pressure drop overestimations predicted by the default formulation in ANSYS Fluent. In addition, a special user-defined function was developed to calculate the heat flux from the heat pipe walls to the calciner bubbling bed, improving the overall predictions with respect to the default DDPM model. More information on the developed DDPM model can be found in [12].

4.3.3 Development of a Solid-Solid Heat Exchanger

Four concepts for a solid-solid heat exchanger were developed to reduce the heat demand of the calciner by transferring heat from the calciner solids flow to the carbonator solids flow (see Figure 6). These concepts are: (i) a regenerative concept involving heat pipes, (ii) a concept utilizing molten salt circulating in tubes (MSHEX), (iii) a concept based on regenerative heating and cooling of a solid (RegHEX), and (iv) an L-valve concept composed of two concentric L-shaped tubes. All concepts were developed based on the same numerical equations and boundary conditions, and assessed in terms of both CAPEX and OPEX.

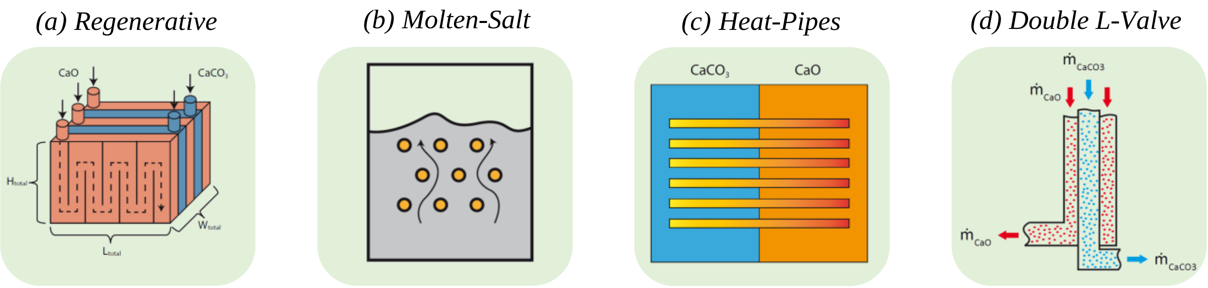

(a) Regenerative *(b) Molten-Salt* *(c) Heat-Pipes* *(d) Double L-Valve*

Figure 6. Assessed concepts for the development of a solid-solid heat exchanger.

The MSHEX concept has limited calciner inlet temperatures ($T_{s,calc,in}$ < 750 °C), making it unsuitable for its intended purpose —$T_{s,calc,in}$ > 800°C is economically crucial. The L-Valve faces similar challenges with low attainable temperatures and a substantial space requirement (up to 5000 m^3) due to suboptimal concurrent-flow heat transfer. The L-valve concept has undergone some preliminary experimental testing in a cold-flow model. For both, the MSHEX and L-valve concepts, the target temperature of 800 °C cannot be achieved. From a technical perspective, the heat pipe concept emerges as the most promising, exhibiting high Technology Readiness Level (TRL) because of the successful heat pipe operation in the 300 kW_{th} pilot plant at TUDA. However, due to the significant demand for steam, the concept incurs considerable OPEX costs, which need to be mitigated in further developments. Economic evaluations, encompassing both CAPEX and OPEX calculations, suggest that the RegHEX solution is the most cost-effective in terms of annualized costs per transferred heat flux.

4.3.4 Development of a Two-Stage Calciner

This task focused on the integration of steam into the carbonate looping process and the evaluation of possible energy savings through the use of waste heat for steam calcination. In the course of the simulation, further interconnection options were tested. It was found that the greatest energy savings could be achieved by integrating a solid-solid heat exchanger and direct steam calcination for the lime plants Hellas (5 MW) and for Hönnetal (19.5 MW).

In addition to investigating the behaviour of calcination, direct steam calcination was also investigated. During the experimental investigations, it was found that the partial pressure at the reactor outlet determines the reaction rate. From this, the reaction time and steam requirement can be calculated. Furthermore, it was shown that experimentally the theoretically calculated equilibrium line does not apply but that an equilibrium occurs for steam at a distance of 10 %, and, for nitrogen, a hysteresis occurs at a distance of 50 % from the equilibrium. Furthermore, it could be shown that calcination with steam is significantly faster due to the catalytic effect described in the literature.

Based on the results and the influence of the partial pressure on the reaction, it is clear that fluidisation must be carried out with 100 % steam (< 900 °C). CO_2 contained in the fluidisation medium prevents further release and less CO_2 can be released by the reaction. The residence time of the calcination would increase further, leading to further sintering and higher energy consumption. Sintering would have a negative effect on the performance in the carbonator.

4.3.5 Development of an Improved Heat Exchanger Arrangement

The optimal heat-pipe arrangement can enhance the heat input into the fluidised bed. To achieve this, concepts for improving heat input were be tested. In the course of addressing the task "Development of an improved heat exchanger arrangement," it was revealed that surface area is the limiting factor for the heat transfer.

Two concepts were tested. Firstly, the influence of fluidisation on the heat input was examined. Additionally, the heat-pipe arrangement was investigated and the impact of particle size was tested. To investigate the concepts, two Plexiglas fluidised beds with different heat-pipe arrangements were constructed. They included heating elements capable of heating the fluidised bed to 100 °C and a precise temperature control system. During the measurement campaign, it was demonstrated that the heat-pipe arrangement causes differences in the heat transfer coefficient of 100 $W/m^2/K$. It was also shown that the optimal heat-pipe arrangement is already utilised in the IHCaL pilot plant at TUDA. Furthermore, fluidisation has a significant impact on heat transfer. Similarly, particle size plays a crucial role; smaller particle sizes exhibit higher heat transfer coefficients than larger ones. However, it is important to note that particle size is process-dependent.

4.3.6 Development of Improved Heat Pipes

For the further development of heat pipes, two aspects should be considered: the optimization of efficiency and the improvement or examination of the material. In the course of the processing and performance tests, it was found that the limiting factor in heat transfer is not the heat pipes themselves but the surface area of the heat pipe. One option is to reduce the resistance of the heat pipe, thus improving the transfer of heat from the combustor to the calciner side. Heat pipes were manufactured with a low-resistance capillary structure, resulting in material and cost savings in the construction of the heat pipe. Consequently, this also leads to enhanced heat transfer, increasing the combustion efficiency.

As part of the task, three materials, namely 1.4841, 1.4835, and 1.4876 were analysed for their use as heat-pipe container materials. The influence of the start-up and shut-down behaviour, the conditions of the calciner and combustor side and material samples of the heat pipe itself were investigated. One challenge of the three materials is that they form phases in the temperature range of 700–800 °C, which leads to a loss of strength. In general, however, the test campaigns of the CARINA [10] and ANICA projects showed that the heat pipes work well as heat exchangers.

4.4 Process Assessment

In order to evaluate the technological risk, techno-economic performance, and environmental aspects of the IHCaL processes, the following tasks have been implemented.

4.4.1 Risk Assessment

This task focused on the assessment of possible risks of the applications of the full-scale IHCaL integration in lime and cement industries. Two assessments have been made: qualitative and quantitative. For qualitative assessment, the Failure Mode, Effect and Criticality Analysis (FMECA) has been used. It had been used with success by ESTRA in previous research projects on CO_2 capture. For the quantitative assessment, Monte Carlo simulation has been used in ANICA probably for the first time in a CO_2 capture project.

<u>Qualitative risk analysis on the lime side</u>

The highest risks detected by FMECA, on the tail end lime case of ANICA have been:

a) Stoppage of the raw material production in the limestone preparation, giving a high risk priority number but with no effect on health and safety
b) Chemical attack or corrosion on seals and parts of <u>flue gas blowers</u>, leading to possible fires or explosions, <u>causing injuries or loss of life</u>, the highest risk in the tail end concept
c) Equipment damage in the carbonator
d) Inefficient CO_2 capture in the carbonator, with a high probability of occurrence
e) Damage of construction in the combustor, with severe consequences
f) Design failure in the solid-solid heat exchanger

The highest risks detected by FMECA on the full integration lime case of ANICA have been:

a) Chemical attack or corrosion on seals and parts of flue gas blowers, leading to possible fires or explosions, causing injuries or loss of life
b) Damage in the calciner, due to overheating, causing leakage of CO_2

<u>Qualitative risk analysis on the cement side</u>

No serious risks to human life or health have been found by the participants of the cement side. The highest risks detected by FMECA on the cement side, have been:

a) Insufficient grinding of the raw material in the mill
b) Insufficient CO_2 capture in the carbonator
c) Quick sorbent decay in the carbonator
d) Inappropriate heat transfer to material in the calciner
e) Insufficient sealings in the calciner
f) Defluidisation in the combustor

<u>Quantitative risk analysis on the lime side and on the cement side</u>

Quantitative risk analysis with the Monte Carlo method is based on system dynamics simulation of the processes, involving statistical distributions to represent the behaviour of components along the time. In ANICA, Powersim Studio system dynamics simulation software was used for this analysis. Detailed information about the cement process has been provided by VDZ and for the lime processes by TUDA and ULSTER. The simulation of the processes on the lime cases and on the cement case has involved the application of appropriate statistical distributions to simulate the occurrence of events and their consequences. The conclusion from the analysis on both sides is that no serious risk exists of perturbation in one component being the cause of more serious perturbations in the other components. However, if in the future partners of ANICA decide to proceed to scale up ANICA technology, further Monte Carlo analyses of the processes would be advisable to ensure completely safe decisions.

4.4.2 Techno-Economic Evaluation of Full-Scale IHCaL Integration

ULSTER and ESTRA performed an economic assessment for the integration of IHCaL into both lime and cement plants. The process definition is summarized in Table 1.To establish techno-economic models, three reference plants were considered. For lime production, the pre-heated rotary kiln (RK) and double shaft kiln (DS) were selected. For cement production, a RK-based cement plant was chosen. The fuel options considered for the combustor were lignite, coal, and solid recovered fuel (SRF). The main economic results of are given in Table 2.

