



ADVANCED INDIRECTLY HEATED CARBONATE LOOPING PROCESS

Integrating the indirectly heated carbonate looping process into the cement and lime industry for a sustainable CO₂-free production through CO₂ capture.

NEWSLETTER IV - SEPTEMBER, 2021

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Project Overview

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The ANICA “Public workshop on the Development of Efficient CO₂ Capture Technologies for Cement and Lime Industries” is coming soon.



PROJECT OVERVIEW

ANICA is an ACT project focused on developing novel integration concepts of the state-of-the-art indirectly heated carbonate looping (IHCaL) process in cement and lime production. The project aims at lowering the energy penalty and CO₂ avoidance costs for CO₂ capture from lime and cement plants. Within 36 months, the project brings the IHCaL technology to a high level of technical maturity by carrying out long-term pilot tests in industry-relevant environments and deploying accurate 1D and 3D simulations.



WHAT HAS BEEN ACHIEVED SO FAR?

So far, concepts for the integration of the IHCaL process into existing lime plants in Hönnetal (Lhoist Group) and Thessaloniki (CaO Hellas) have been developed (see Newsletter III, page 3). The corresponding one-dimensional simulations were successfully carried out. The first results were published at the GHGT-15 Conference (Greco-Coppi et.al.). Further results were presented in the 11th Trondheim Carbon Capture & Storage Conference.

In parallel, VDZ assessed concepts for the high level integration of the IHCaL process into a BAT (Best available technic) -cement plant.

Regarding the experimental work, some experimental results are already available and fascinating tests are being prepared right now at TUDA and FAU. Long-term test-campaigns at TUDA will take place towards the end of the year 2021 in the 300kW_{th} IHCaL testing facility.

Furthermore, the first direct separation concepts for cement production are available, and the up-scaling works for an industrial-scale IHCaL facility are being performed, including technical and economical analysis as well as risk assessments with Monte Carlo simulations.

Finally, important results from the 1-D and 3-D simulations were produced. More information on the CFD simulations can be found on Newsletter III, page 4.

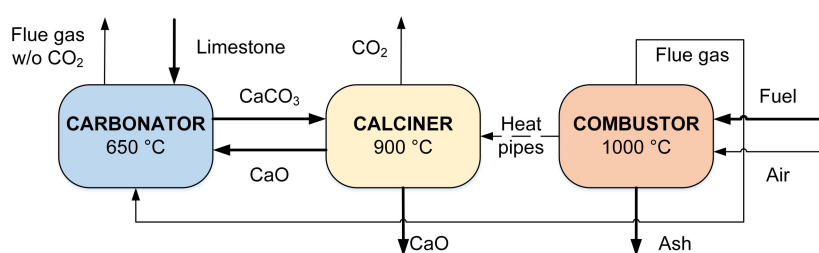


Figure1: IHCaL process flow diagram

K. Peloriadi, K. Atsonios, A. Nikolopoulos, K. Intzes, G. Dimitriadis, N. Nikolopoulos, *Process Integration of Indirectly Heated Carbonate Looping in Lime Plant for Enhanced CO₂ Capture*.

TCCS-11 – Trondheim Conference on CO₂ Capture, Transport and Storage : June 21–23, 2021, p. 529-535, Trondheim, Norway.

ISBN: 978-82-536-1714-5

J. Ströhle, C. Hofmann, M. Greco-Coppi, B. Eppe, *CO₂ Capture from Lime and Cement Plants using an Indirectly Heated Carbonate Looping Process – The ANICA Project*.

TCCS-11 – Trondheim Conference on CO₂ Capture, Transport and Storage : June 21–23, 2021, p. 529-535, Trondheim, Norway.

ISBN: 978-82-536-1714-5

M. Greco-Coppi, C. Hofmann, J. Ströhle, D. Walter, B. Eppe, *Efficient CO₂ Capture from Lime Production by an Indirectly Heated Carbonate Looping Process*.

Abu Dhabi UAE, 15th International Conference on Greenhouse Gas Control Technologies GHGT-15.

DOI: <http://dx.doi.org/10.2139/ssrn.3817331>

PROCESS DEVELOPMENT

Preliminary concepts were developed for the integration of the IHCaL process into lime and clinker burning process. By implementing the process, priority was always given to high plant efficiency with maximum utilization of thermal energy.