Table 1. Process definition for the process assessed in the techno-economic evaluation.

Process Number	Scenario	Description
PN1	Reference-RK-Lime plant	Reference rotary kiln lime plant using lignite as fuel for the kiln/combustor w/o CO_2 capture
PN2	Reference-DS-Lime plant	Reference double shaft lime plant using petroleum coke as fuel for the kiln/combustor w/o CO_2 capture
PN3	Reference-RK Cement plant	Reference rotary kiln cement plant using the coal as fuel for the kiln/combustor w/o CO_2 capture
PN4	Tail-End RK-Lime-lignite	Rotary kiln lime plant using lignite or SRF as fuel for the kiln/combustor with tail-end CCL carbon capture
PN5	Tail-End-RK-Lime-SRF	
PN6	Integrated-RK-Lime-lignite	Rotary kiln lime plant using lignite or SRF as fuel for the kiln/combustor with integrated CCL carbon capture
PN7	Integrated-RK-Lime-SRF	
PN8	Tail-End DS Lime	Double shaft kiln lime plant using petroleum coke as fuel for the kiln/combustor with tail-end CCL carbon capture
PN9	Integrated-DS Lime	Double shaft kiln lime plant using petroleum coke as fuel for the kiln/combustor with integrated CCL carbon capture
PN10	Tail-End-RK Cement-Coal	Rotary kiln cement plant using Coal or SRF (20% SRF: 80% Coal) as fuel for the kiln/combustor with tail-end CCL carbon capture
PN11	Tail-End-RK Cement -SRF	
PN12	Integrated-RK Cement-Coal	Rotary kiln cement plant using Coal or SRF (20% SRF: 80% Coal) as fuel for the kiln/combustor with integrated CCL carbon capture
PN13	Integrated-RK Cement-SRF	

Table 2. Main economic results.

Process Number	Installed cost increase (M€)	O&M cost (M€)	CO_2 Capture cost (€/$t_{CO2,capt}$)	CO_2 Avoidance cost (€/$t_{CO2,av}$)
PN1	--	11.2	--	--
PN2	--	3.6	--	--
PN3	--	51.8	--	--
PN4	245.9	29.7	26.5	41.1
PN5		15.4	14.2	18.4
PN6	70.1	13.5	30.2	40.2
PN7		8.2	17.4	20.2
PN8	31.1	12.4	120.7	186.9
PN9		6.0	73.8	107.9
PN10	415.3	67.0	20.7	36.3
PN11		56.5	15.8	30.6
PN12	271.9	60.3	25.3	26.6
PN13		56.1	22.1	26.1

4.4.3 Environmental Assessment via Life Cycle Analysis

This task is an environmental analysis for both lime and cement plants integrated with IHCaL using Life cycle assessment (LCA) software (SimaPro).

RK lime plant endpoint and single score results lignite:

Both capture technologies perform environmentally better than the base case with a 61% and 47% reduction in the single score. Compared to the reference case, the Human Health indicator is reduced by 45% and 60 % for the tail-end and fully integrated cases, respectively. The Ecosystems indicator is reduced by 63% and 66% for the tail-end and fully integrated cases, respectively. The Resource indicator is negative for the tail end case meaning a positive environmental impact, and the fully integrated case is 55% lower than the reference case.

RK lime plant midpoint results lignite:

The Global Warming and Mineral Resource Scarcity indicators are reduced for both capture technologies compared to the reference case. The tail-end option has a greater reduction than the integrated case. The Fossil Resource Scarcity indicator is increased for the capture technologies compared to the reference case plant. Both capture technologies use more lignite however, the tail-end case uses 5.7 times more lignite than the reference case plant. This fact also accounts for the increased electricity generation.

In each case, the introduction of carbon capture, reduced the environmental impact compared to the reference case, even when considering the increase in raw meal and fuels. Negative single scores are achieved when carbon capture is used with solid recovered fuel (SRF).

DS lime plant endpoint results:

Similar to what was seen for the RK lime set up, both capture technologies perform environmentally better for the DS set up than the base case with a 43% and 94% reduction in the single score compared to the base case. For human health, ecosystems, and resources the petroleum coke fuel has the greatest impact on the indicators followed by the unprocessed limestone. While the impact of the petroleum coke is highest for the tail end assembly, it is reduced significantly for the integrated assembly for all three indicators. The greater impact compared to the base case can be attributed to the increased consumption of petroleum coke for the integrated and tail end cases when compared to the base case.

DS lime plant midpoint results:

The Global Warming potential of the systems is decreased looking at the tail end and integrated case compared to the base case with reduction in the CO_2 emitted from the plant. Integrated has an even greater reduction than tail end. Mineral and fossil resources increase for the tail end assembly when compared to the base case due to the increased use of petroleum coke within the case. Mineral resources decrease for the integrated case compared to the base and tail end cases while also using fewer fossil resources than the tail end too.

LCA for cement plants:

The cement plant with solid recovered fuel (SRF) has an environmental benefit even without carbon capture. Deploying SRF with carbon capture increases the benefits. Electricity generation from heat generated onsite has a large impact in the favourable environmental profile and this is especially so for the cement plant with the tail-end capture technology.

Other factors should be considered. For a retrofit option, the tail-end case would be the only choice. However, for a new plant the economic assessment, including capital and operational costs, along with the environmental analysis would have to be considered. If resource reduction is a primary focus, for the hard coal fuelled plant, the integrated case would be the better option, however for the SRF option, the tail case would be better.

4.5 Design of a Fluidized Bed Demonstration Plant

4.5.1 Basic Plant Layout

Considering the large footprint and the risks of a 20 MW_{th} demo plant, it was decided to make a smaller step for the scale-up. Consequently, the demonstration plant was designed considering a fuel input of 2 MW_{th}. The heat and mass balances were calculated based on the exemplary off-gas composition from the rotary kiln line of the lime plant Hönnetal of LGE.

The extraction of the CO_2-containing flow from the lime plant was chosen to be downstream of the bag filter. The demonstration plant is connected to the production plant through the gas extraction, power line, water pipe and a sewer. The demonstration plant has its own water-cooling cycle. The fuel, sand and sorbent silos are dimensioned for four days non-stop operation. The process flow diagrams were defined (see simplified scheme in Figure 7), taken into account the results from previous tasks (see Sections 4.1 and 4.3.1).

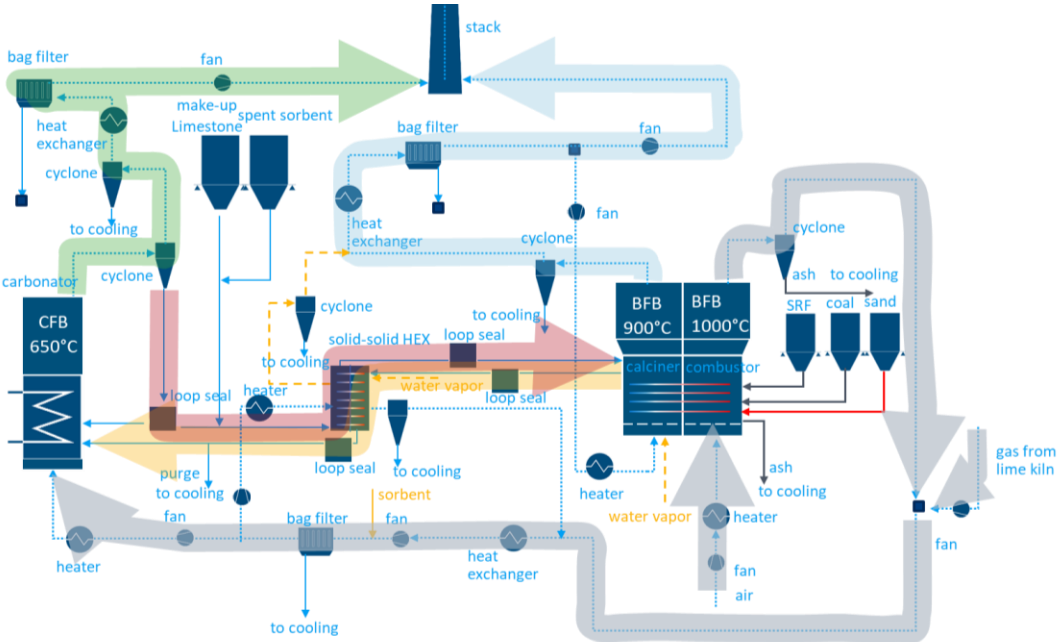


Figure 7 Simplified flow diagram of the demonstration plant

The heat for the calcination process is generated in the combustor through the combustion of coal dust and/or solid recovered fuel (SRF). The fuel burns in a sand bubbling bed reactor with preheated air. The ash in the flue gas is separated in a cyclone.

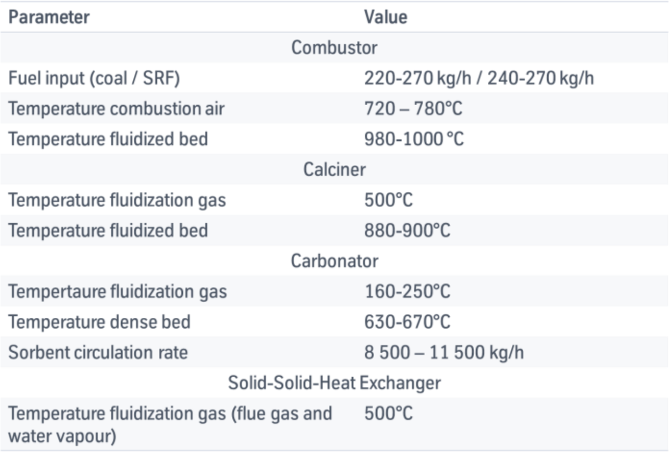
A slip stream from the lime plant is mixed with the flue gas from the combustor. The resulting gas stream is cooled down in an air-gas heat exchanger and dedusted in a bag filter. Between the heat exchanger and the filter, there is a possibility of the injection of dry sorbent for experimental purposes. The CO_2-containing flue gas is fed in the carbonator through a fan and a gas preheater. In the carbonator, the CO_2 from the gas reacts with some of the CaO-rich sorbent in the circulated fluidising bed to form $CaCO_3$. This reaction is exothermal and the excess heat is removed from the system through water cooling of the reactor. The CO_2-lean gas is dedusted in two subsequent cyclones, cooled in an air-gas heat exchanger, dedusted in a bag filter and directed to the stack.