LIME BURNING PROCESS

Two concepts for the integration of the IHCaL process into the lime process were developed. One concept corresponds to a tail-end configuration, in which the IHCaL facility for the carbon capture is attached to an existing plant as an end-of-pipe addition. The second concept (see Figure 2) consists of a highly integrated solution, suitable for entirely new facilities. The design and integration of these plants was based on a reference lime plant of LGE in Hönnetal, Germany.

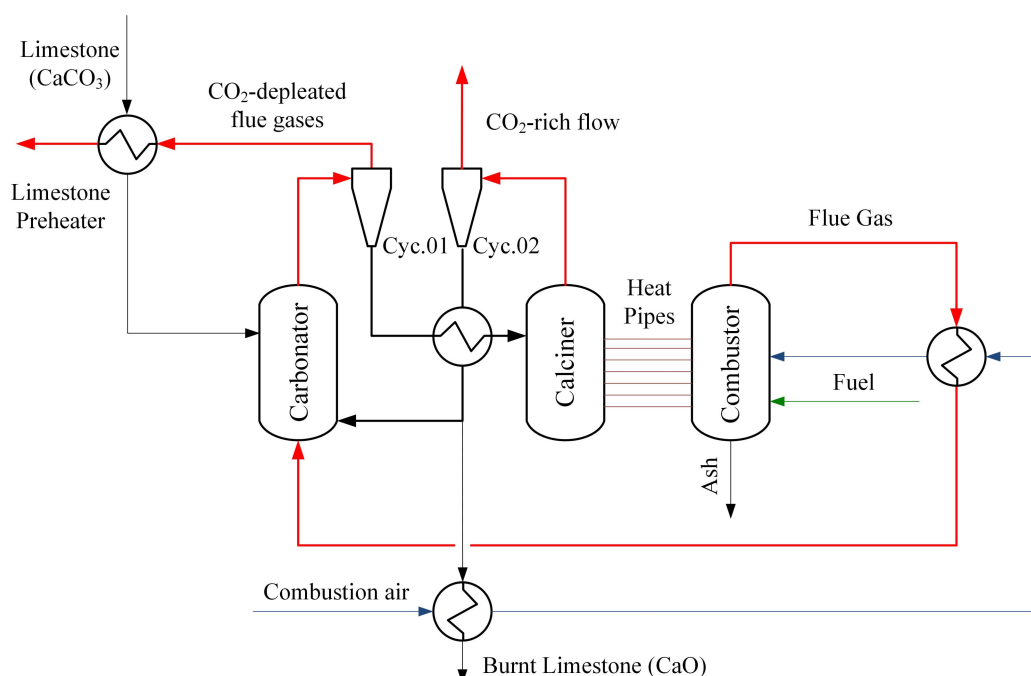


Figure 2: Process flow diagram of the integrated IHCaL process for lime production and carbon capture.

The corresponding numerical models in Aspen Plus were developed and the energy & mass balances were performed. Furthermore, a sensitivity analysis was carried out to assess the influence of the main process parameters. Critical points for integration are the preheating of the combustion air, the efficiency of the sorbent solid-solid heat exchanger, and the utilization of the sorbent purge as lime product. The first results were published in M. Greco-Coppi et al. 2021. The developed models and the obtained results will be used to further develop the integration of the IHCaL into lime plants by experimental and numerical methods.

CLINKER BURNING PROCESS

Preliminary concepts for the integration of the IHCaL process into clinker burning plants were developed. Compared to the lime burning process, the raw material mixture in the clinker burning process consists not only of pure limestone,

but of other components. Portland cement clinker is produced from a raw material mixture consisting mainly of calcium oxide (CaO), silicon dioxide (SiO₂), aluminium oxide (alumina (Al₂O₃) and iron oxide (Fe₂O₃). These chemical constituents are supplied by limestone, chalk and clay or their natural blend, marl. Clay minerals and feldspar are compounds of aluminium oxide and silicon dioxide (aluminosilicates) with alkalis, such as sodium and potassium. The iron oxide required for melt formation is either contained in the clay minerals in the form of ferrous hydroxide or it is added in the form of iron ore. To ensure that the cement meets the required quality standards, a precisely defined raw material composition must be maintained.

Due to the complexity of the process, the preliminary concepts have been continuously revised and enhanced on basis of experimental work and literature data, in order to find a validated process design. For this purpose, the existing VDZ process model for the clinker burning process was adapted to the new requirements. Figure 3 shows an example of the current flow diagram of the concept variant with integrated IHCal process, which is currently being simulated in the VDZ process model on the basis of a BAT plant.

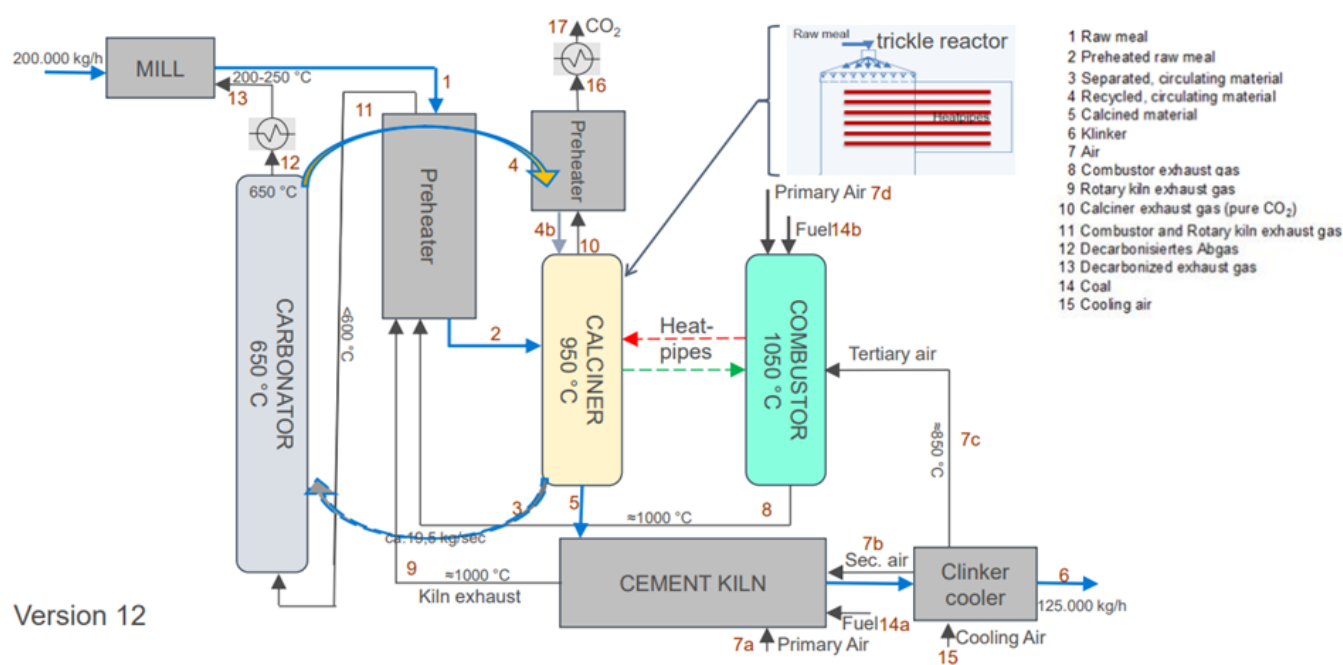


Figure 3: Flow diagram of a cement plant with integrated IHCal process.

EXPERIMENTS ON CEMENT RAW MEAL

During the calcination and carbonation tests, challenges arose with regard to reactivity, agglomeration tendency and fluidization behaviour of cement raw meal, which differed from the behaviour of pure lime, observed in previous work. Therefore, additional investigations were carried out on raw meal in fine powder and pelletized form. First agglomeration test shows that above a temperature of 820 °C the pellets begin to agglomerate, which leads to a process stop. In contrast, the fine raw meal shows no agglomeration in the required temperature range, but has problems with fluidization in terms of channel formation and material discharge. In addition, thermogravimetric investigations under CO₂ atmosphere and temperatures between 630 and 980 °C revealed reduced reactivity, which can be attributed to a low free lime content and increased belite and spurrite phase formation.

PILOT TESTS

During the first half of the project, the pilot plant was adapted to be prepared for the testing under cement and lime plant conditions. On the basis of preliminary heat and mass balances, operational modes of the existing equipment have been identified and the design conditions for the new equipment were elaborated. Concept, basic and detailed engineering of the major upgrades of the pilot plant, namely the flue gas path and solid feeding system, have been performed. In order to improve the operability of the plant, smaller modifications like purging, sampling and feeding system of the sorbent (CaO/CaCO_3) are being installed.