Sorbent is extracted from the carbonator and directed together with fresh limestone to the Solid-Solid-Heat-Exchanger (SSHEX), heated up and fed to the calciner. The SSHEX is calculated as a heat pipes bubbling bed SSHEX (see Section 4.3.3), fluidised with flue gas extracted from the dedusted CO_2-rich gas. In the calciner, the sorbent containing $CaCO_3$ is fluidised with preheated water vapour and CO_2-rich gas. The gas is cooled and dedusted. One part of it is recirculated for the fluidisation of the calciner. The remaining part is directed to the stack.

The hot sorbent is fed on the second half of the SSHEX, is fluidised by water vapour, heats up the SSHEX-heat pipes and exits the SSHEX on the opposite side. Part of the $CaCO_3$-lean sorbent is extracted of the system as purge and the other part is directed to the carbonator. In this way the sorbent loop is closed. The separated sorbent and ash particles from the cyclones and the purge are cooled down in water cooled screw conveyors and pneumatically conveyed to the respective silo. The silos were dimensioned using the mass and energy balances. The filter dust from each filter is separately stored in IBC-stainless steel silo containers.

A detailed instrumentation plan with about 280 measurement devices was elaborated. As the exhaust gas of the IHCaL plant is emitted through a separate stack than the exhaust gas of the lime plant, a full emission measurement according to the German legislation BImSchV 17 is planned for its stack.

The heat and mass balances of the 2 MW_{th} plant were calculated based on the validated steady state process model in Task 3.1 for two different fuels (coal and SRF) and two different H_2O-contents of the calciner fluidisation gas. The main process parameters that result from the mass and heat balance are summarized in Table 3. In order to keep the heat pipes within the 1000 °C temperature limit, a maximum temperature of 900 °C was set for the calcination. The calcination temperature and the fluidised bed geometry define a maximum CO_2 content of the calciner fluidising gas of 30 vol%.



Parameter	Value
Combustor	
Fuel input (coal / SRF)	220-270 kg/h / 240-270 kg/h
Temperature combustion air	720 – 780°C
Temperature fluidized bed	980-1000 °C
Calciner	
Temperature fluidization gas	500°C
Temperature fluidized bed	880-900°C
Carbonator	
Tempertaure fluidization gas	160-250°C
Temperature dense bed	630-670°C
Sorbent circulation rate	8 500 – 11 500 kg/h
Solid-Solid-Heat Exchanger	
Temperature fluidization gas (flue gas and water vapour)	500°C

Table 3. Main process parameters

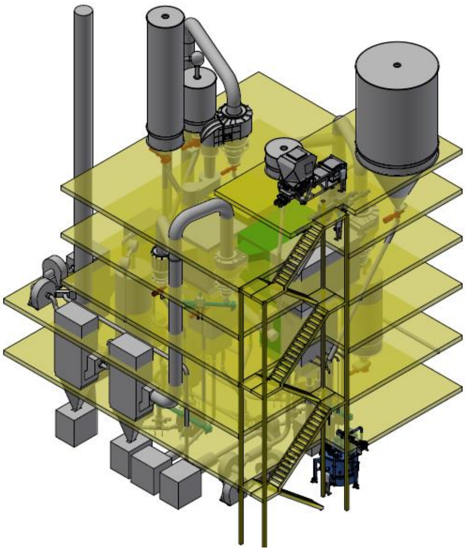


Figure 8. Demonstration plant 3D render

4.5.2 Design of the Reactor System

The **design of the reactors and the auxiliary components** was based on the mass and heat balances, and the experimental results of pilot tests. The carbonator was designed with a free gas velocity of 7 m/s. This results in a cross-sectional area of 0.37 m². The height of the carbonator results from model simulations and layout limitations. It was determined that 13m are necessary for the achievement of the retention time for the carbonation reaction and the sorbent transport between the three reactors. The calciner and the two sides of the SSHEX were designed for a free gas velocity of 0.31 m/s, and the combustor for 1 m/s. Together with the determined volume flows from the heat and mass balance, the following cross-sectional areas were determined for: the calciner 2,26 m², and each of the SSHEX-sides 1,85 m². In the calciner/combustor, there are 695 heat pipes installed, and 394 in the SSHEX, with a diameter of 33.7 mm and a length of 2.2 m, in a staggered arrangement with a pitch equal to two heat-pipe diameters. All reactors and cyclones are refractory-lined. A detailed plant layout was designed using 3D CAD software (see Figure 8). The 3D plant layout supports the cost estimation of the piping and the steel building, among others, lowering the uncertainties in the calculations.

Within this task, the performance of the calciner reactor in the scaled-up design of the 2 MW_{th} demonstration plant was verified using **CFD simulations**. The CFD analysis of the calciner provides valuable insights that can contribute to the optimization of this central component of the IHCaL process. Considering the high computational cost associated already with the 300 kW_{th} pilot plant simulations of Task 3.2, and to make the simulations of the up-scaled design feasible in terms of computational requirements, the porous media model is employed, thereby avoiding the need to create a mesh around a geometrically complex tube bundle. Specifically, it is estimated that modelling the actual heat pipes would have required a mesh of 4.5 million cells, 64 times more than the 70,000 cells finally used for the 3D porous media simulations.

Since this was one of the first applications of the porous media model to simulate a dense two-phase gas-solid bubbling flow around an immersed heat exchanger, the porous media model is first validated in a 2D representation of the calciner, by comparing its results with TFM simulations of the actual geometry containing the heat pipes. For the 3D simulations of the upscaled calciner, the TFM model used in the CFD simulations of Task 3.2 described in D3.3 is largely retained, with the added element of the porous media model to include the effect of the heat pipes. Regarding the case set-up two approaches are followed: (i) one that effectively imposes an inventory equal to the one considered by TKIS for the 0-D mass and energy balances; This is achieved by establishing a constant flux of sorbent out of the calciner equal to TKIS's considerations, and (ii) another that maintains constant pressure in this boundary. From the two, the second approach resembles more the actual conditions in the calciner, since it allows for the sorbent to freely flow out of the calciner which is what happens in real operation. For this approach (constant pressure boundary condition), two calcination reaction rates are tested, one from Labiano et al. [13] (Case B1) and another developed and calibrated by TUDA in the context of ANICA and presented in Deliverable 3.1. (Case B2) [14].

The calciner bulk bed temperature remained at around 900 °C, as designed by TKIS. The pressure drop was estimated at 170 mbar and the inventory stabilized at roughly 2930 kg on average. As for the calcination efficiency, high values in the order of 99 % are observed in both simulations, regardless of the applied calcination kinetics. As for the CO_2 production, it is estimated to be 674 kg/h and 641 kg/h for cases B1 and B2, respectively. CO_2 production is marginally lower in case B2, because the calcination rate of TUDA is slower. However, the discrepancy is minor, consistent with the calcination efficiency. This mainly happens because calcination is relatively quick, with a short characteristic time relative to the residence time of the particles in the reactor.

4.5.3 Cost Estimation

The estimations of the capital (CAPEX) and operational (OPEX) cost for the 2-MW_{th} IHCaL demonstration plant is based on the results from Task 5.1 and Task 5.2. The cost estimation corresponds to class 3 in the matrix for the process industry resulting in an expected accuracy range of -20% to +30%. The cost basis is 09.2023.

The CAPEX cost of the 2 MW_{th} demonstration plant is 31.5 M€. This includes the equipment (reactors, auxiliary systems, steel building, piping, electrical equipment, automation and instrumentation), the engineering and the project management, the erection and commissioning, and a safety factor for contingencies. The OPEX cost was calculated for a three-year period plant operation (overall 10,000 h) to 8.6 M€ and contains the following positions: personal cost for the operation of the plant, the cost of the consumables (fuel and utilities), maintenance cost.

The relatively high costs determined above are plant specific and are related to the high plant flexibility, the intensive instrumentation, the high utility and personnel costs and prototype risks and should not be used for a proportionate cost estimation of a commercial IHCaL plant.

4.6 Direct Separation–IHCaL roadmap

The main aim of the work reported in this section was to assess the potential synergies between the IHCaL and Leilac technologies when applied to full-scale cement and lime plants. There were four principal deliverables (D6.1 to D6.4) with their specific aims and results summarized in the respective sections below.

4.6.1 IHCaL-DS Basic Plant Layout

The work completed as part of D6.1 served as the foundation for all other deliverables within WP6, and fundamentally laid out how IHCaL and Leilac technologies might be integrated with cement and lime plants. Several different process layouts were developed for combination with Leilac, namely with the IHCaL technology in the (i) tail-end, and (ii) fully integrated configurations. For each configuration, a high-level block flow diagram (BFD) was generated showing the main flows of material and energy around the process.

Furthermore, the conceptual performance of each option was explored with qualitative analysis providing insight into the perceived impact of IHCaL and Leilac. Technical aspects were considered, which ranged from those related to sorbent, e.g. sorbent material, particle size distribution, fluidisation behaviour); to maintaining effective operation, e.g. the requirement for bypass to prevent impurity build-up; or other miscellaneous considerations, e.g. the location(s) and potential(s) for waste heat recovery.