The major changes that have already been



Figure 4a. Updates at the pilot plant. The in-series blowers are displayed in this photograph. These are key assets for the newly installed flue gas paths.



Figure 4b. Updates at the pilot plant. The photograph depicts a detail of the new flue gas path under construction. The new cooler and the filter can be seen.

implemented and are illustrated in Figures 4a and 4b. The mechanical and electrical implementations of the upgrades are ongoing, while the cold commissioning of the main components of the flue gas path has been achieved. The boundary conditions of the first test campaign under lime plant conditions are elaborated and the pilot plant is being prepared for these tests, aiming to deliver purged sorbent (CaO) to analyse it and assess its quality for lime applications.

While using cement raw meal as sorbent, proposed for the highly integrated solution under cement plant conditions, different challenges occurred, addressed in WP 1.5. Depending on these results and additional tests in batch conditions, e.g. investigations on hydrodynamic behaviour, operational modes for using cement raw as sorbent will be identified.

In the second half of the project, the solid feeding system will be installed and pilot tests performed. First experimental results are expected by the end of the year 2021.

REACTOR DEVELOPMENT

PROCESS MODEL DEVELOPMENT

To further improve the simulation and design of the fully integrated IHCaL process, a 1D-Model of the 300kWth pilot plant reactor at TU Darmstadt is developed in Matlab and validated. This model is used to calculate the heat and mass balances of the process, which will be used to upscale the process models. Two different calcination models, one from Labiano and one from Martinez, are compared, to find the one that predicts the most accurate. First simulation results suggest that, although the calciner is fluidised by air, the amount of released CO_2 is so high, that no calcination takes place in the colder regions of the fluidised bed. For modelling the temperature distribution in the calciner, a one-dimensional steady conduction model is added to the heat transfer model. The validated model will be implemented into the already existing process model in ASPEN Plus. This will help to scale up the IHCaL process precisely.

CFD SIMULATIONS OF THE CALCINER

One of CERTH's major contributions into ANICA Project is to provide a validated transient CFD model of the 300 kW bubbling calciner. This model will be used to investigate the effect of several operating conditions (e.g. Geldart A vs. Geldart B particles, operating regimes) and heat pipes arrangements on the performance of the heat pipe heat exchanger. The simulations are conducted within the commercial platform of ANSYS Fluent, using numerous in-house built subroutines regarding the Kinetic Theory of Granular Flows (KTGF) closure terms, reaction kinetics, and drag force models.

The granular flow in the calciner is modelled following an Euler-Euler (TFM) approach. CERTH applies an in-house version of the sophisticated sub-grid EMMS (Energy Minimization Multi-Scale) drag model, able to accurately take into account the effect of the flow heterogeneity. The TFM model is validated against experimental data from the previous CARINA project, concerning heat transfer coefficient, CO_2 production and pressure profile.