4.6.2 Design of IHCaL-DS Reactor System

Continuing from D6.1, deeper consideration of the process was achieved by modelling these processes on Aspen Plus, full details of which are given in D6.2. However, for the novel cases, time constraints necessitated focus on Leilac with the tail-end IHCaL. To contextualize the discussion below, the main cases that were considered have been summarized in Table 4.

Table 4: The principal cases considered in D6.2 and D6.4

Case	Description	Design Capture Rate (%)	Integration (-)	Capture Technology (-)
1	Unabated Cement Plant	None	None	None
2	Case 1 + Amines	~90%	Tail-end	Amines
3	Case 1 + Leilac	~60%	Integrated	Leilac
4	Case 1 + IHCaL	~90%	Tail-end	IHCaL
5	Case 3 + IHCaL	~95%	Tail-end	Leilac & IHCaL

A selection of results is shown in Figure 9, illustrating the simulated thermal duty (GJ/t_{clk}) and overall avoidance rate (%) for the studied cases. The results confirm that the Leilac technology offers considerable advantages when applied for abatement of cement plants, namely offering the lowest thermal penalty (+0.3 GJ/t_{clk}) of the cases examined and significant CO_2 avoidance (70 %). Nevertheless, whilst most of the process CO_2 can be captured very efficiently, additional efforts are required to mitigate the residual emissions (mainly comprising fuel CO_2) in order to reach Net Zero. When deployed in the tail-end configuration, IHCaL appeared challenged by poor heat integration which led to excessively high thermal penalty (+8.9 GJ/t_{clk}), albeit with substantial avoidance of CO_2 (93 %). This result was largely intuitive since the tail-end configuration can only use waste heat for power generation and not for preheating meal (as in the integrated IHCaL), resulting in large amounts of *additional* CO_2 (+150 %) which thereby also required *additional* capture. However, when combined with Leilac, tail-end IHCaL enabled significantly higher avoidance rates (98 %) highlighting the potential of integrating the two technologies. The use of

Leilac reduced the quantity of CO_2 requiring abatement by IHCaL, thereby minimizing the heat integration issues highlighted above and resulting in a substantially improved thermal penalty (+4.0 GJ/t_{clk}).

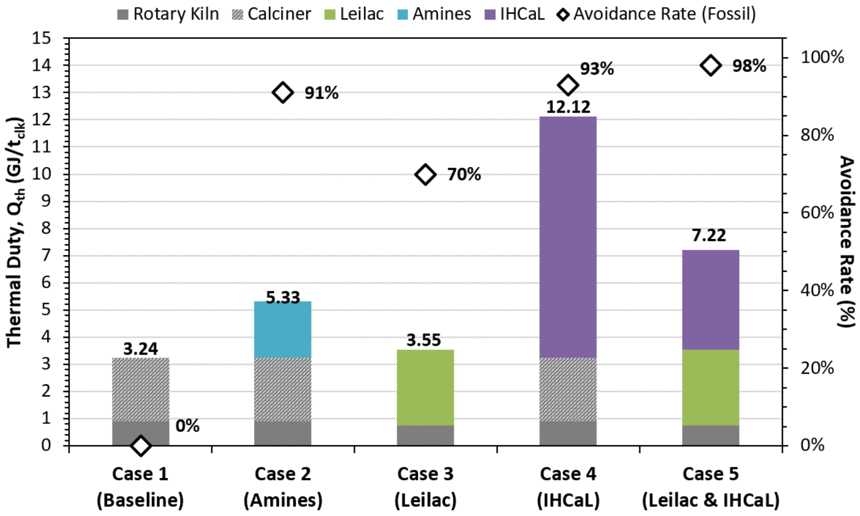


Figure 9: Thermal duty (GJ/t_{clk}) with breakdown by technology and avoidance rate (%) for the baseline (Case 1), amine scrubbing (Case 2), Leilac (Case 3), tail-end IHCaL (Case 4), and combined Leilac & tail-end IHCaL (Case 5). Avoidance rate calculated as the fraction of fossil CO_2 avoided relative to Baseline.

4.6.3 Cost Estimation and IHCaL-DS Roadmap

The studies in D6.3 aimed to provide a better understanding of the sorbent inlet conditions required for effective calcination by Leilac. A wide range of conditions were explored, with simulations varying the (i) wall temperature, (ii) inlet meal temperature, (iii) meal composition, (iv) mean particle diameter, (v) calciner throughput, (vi) partial pressure of CO_2, and (vii) specific surface area of the meal. A full account of these simulations has been given in D6.3, with an example of one provided for discussion in Figure 10.a–b which considers an isothermal calciner.

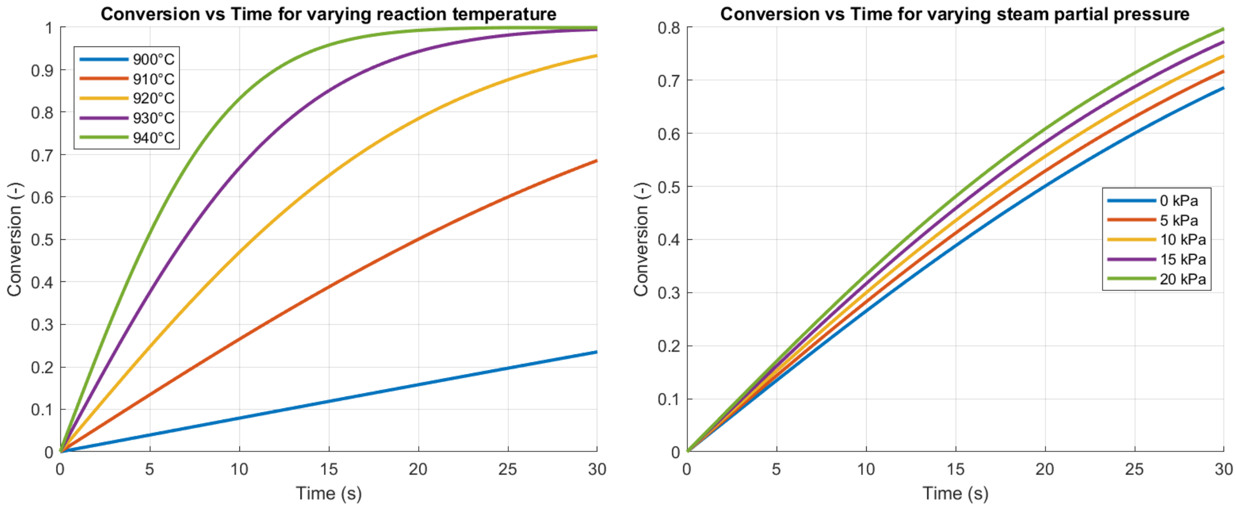
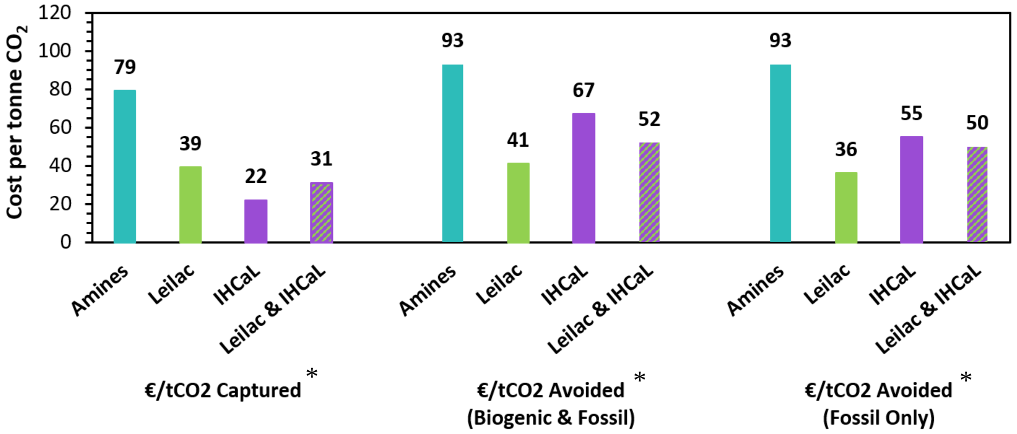


Figure 10: Calcination profiles when varying: reaction temperature (left) and steam partial pressure (right)

Results in Figure 10.a highlight the necessity to achieve more than ~920 °C within the calciner for comprehensive conversion (> 90 %) within the expected residence time, whereas Figure 10.b shows calcination can be slightly improved with relatively modest injections of steam (+5 %, when increasing from roughly 0 to 10 % steam). This is favourable since (as previously highlighted), the tail-end IHCaL technology has large potential for waste heat recovery steam generation, a portion of which could be used to boost and/or guarantee extents of calcination within Leilac. Promisingly, this is likely also true for the integrated IHCaL configuration, which also benefits from preheating of looped meal due to the exothermic carbonation reaction, also ensuring elevated meal inlet temperatures to Leilac.

Finally, a techno-economic analysis (TEA) was performed to assess the potential of each technology, focusing on the cases shown in Table 4. As previously, a full account of all assumptions has been given in D6.4, with Figure 11 only showing a selection of the main results. Some key assumptions in this analysis were more conservative than those applied in the comprehensive TEA of WP 4 (see section 4.4.2). In particular, higher costs for SRF and lower benefits from electricity export were assumed here. Because of this, there are some differences in the reported figures.

The results of the comparative TEA showed that on *per tonne captured* basis, all the capture technologies except for Amines offered competitively low costs ranging from 22–39 €/t_{CO2}. Notably, the tail-end IHCaL configuration offered excellent performance costing just 22 €/t_{CO2}, with this likely achieved in part due to (i) net export of excess power generation, and (ii) large amounts of capture of *additional* CO_2. This can be seen in the results on a *per tonne fossil avoided* basis, where the tail-end IHCaL increases to 55€/t_{CO2}. Contrastingly, the Leilac technology remains relatively consistent across this comparison emphasizing the efficiency of capture (with the trade-off being lower avoidance rates). Adoption of both Leilac and tail-end IHCaL could yield costs of just 50 €/t_{CO2} avoided whilst achieving an excellent avoidance rate (98 %, as in Figure 24). Promisingly, even better performance could likely be realized by coupling Leilac and the integrated IHCaL process.