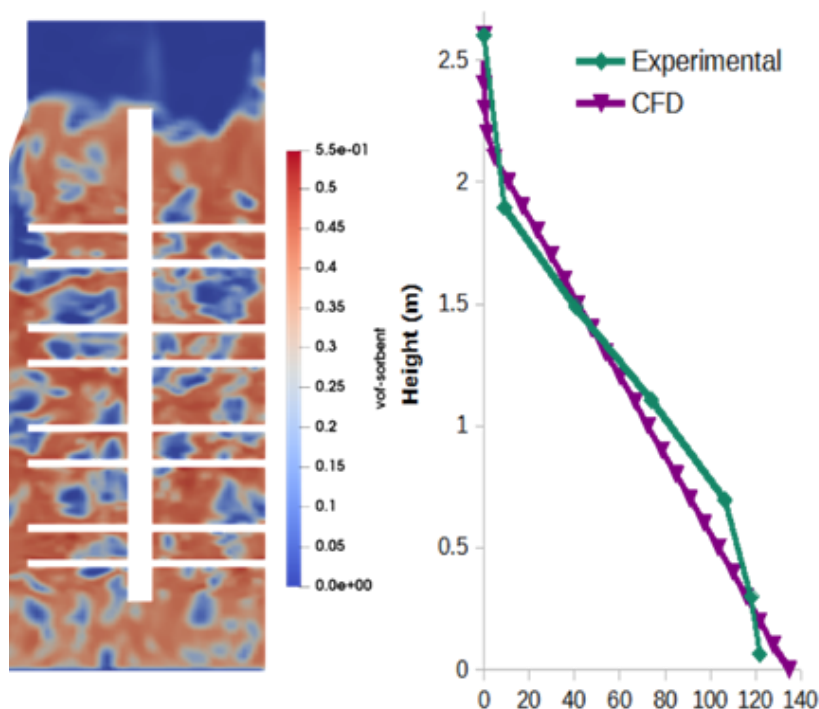


Figure 5: Euler-Euler model: Volume fraction field in the calciner (left) and pressure profile validation (right)

In parallel, CERTH is currently developing an Eulerian-Lagrangian model, i.e. the Dense Discrete Phase Model (DDPM), which utilizes closure terms from the KTGF. The main advantage of the DDPM methodology is that it is capable of incorporating a PSD into the model, which is a key parameter to improve the heat transfer and reaction modelling accuracy. However, DDPM has not been extensively validated especially for dense flows, compared to the TFM. As a result, its successful implementation in the complex calciner geometry is quite challenging, often requiring special numerical treatment.

REACTOR DEVELOPMENT

In the first half of the ANICA project, the batch calciner was successfully recommissioned. A concept for a second stage calciner was developed and adapted. Before investigations regarding the second stage calciner are conducted, first tests were carried out with limestone and cement raw meal to investigate whether also cement raw meal could be a possible sorbent in a highly integrated variant of the IHCaL. Contrary to expectations, the carbonation of cement raw meal was quite complicated in the batch calciner. In summary, three challenges can be named: (1) low reaction rates of cement raw meal with CO_2 in comparison to lime, (2) agglomerations that lead to defluidisation, and also (3) a difficult fluidisation behaviour due to a broad particle size distribution and stickiness of the cement raw meal and cement pellets.

All three challenges could become a reason that the highly integrated variant of the IHCaL is not applicable for cement raw meal. An objective of the ANICA project is to propose a realistic concept – tail-end or highly-integrated – for the IHCaL in a cement plant. Due to this, FAU decided to adjust its focus to investigate this open questions regarding reactivity, agglomeration and fluidisation of cement raw meal. A new task 1.5 is identified and the first results are shown in WP1.

DEVELOPMENT OF IMPROVED HEATPIPES

For an improved heat transfer between the calciner and combustor of the IHCaL, an optimization of the heat pipes is part of the project. In the IHCaL, horizontal heat pipes are used. The heat transfer capacity of horizontal heat pipes is limited by their capillary limit and entrainment limit. An optimization calculation was made to find the best compromise between the mesh structure for the special needs of the capillary and the vapor area. Furthermore, FAU calculated that the entrainment limit is negligible with one layer of a fine pore structure at the inside of the capillary system. Larger pores at the outside of the capillary system could make a further improvement in the backflow of the medium from the condensation to the evaporation zone. One point that should be further considered is the filling level of the working fluid in the heat pipe.

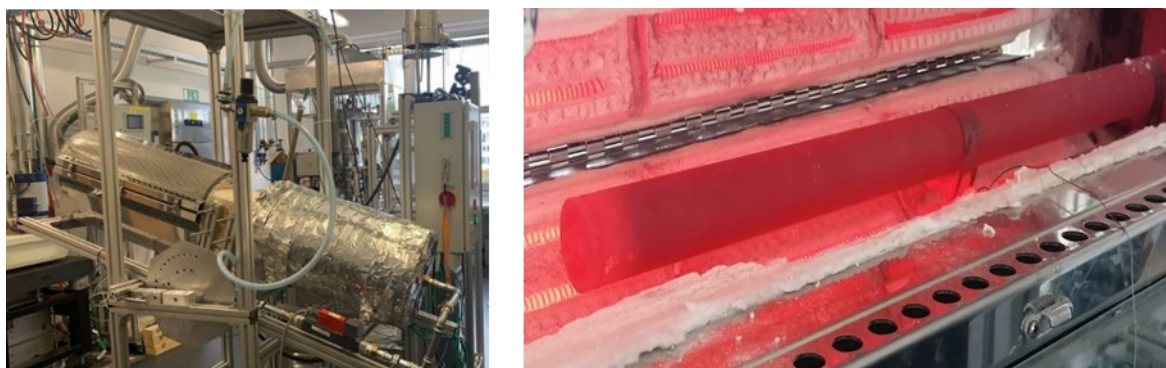


Figure 6: Picture of the test rig to measure the performance of the heat pipes (l). The test rig is rotatable to measure the capillary limits of the heatpipes. The picture (r) shows a detailed view of a heat pipe in the test rig at 800°C.