* With conservative assumptions on fuel costs and income from electricity export.

Figure 11: Costs in terms of (i) captured CO_2, (ii) avoided CO_2 (biogenic & fossil), and (iii) avoided CO_2 (fossil only) for amine scrubbing, Leilac, Tail-End IHCaL, and combined Leilac & IHCaL (Tail-End)

4.7 Dissemination and Exploitation

4.7.1 Project Logo and Website

TUDA created a project logo (see front page) that includes the project name and a colour coding that reflects the objective of the project. The logo was used in all internal documents, presentations, deliverables, and external communication activities (e.g. public workshops, newsletters.).

Furthermore, a website was created during the first quarter of the project. The website (*https://act-anica.eu/*) was regularly updated and included information of the project, reporting of the progress, news about publications, etc.

4.7.2 Public Workshops

Two public workshops were organized throughout the ANICA project. The first workshop took place online and was hosted by TUDA. The recordings and presentation are available on the project website (*https://act-anica.eu/anica-virtual-workshop/*).

The second workshop took place at the premises of VDZ, in Düsseldorf. It was a hybrid workshop with the possibility to participate either in person or online. The workshop was co-hosted with the AC^{2}OCem project (*https://ac2ocem.eu-projects.de/*). Around 120 representatives from various countries and industries participated.

4.7.3 Publications

The publications list is provided in Chapter 8. The objective of 10 conference publications was not only achieved, but surpassed. The ANICA project was represented in the most important international conferences related to carbon capture technologies, including the Greenhouse Gas Control Technologies conference (editions 15 and 16), the second International Conference on Negative CO_2 Emissions, and the Trondheim Conference on Carbon Capture, Transport and Storage (editions 11 and 12) .

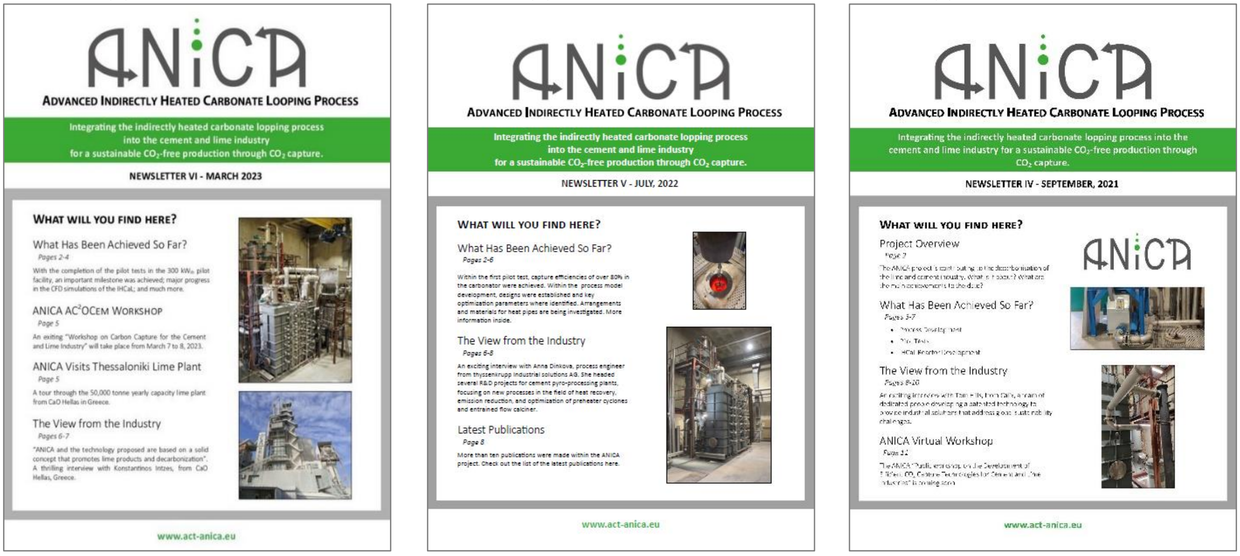
To the date of submission of this report, six publications in peer-reviewed journals are available. Two additional publications are undergoing peer-reviewing process, and two more publications are planned.

4.7.4 Industrially Oriented Newsletters

Six industrially-oriented newsletters were published. The newsletters were sent to the subscribers that registered through the project website (40 approx.). They are also publicly available on a *webpage of the project website*. The issues include information about the project results, upcoming meetings (such as the workshops), and highlights about publishing activities.

The approach adopted to write the newsletters was industrially-oriented, with focus on main results rather than detailed methodological explanations. Furthermore, five newsletters featured interviews with the ANICA partners from the industry, which focused on the view from the industry on topics such as decarbonisation of the lime and cement industry, carbon capture, and the roadmap to industrial deployment of the IHCaL process.

The newsletters included photographs of the project partners, the research facilities, and the relevant lime and cement production facilities what were analysed within the ANICA project. All the partners participated with their input. TUDA was in charge of the coordination, edition and publication of each newsletter. The cover pages of the last three newsletters are included in Figure 12, for reference.



(a) Newsletter VI

ANICA

ADVANCED INDIRECTLY HEATED CARBONATE LOOPING PROCESS

Integrating the indirectly heated carbonate lopping process
into the cement and lime industry
for a sustainable CO_2-free production through CO_2 capture.

NEWSLETTER VI - MARCH 2023

WHAT WILL YOU FIND HERE?

What Has Been Achieved So Far?
Pages 2-4

With the completion of the pilot tests in the 300 kW_{th} pilot facility, an important milestone was achieved; major progress in the CFD simulations of the IHCaL; and much more.

ANICA AC^{2}OCEM WORKSHOP
Page 5

An exiting "Workshop on Carbon Capture for the Cement and Lime Industry" will take place from March 7 to 8, 2023.

ANICA Visits Thessaloniki Lime Plant
Page 5

A tour through the 50,000 tonne yearly capacity lime plant from CaO Hellas in Greece.

The View from the Industry
Pages 6-7

"ANICA and the technology proposed are based on a solid concept that promotes lime products and decarbonization". A thrilling interview with Konstantinos Intzes, from CaO Hellas, Greece.

www.act-anica.eu

(b) Newsletter V

ANICA

ADVANCED INDIRECTLY HEATED CARBONATE LOOPING PROCESS

Integrating the indirectly heated carbonate lopping process
into the cement and lime industry
for a sustainable CO_2-free production through CO_2 capture.

NEWSLETTER V - JULY, 2022

WHAT WILL YOU FIND HERE?

What Has Been Achieved So Far?
Pages 2-6

Within the first pilot test, capture efficiencies of over 80% in the carbonator were achieved. Within the process model development, designs were established and key optimization parameters where identified. Arrangements and materials for heat pipes are being investigated. More information inside.

The View from the Industry
Pages 6-8

An exciting interview with Anna Dinkova, process engineer from thyssenkrupp Industrial solutions AG. She headed several R&D projects for cement pyro-processing plants, focusing on new processes in the field of heat recovery, emission reduction, and optimization of preheater cyclones and entrained flow calciner.

Latest Publications
Page 8

More than ten publications were made within the ANICA project. Check out the list of the latest publications here.

www.act-anica.eu

(c) Newsletter IV

ANICA

ADVANCED INDIRECTLY HEATED CARBONATE LOOPING PROCESS

Integrating the indirectly heated carbonate lopping process into the cement and lime industry for a sustainable CO_2-free production through CO_2 capture.

NEWSLETTER IV - SEPTEMBER, 2021

WHAT WILL YOU FIND HERE?

Project Overview

What Has Been Achieved So Far?

The View from the Industry

ANICA Virtual Workshop

www.act-anica.eu

Figure 12. Cover pages of the last three industrially-oriented newsletters from the ANICA project, which are publicly available on the *project website*.

4.7.5 Exploitation Plan

The results of the ANICA projects will provide CALIX with deeper insights into the challenges of eliminating CO_2 emissions from the manufacture of lime and cement, and enable them to improve their offering of the integration of the Leilac calciner into the calcium looping process. CALIX will get a stronger view on the competitiveness of our technology and the potential for increasing its competitiveness in combination with other technologies. It will enhance the potential for collaboration with other partners in the commercialization.

CERTH will apply the developed tools and knowledge on new research projects that require to model fluidised beds with an immersed heat exchanger. In the future the developed DDPM model will be used in several research projects simulating bubbling beds. As part of a long-term strategy, improvements will be made to address any challenges identified in Task 3.2.

CERTH can utilize the developed novel solid-solid heat exchanger based on the double L-valve concept and the respective models to design experiments of solid-solid heat exchangers operating during "hot" state as well as in larger scale (pilot-scale for example). This will facilitate a step towards commercialization of the technology.

DYCK expects the IHCaL process to be employed to be installed in the existing production plants with as little effort as possible. Furthermore, the sorbent, e.g. limestone or raw meal, which is used for the "carbonate looping" should be added to the cement production process after use without any loss of quality.

The proposition of application of the Monte Carlo method to any new field must be done carefully, as availability of the necessary data for such an application must be verified. However, the sort of trustworthy data that must be available for a high quality Monte Carlo risk analysis is now clear to ESTRA.

For the scale-up analysis of the process, heat pipes are to be used in quantity on an industrial scale. With its know-how and a plant manufacturer, FAU is designing a concept for industrial production. The results and innovation show how the indirectly heated carbonate looping process can be reasonably operated with steam instead of CO_2. These findings gave FAU further knowledge in calcination. The knowledge can be used in further projects, making FAU a suitable partner for the industry for indirect fluidised bed calcination.

LGE may use data from the ANICA project for integrating an IHCaL-process (either as tail-end solution or as an integrated solution) into an existing lime plant under real conditions.

The design of the demonstration plant will be the basis for (i) contacting potential customers and/or (ii) the application for a public funding for the detailed engineering and erection of a IHCaL demonstration plant based on fluidised beds.

The knowledge acquired through the process simulations and the experimental work at the 300 kW_{th} pilot plant will be used by TUDA in future research projects, training and consulting services, and education (e.g. lectures). The upgraded pilot plant may be used for further experimental investigations in following projects addressing the IHCaL process. The gained knowledge in CO_2 separation technologies may facilitate new research at TUDA in this field. The newly developed concepts for integrating IHCaL into lime plants were patented in cooperation with LGE. The newly developed concepts for a solid-solid heat exchanger may be patented.

Techno-economic assessment results can be used by ULSTER as a bench mark for the future commercialization of the IHCaL technology. The lifecycle assessment (LCA) may help to identify potential environmental issues that need to be addressed or mitigated prior commercialization.

4.8 Coordination

4.8.1 Coordination

TUDA was responsible for the coordination of the ANICA project. The main results and highlights corresponding to this task are included in Chapters 6 and 7.

4.8.2 Project Administration

TUDA was responsible for the administration of the ANICA project. The main results and highlights corresponding to this task are included in Chapter 7.

4.8.3 Management of Research Data

TUDA was responsible for the management of research data within the ANICA project. A data management strategy was implemented and reported in the corresponding deliverable (D8.1).

The data from the project was stored on a *HessenBox* server on the cloud. All the partners had access to this server and could use it for data-exchange. The documents available in the HessenBox are confidential. The public information from the ANICA project was published through the *ANICA website*.

4.8.4 Innovation Management

Innovations within the ANICA project were collected quarterly in an innovation management report. CALIX was responsible for coordinating this task.

A patent application was made with the developed concepts for the IHCaL process integration in the lime industry. The title of the patent is “Apparatus and Method for Producing Lime”, application number 10 2023 114 354.9. It is currently pending before the German Patent and Trademark Office to TUDA and LGE.

Apart from this, new models for the simulation of IHCaL components were developed by TUDA. These models, reported in the corresponding deliverables (see section 4.3.1), are useful to design and up-scale IHCaL facilities for lime and cement production with integrated CO_2 capture.

Additionally, CERTH developed a process model that can provide technical assessment of CO_2 capture processes for lime plants. This model can be used in consulting services for lime producers to evaluate their CO_2 capture schemes.

4.9 Financial Overview

In Table 5, an overview of financial progress is presented. An indicative account of the project budgets spent by the partners per work package is given. If any deviation from planned budget is stated, an explanation is given in Table 6. For the UK partners, the values in pounds were converted to euros (1£=1.14€).

Table 5 Financial progress of the ANICA project of the whole project

Partner	WP1 [k€]	WP2 [k€]	WP3 [k€]	WP4 [k€]	WP5 [k€]	WP6 [k€]	WP7 [k€]	WP8 [k€]	Total at the end of project [k€]	% of total budget(actual spent in the project)
TUDA	62.52	520	75	–	66.2	–	39	39	**801.0**	101%
FAU	–	30	178.6	17.8	30	–	15.5	–	**271.9**	100%
VDZ	541	–	–	–	–	–	9	–	**550.0**	108%
TKIS	23.52	9.80	3.04	3.92	408.53	–	8.82	–	**457.6**	114%
LGE	10.85	28.65	–	0.56	–	–	5.64	–	**45.7**	91%
DYCK	11.5	13.3	–	–	–	–	–	–	**24.7**	26%
PREZ	–	–	–	–	–	–	–	–	**0.0**	0%
ESTRA	–	–	–	202.8	–	–	–	–	**202.8**	128%
ULSTER	–	–	–	162.7	–	–	26.8	–	**189.9**	98%
CALIX	–	–	–	–	–	258	–	60	**318.0**	74%
CERTH	20.971	–	63.221	–	33.567	–	6.301	17.371	**141.4**	96%
CH	29.703	10.876	3.566	2.700	–	–	8.370	–	**55.2**	76%
TOTAL	**689**	**599**	**323**	**390**	**538**	**258**	**144**	**116**	**3058**	**101%**

Table 6 Explanation of significant divergences on financial progress

Partner	Explanation
TUDA	–
FAU	–
VDZ	–
TKIS	The potential host plant for the demonstration plant was changed after a first estimate of the reactor size was calculated. Respectively the heat and mass balances had to be recalculated. The engineering complexity of the pilot plant raised due to the need of a solid-solid heat exchanger.
LGE	–
DYCK	Since pilot tests with cement raw meal were not possible, much less laboratory analyses were required.
PREZ	Type of waste-derived fuel produced by PREZ was not suitable for the pilot plant.
ESTRA	The risks assessment required more resources than originally planned.
ULSTER	–
CALIX	Calix commercial expansion demanded more work-force as originally planned. The ANICA work was completed after budget period had ended.
CERTH	–
CH	There was lower equipment depreciation than expected due to the delay in the approval of the project from the Greek authority. Moreover, COVID 19 restrictions prevented most of the travelling. Because of this, around 25% of the original budget was not spent.

The funding information per country and type of funding is provided in Table 7.

Table 7. Financial overview per country and type of funding

Country	ACT funding	Other public funds	Private funding, R&D institution	Private funding, industry	In-kind, R&D institution	In-kind, industry	Other funds	Total at the end of project per partner	Total at the end of project per country
Germany (k€)	1638	0	0	0	165	348	0		2151
TUDA	801.0	–	–	–	–	–	–	801.0	
FAU	271.9	–	–	–	–	–	–	271.9	
VDZ	385.0	–	–	–	165.0	–	–	550.0	
TKIS	180.0	–	–	–	–	277.6	–	457.6	
LGE	–	–	–	–	–	45.7	–	45.7	
DYCK	–	–	–	–	–	24.7	–	24.7	
PREZ	–	–	–	–	–	–	–	0.0	
UK (k€)	554	0	0	0	0	156	0		711
ESTRA	142.0	–	–	–	–	60.84	–	202.8	
ULSTER	189.9	–	–	–	–	–	–	189.9	
CALIX	222.6	–	–	–	–	95.4	–	318.0	
Greece (k€)	141	0	0	0	0	55	0	141	197
CERTH	141.4	–	–	–	–	–	–	141.4	
CH	–	–	–	–	–	55.2	–	55.2	

4.10 Deliverables

A list of the deliverables written within the ANICA project is provided in Table 8.

Table 8. List of deliverables from the ANICA project

No.	Title	Lead	Date received
D1.1	Preliminary concept for integrating IHCaL into a lime plant	TUDA	28.12.2020
D1.2	Characterization of spent sorbent regarding utilization in the lime process	LGE	15.09.2023
D1.3	Final concept for integrating IHCaL into a lime plant	TUDA	25.10.2023
D1.4	Preliminary concept for integrating IHCaL into a cement plant	VDZ	28.01.2021
D1.5	Characterization of spent sorbent regarding utilization in the cement process	VDZ	31.12.2023
D1.6	Final concept for integrating IHCaL into a cement plant	VDZ	29.09.2023
D1.7	Experiments on cement raw meal for fully-integrated solution	FAU	03.04.2023
D2.1	Design of pilot plant upgrades	TUDA	15.07.2020
D2.2	Pilot test at lime plant conditions	TUDA	15.09.2023
D2.3	Pilot test of tail-end integration into a cement plant	TUDA	15.09.2023
D2.4	Pilot test of high integration into a cement plant	TUDA	15.09.2023
D3.1	IHCaL process model for lime applications	TUDA	07.07.2023
D3.2	IHCaL process model for cement applications	TUDA	15.08.2023
D3.3	CFD model using the dense discrete particle model	CERTH	03.05.2023
D3.4	Comparative assessment of solid-solid heat exchanger concepts	TUDA	16.05.2023
D3.5	Concept of a two-stage calciner for limestone	FAU	23.11.2023
D3.6	Improved heat exchanger arrangement	FAU	27.11.2023
D3.7	Improved heat pipe design	FAU	24.01.2023
D3.8	Long-term heat pipe tests	FAU	27.11.2023
D4.1	Risk mitigation plan for IHCaL integration in a lime plant	ESTRA	01.06.2023
D4.2	Risk mitigation plan for IHCaL integration in a cement plant	ESTRA	01.06.2023
D4.3	Techno-economic assessment of IHCaL integration in a lime plant	ULSTER	15.02.2023
D4.4	Techno-economic assessment of IHCaL integration in a cement plant	ULSTER	15.02.2023
D4.5	Life-cycle analysis of IHCaL integration in a lime plant	ULSTER	15.02.2023
D4.6	Life-cycle analysis of IHCaL integration in a cement plant	ULSTER	15.02.2023
D5.1	Basic process layout of the fluidized bed demonstration plant	TKIS	31.12.2023
D5.2	Basic design of the fluidized bed reactor system of the demonstration plant	TKIS	31.12.2023
D5.3	CFD simulations of the fluidized bed reactors of the demonstration plant	CERTH	10.09.2023
D5.4	Cost estimation of the fluidized bed demonstration plant	TKIS	31.12.2023
D6.1	Basic process layout of the Direct Separation – IHCaL demonstration plant	CALIX	31.12.2023
D6.2	Basic design of the Direct Separation – IHCaL demonstration plant	CALIX	31.12.2023
D6.3	CFD simulations of the Direct Separation – IHCaL demonstration plant	CALIX	31.12.2023
D6.4	Cost estimation of the Direct Separation – IHCaL demonstration plant and roadmap	CALIX	31.12.2023
D7.1	Project logo and website	TUDA	20.12.2019
D7.2	*First public workshop*	TUDA	06.10.2021
D7.3	*Second public workshop*	TUDA	08.03.2023
D7.4	*Industrially oriented newsletters*	TUDA	02.03.2023
D7.5	Exploitation plan	TKIS	31.12.2023
D8.1	Project Management Plan	TUDA	07.05.2020

5 Project Impact

5.1 Contribution to the Facilitation of the Emergence of CCUS

The ANICA project demonstrated the technical and economic feasibility of integrating IHCaL technology in lime and cement plants. This is an important milestone towards the decarbonisation of the industry through deployment of carbon capture and storage (CCS).

The experimental work, including the two pilot test campaigns in 300 kW_{th} scale, showed that developed processes are valid, and that high capture rates of over 90% are achievable. The successful operation of the combustor flue gas recirculation path demonstrated that the IHCaL concept is feasible for high decarbonization rates, including the capture of combustion CO_2 emissions. Furthermore, the results from the purged sorbent samples suggest that the use of the spent sorbent is possible.

The techno-economic assessment of the integration concepts revealed a high potential for the IHCaL technology applied to the lime and cement industries. Low CO_2 avoidance costs of around 20 $€/t_{CO2,av}$ may be achieved if solid recovered fuels (SRF) are burnt in the combustor to obtain the heat for sorbent regeneration. This is a low figure compared with the cost of other technologies for similar applications (cf. [15]).

Because of the high potential of IHCaL technology, the members of the consortium intend to upscale the process by means of a demonstration plant operating next to an existing cement or lime facility. If the next experimental steps required to assess the remaining technical questions (e.g. operating life of heat pipes) are successful, the IHCaL carbon capture technology may become commercial by early 2030. This would be a significant contribution to the decarbonisation of the cement and lime industries, which are responsible of the majority of the industrial CO_2 emissions worldwide.

5.2 Strengthen the Competitiveness and Growth of European Companies

The cement and lime industry needs to be carbon neutral by 2050. Due to the unavoidable process CO_2 emissions, CCUS is necessary to achieve this target.

Within the ANICA project, important European companies from the lime and cement sectors (LGE, CAO, DYCK) worked together to develop IHCaL technology. The association for the German cement industry (VDZ) was another relevant partner of this project. Project results were published in important scientific journals for advances on CO_2 capture (e.g. *Fuel*, and *International Journal of Greenhouse Gas Control*), as well as in conferences relevant for academic and industrial stakeholders (e.g. Symposium on UK-Lime Research, and Greenhouse Gas Control Technologies conference). Furthermore, the two public workshops involved lime and cement producers, raising awareness on carbon capture and the IHCaL technology.

The successful deployment of the IHCaL process in commercial plants would allow to drastically reduce CO_2 emissions from industrial sources without incurring high economic penalties from escalating CO_2 taxes. This is crucial for the competitiveness of European companies.

5.3 Other Environmental or Socially Important Impacts

The high potential of the IHCaL technology with respect to environmental impact was verified with the life cycle analysis (LCA) (see Section 4.4.3). The technology not only performs well in terms of economic indicators, but also regarding resources saving, reduction in global warming potential, and improved human health. The best results were achieved with the concepts using solid recovered fuels (SRF) in the IHCaL combustor. The implementation of carbon capture technologies, such as the IHCaL process, will generate new jobs and company growth for lime and cement producers, as well for equipment suppliers.

Regarding the role of the ANICA project in CCUS public acceptance, dissemination activities for the general public were performed. This included publishing six public newsletters, posting the project progress on LinkedIn, and hosting two public workshops.

5.4 Chances for Commercializing the Technology Further

The high potential of IHCaL makes it a possible candidate among CO_2 capture technologies for implementation in the commercial scale. The economic indicators (see Section 4.4.2) reveal the competitiveness of the IHCaL process. The participation of industrial partners, as well as the implication of the cement and lime industry (see Section 4.7), were key factors for the success of the ANICA project and to boost the technology towards commercialisation.

LGE is one candidate to be a future host of a full-scale IHCaL plant. LGE gained important information on the operability of the pilot plant and see the technology as a possible candidate for decarbonisation. Considering the results from purged material analysis, they see a new market opportunity arising for the commercialization of fine purged material from the IHCaL process. Lhoist may host the first IHCaL demonstrator in one of their production sites. Such a demonstrator is a necessary step to enable the commercial implementation of IHCaL technology in the lime industry.

DYCK is an important cement producer that may implement the IHCaL technology to capture CO_2 emissions from cement kilns in the future. DYCK is interested in further investigations into this technology. This includes the installation of an IHCaL demonstrator in an industrial environment in order to further advance the technological and commercial maturity.

5.5 Gender Issues

In the frame of the ANICA consortium, there were no known or documented gender equality issues. A total of 33 men and 10 women participated in the ANICA consortium, without counting students.

6 Implementation

The ANICA project addressed one of the three **SET-Plan**'s key objectives for CCUS R&I, namely the reduction of CO_2 capture costs [16]. The cost factor of CCS is decisive to enable commercial deployment. It is a key challenge to reduce the costs and the energy penalty associated with the carbon capture. To respond to this issue, pilot projects that demonstrate the technologies are required [16].The development of next-generation CO_2 capture technologies, such as the IHCaL process, correspond to the sixth R&I Activity of the 2017 SET-Plan [17].

Through process development tasks within the ANICA project (see Section 4.1), process integration options to increase the energy efficiency were identified. The low values of specific primary energy consumption per CO_2 avoided (*SPECCA*) of the lime integrated configurations indicate low energy penalties associated with the CO_2 capture using IHCaL technology (0–2.5 $MJ_{LHV}/t_{CO2,av}$). The results from the process development were published in international conferences, public workshops, and peer-reviewed journals. A collaborative patent (TUDA-LGE) was submitted.

The synergies with the lime and cement industry, as well as the efficient energy utilization, generate a cost reduction in the IHCaL capture facilities. For lime and cement plants, low costs of CO_2 avoided are achievable if solid recovered fuel is deployed to generate the heat for the calcination ($CCA < 25$ €/$t_{CO2,av}$). In this sense, the IHCaL is competitive against other technologies such as amine scrubbing or membrane-assisted CO_2 liquefaction, which incur much higher cost penalties ($CCA > 60$ €/$t_{CO2,av}$) [15].

The IHCaL process was validated within two pilot campaigns. In this way, the promising performance predicted by model estimations was supported with empirical proof from experiments in the 300 kW_{th}-scale.

The ANICA project made an important contribution in the development of the IHCaL CO_2 capture technology, which may be a key asset to strongly reduce the CO_2 emissions of two carbon-intensive industries: cement and lime. In this line, the project addressed the *Net-Zero Industries Mission* within the **Mission Innovation** initiative [18]. It was even shown that the developed processes have the potential to enable net-negative lime plants [2].

A total of five **industrial partners** were involved in the ANICA project: two from the lime industry (LGE, CH), one from the cement industry (DYCK), and two technology providers (TKIS and CALIX). LGE, CH and DYCK were involved in the process development as well as the assessment of solid samples from the pilot tests. TKIS lead the design of the IHCaL demonstration plant for the technology scale-up, and CALIX was de main responsible of developing the roadmap for IHCaL deployment combined with Direct Separation technology. Other companies from the **industrial sector** were involved through dissemination activities, such as the bi-annual newsletters, and the two public workshops (see Section 4.7 and Chapter 8).

7 Collaboration and Coordination within the Consortium

The ANICA consortium was composed of twelve partners from three European countries, namely Germany, United Kingdom, and Greece. TUDA was responsible for the project coordination.

In every work package, at least two of the three nationalities were represented. Collaboration was crucial for the success of the project. To ensure regular communication among the partners, monthly online *Steering Committee (SC) Meetings* were organized, as well as two in-presence[2] *General Assembly (GA) Meetings* per year. The SC-Meetings were one-hour meetings used to discuss main results and organization issues. The GA-Meetings lasted one or two days. They were useful to present and examine results in detail, discuss project deviations, and explore collaboration opportunities between partners.

To ensure a proper coordination of the dissemination activities, publication plans were informed in advanced and recorded by TUDA. TUDA kept the lists of publications (included planned publications), which were reported quarterly within the *Traffic Light Reports*. Many publications from the ANICA project were collaborative works from partners from different countries, which highlights the importance of trans-national collaboration within the project (see Section 8.1).

The innovation management plan was regularly updated by CALIX with input from all partners. This ensured that all relevant innovations were properly assessed. The reported innovations included scientific and technical knowledge, products, and services susceptible to be exploited.

Apart from the collaborations within the consortium, collaborations with other ACT projects were realised. These included the participation of the AC^{2}OCem project (*https://ac2ocem.eu-projects.de/*) in the first ANICA public workshop with a presentation, as well as the co-hosting of the second public workshop together with the AC^{2}Ocem consortium (*https://act-anica.eu/anica-ac2ocem-workshop-on-carbon-capture-for-the-cement-and-lime-industry/*).

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[2] The General Assembly Meetings before 2021 were held online due to COVID travel restrictions.

8 Dissemination Activities

The partners of the ANICA consortium were active in publishing results from the ANICA project. The publications included oral and poster presentations in international conferences, participation in workshops as speakers, periodic publication of newsletters, activity in social media, and publication of scientific articles in peer-reviewed journals.

8.1 Publications

The participation in conferences and workshops is detailed in Table 9 and Table 10. More than 30 presentations were made, including oral and poster presentations. Some participations were focused in the academic aspects of the investigations (e.g. International Conference of Greenhouse Gas Control Technologies), while others were aimed to broader audiences, including stakeholders from the cement and lime industry (e.g. public workshops).

A list of the articles published in peer-reviewed academic journals is given in Table 11. To the date, seven peer-reviewed journal publications with results from the ANICA project are available. These include publications in well-established journals for advances in CO_2 capture such as *Fuel* and the *International Journal of Greenhouse Gas Control*. Apart from the published articles, one additional article is currently undergoing peer-reviewing and will be published soon. Further publications are planned for the year 2024.

Table 9. List of oral presentations in conferences and workshops

Conference	Presentation Title	Partners*	Date
Carbon Capture and Storage in the cement industry, Wiesbaden	ANICA project—Advanced Indirectly Heated Carbonate Looping Process	TUDA	5.12.2019
8th High Temperature Solid Looping Cycles Network Meeting, Geleen	Advanced CO_2 capture from lime and cement plants by integration of an indirectly heated carbonate looping process	TUDA	20.02.2020
15th Greenhouse Gas Control Technologies	Efficient CO_2 Capture from Lime Production by an Indirectly Heated Carbonate Looping Process (*full paper available*)	TUDA	16.03.2021
11th Trondheim Conference on CO_2 Capture, Transport and Storage, Trondheim	CO_2 Capture from Lime and Cement Plants using an Indirectly Heated Carbonate Looping Process—The ANICA Project	TUDA	21–23.06.2021
ANICA-Workshop on Advanced CO_2 Capture Technologies For Cement and Lime Industries	Integration of the IHCaL Process into Lime Plants	TUDA	6.10.2021
	IHCaL Pilot Testing at the TU Darmstadt	TUDA	6.10.2021
	Integration of the IHCaL Process into Cement Plants	VDZ	6.10.2021
	Experimental Characterization of Cement Raw Meal for Application in the IHCaL Process	FAU	6.10.2021
	Integration of the Direct Separation into the IHCaL Process	CALIX	6.10.2021
Symposium on UK-Lime Research	Technical and Environmental Analysis of Calcium Looping Carbon Capture for Rotary Kiln Lime Plants	ULSTER	13.10.2021
13th European Conference on Industrial Furnaces and Boilers (INFUB-13),	Adaption of a 300 kW_{th} Pilot Plant for Testing the Indirectly Heated Carbonate Looping Process for CO_2 Capture from Lime and Cement Industry	TUDA	04.2022

Conference	Presentation Title	Partners*	Date
24th International Conference on Fluidized Bed Conversion	Operation of a 300 kW_{th} Indirectly Heated Carbonate Looping Pilot Plant for CO_2 Capture from Lime Industry	TUDA	05.2022
	CFD modelling of an indirectly heated calciner reactor, utilized for CO_2 capture, in an Eulerian framework	CERTH	05.2022
	Development and numerical investigation of a DDPM-KTGF model for modeling flow hydrodynamics and heat transfer phenomena in a bubbling calciner reactor	CERTH	05.2022
2nd International Conference on Negative CO_2 Emissions	Negative CO_2 Emissions in the Lime Production Using an Indirectly Heated Carbonate Looping Process	TUDA	06.2022
14th International Conference on Applied Energy	Reducing CO_2 Emissions from Lime Plants. A Techno-economic and Environmental Assessment (*full paper available: energy proceedings*)	ULSTER	10.08.2022
16th International Conference on Greenhouse Gas Control Technologies	Pilot Testing of the Indirectly Heated Carbonate Looping Process for Cement and Lime Plants	TUDA	10.2022
12th Mediterranean Combustion Symposium MCS-12	Pilot Testing of the Indirectly Heated Carbonate Looping Process for CO_2 Capture From Lime Industry	TUDA	01.2023
ANICA-AC2OCem Workshop on Carbon Capture for the Cement and Lime Industry	Pilot Testing of the IHCaL Process	TUDA	03.2023
	Reactor Development for IHCaL Technology	FAU	
	Scale-Up of the IHCaL Process for the Lime Production	TKIS, TUDA	
	Techno-Economic Assessment of IHCaL Integration in Lime and Cement Plants	ULSTER	
	Capturing Unavoidable Carbon Emissions in the Cement and Lime Industry	CALIX	
IEAGHG 9th High Temperature Solid Looping Cycles Network	Results of 300kW_{th} IHCaL pilot plant	TUDA	03.2023
Jahrestreffen der DECHEMA Fachgruppe Hochtemperaturtechnik	Influence of steam on the calcination reaction	FAU	03.2023
Fluidization XVII 2023, Edinburgh	Performance of a Limestone-Based Coupled Fluidized Bed Reactor System Aiming CO_2 Capture in a 300 kW_{th} Pilot plant	TUDA	05.2023
12th Trondheim Conference on CO_2 Capture, Transport and Storage, Trondheim	Efficient CO_2 Capture from Lime Plants: Techno-economic Assessment of Integrated Concepts using Indirectly Heated Carbonate Looping Technology	TUDA	06.2023
13. Österreichisches IEA Wirbelschichttreffen, Wien	Wasserdampf in der Kalkherstellung/ Kalzinierung	FAU	20–22.09.2023
7th Post Combustion Capture Conference, Pittsburgh (PA)	Design of a 2 MW_{th} IHCaL demonstration facility at a lime plant in Germany	TUDA, TKIS, LGE, FAU	27.09.2023

* The affiliation of the presenter is shown first, followed by the rest of the affiliations, corresponding to the authors list.

Table 10. List of poster presentations in conferences and workshops

Conference	Poster Title	Partners*	Date
Trondheim Conference on CO_2 Capture, Transport and Storage	Process Integration of Indirectly Heated Carbonate Looping in Lime Plant for enhanced CO_2 Capture	CERTH, CH	21–23.06.2021
Fluidization XVII 2023, Edinburgh	Proof of concept calcination kinetic in TGA and fluidized bed reactor	FAU	05.2023
	Proof-of-Concept of a Novel Solid-Solid Heat Exchanger Based on a Double L-Valve Concept	CERTH	05.2023

* The affiliation of the presenter is shown first, followed by the rest of the affiliations, corresponding to the authors list.

Table 11. List of peer-reviewed journal publications

Authors and Title	Journal	Partners	Date
M. Greco-Coppi et al., *Efficient CO_2 Capture from Lime Production by an Indirectly Heated Carbonate Looping Process*	International Journal of Greenhouse Gas Control; SI GHGT-15	TUDA, LGE	09.2021
G. Kanellis et al., *CFD modelling of an indirectly heated calciner reactor, utilized for CO_2 capture, in an Eulerian framework*	Fuel	CERTH	04.2023
M. Greco-Coppi et al., *Negative CO_2 Emissions in the Lime Production Using an Indirectly Heated Carbonate Looping Process*	Mitigation and Adaptation Strategies for Global Change; SI: 2nd Int. Conf. Negative CO_2 Emissions	TUDA, LGE	06.2023
G. Kanellis et al., *Development and numerical investigation of a DDPM-KTGF model for modeling flow hydrodynamics and heat transfer phenomena in a bubbling calciner reactor*	Fuel	CERTH	06.2023
Ch. Papalexis et al., *Proof-of-Concept of a Novel Solid-Solid Heat Exchanger Based on a Double L-Valve Concept*	Energies	CERTH	08.2023
C. Hofmann et al., *Enhancement of a 300 kW_{th} Pilot Plant for Testing the Indirectly Heated Carbonate Looping Process for CO2 Capture from Lime and Cement Industry*	Experimental Thermal and Fluid Science; SI MCS-12	TUDA	11.2023
M. Greco-Coppi et al., *Efficient CO_2 Capture From Lime Plants: Techno-Economic Assessment of Integrated Concepts Using Indirectly Heated Carbonate Looping Technology*	Carbon Capture Science & Technology; SI: TCCS-12	TUDA, ULSTER, ESTRA	12.2023
S. Rezvani et al., Techno-economic-analysis of indirectly heated carbonate looping cycles for CO_2 sequestration within full-scale cement plants	International Journal of Greenhouse Gas Control	ESTRA, ULSTER, FAU	In review
M. Greco-Coppi et al., The Impact of the Calcination Rate in the Indirectly Heated Carbonate Looping Process—Development of a Rigorous Carbonator Model with Experimental Validation	Chemical Engineering Journal	TUDA	Planned (2024)
M. Greco-Coppi et al., Modelling of the Bubbling Bed Calciner of an Indirectly Heated Carbonate Looping Process for Efficient CO_2 Capture	Fuel	TUDA	Planned (2024)

8.2 Newsletters, Website Articles, and Other Activities

Apart from the publications in peer-reviewed journals, and the involvement in international conferences and workshops, there were other dissemination activities focused on the lime and cement industry, as well as the general public (see Table 12, Table 13, and Table 14). These contributions were not only intended to disseminate the ANICA project and the IHCaL technology, but also to raise awareness on the necessity of CCUS in the lime and cement production.

The dissemination activities included the publication of six industry-oriented newsletters. The newsletter featured photographs from the test facilities and project highlights for the general public and the industry. Apart from this, the ANICA project had a presence in the social media through LikedIn. The project was also disseminated through the project website (*https://act.anica.eu*) and the information on the institutional websites of the ANICA partners.

Table 12. List of newspaper or magazine articles published

Newspaper/Magazine, Article	Partner	Date
ANICA Newsletter No. 1	TUDA	30.04.2020
ANICA Newsletter No.2	TUDA	05.11.2020
ANICA Newsletter No.3	TUDA	28.06.2021
ANICA Newsletter No.4	TUDA	01.10.2021
ANICA Newsletter No.5	TUDA	01.07.2022
ANICA Newsletter No.6	TUDA	02.03.2023

Table 13. List of website articles or postings

Website, Group	Partner	Date
www.researchgate.com ANICA ACT project, Advanced Indirectly Heated Carbonate Looping Process	TUDA	until 31.03.2023
www.linkedin.com Hashtag: #ANICAact	TUDA	ongoing
ANICA Website: *https://act.anica.eu*	TUDA	ongoing

Table 14. List of activities

Activity	Partner	Date
ANICA logo included on company website and in relevant brochures	LGE	ongoing
ANICA project description included on company website	LGE	ongoing
ANICA logo and project description included on company website	VDZ	ongoing
ANICA newsletters linked on the company website	VDZ	ongoing

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