THE VIEW FROM THE INDUSTRY



CALIX

Calix is a team of dedicated people developing a patented technology to provide industrial solutions that address global sustainability challenges.

The core technology is being used to develop environmentally friendly solutions for advanced batteries, crop protection, aquaculture, wastewater, and carbon reduction.

Calix's core technology has attracted considerable recognition, and is the reason behind many of our business partnerships, including Project ANICA. Calix is also working with a number of universities, research institutes, governments and industry partners across many other projects globally.

THE INTERVIEW

Calix has a very impressive company's motto: "Mars is for quitters". Could you explain what this means, and to what extent the phrase summarizes the company's vision?

Mars is for quitters is our 'why', the reason why we exist. While all our staff brought different perspectives to bear on why they do what they do, we initially found it difficult to wrap it into a common, pithy theme that resonated with all. After a suggestion that Elon Musk's "SpaceX" had their "why" nailed – a picture of Mars – a theme emerged that quickly brought together the different perspectives, as well as reflect Calix's personality – a little bit irreverent and fun at the surface, but very serious underneath.

OUR INTERVIEWEE: TOM HILLS

Tom Hills is a Research Engineer at Calix. He is involved in the LEILAC-2, SOCRATCES and ANICA projects, amongst others, and led key work in LEILAC-1. He has previous experience in industrial carbon capture and storage, including research at Imperial College London and consultancy for industrial and governmental organisations. He holds a PhD in Chemical Engineering from Imperial College London on the topic of CCS in the cement industry.



Calix is well known in the industry and the academia for the LEILAC Project. Could you tell us about this project? What makes it so special?

Over half of the CO₂ coming from cement and lime manufacture comes from the mineral feedstocks themselves – mainly calcium carbonate (limestone), which is calcined to calcium oxide (lime). This is a highly energy-intensive process and in conventional lime and cement plants, the feedstock is injected into a combusting mixture of fuel and air which provides the temperature and energy to drive the calcination reaction. The resulting flue gas (or exhaust) is around 20–30% CO₂, most of which comes from the feedstock (also known as 'process CO₂') while most of the rest comes from the fuel (also known as 'combustion CO₂'). This stream of CO₂ and other gases is not suitable for transport and permanent storage; it must be concentrated up in a 'post-combustion capture plant'. This additional process is energy intensive and therefore expensive. If the energy used comes from carbon fuels, then more CO₂ has to be captured, adding onto the overall costs.

In Projects LEILAC 1 and 2, the fuel combustion is kept separate from the calcination. Combustion occurs in a furnace surrounding the tube, heating it up. The tube then re-radiates the heat to the limestone that's passing through on the inside. Because the furnace gases are kept separate, and the gases emanating from the feedstock are overwhelmingly (>95%) CO₂, the stream is suitable for transport and storage once cooled, de-dusted and compressed. From a process perspective, the cost of the process should be comparable to the conventional process.

In the LEILAC-1 project we designed, built and successfully operated a 10 tonne per hour pilot plant at a cement plant in Lixhe, Belgium. Despite that, it's only around 5% of the size of what would be needed at full scale application! We are now in the LEILAC-2 project, where we are building a demonstration plant in Hannover, Germany that will be around 4 times the size of the LEILAC-1 plant, and will calcine the equivalent of 20% of the main plant's feedstock. As well as scaling up, we're focusing on integrating LEILAC-2 into the main cement plant, using the same fuels. This blend of R&D and engineering will reduce the technical and financial risks associated with applying the technology commercially at full scale. Both LEILAC projects were funded through the Horizon 2020 scheme of the European Commission, and have involved an array of partners from industry, commerce and academia.

In Calix's vision, what role does the ANICA project play?

LEILAC is a great technology for capturing the process CO_2 , but it doesn't capture or directly reduce the combustion CO_2 emissions. Other solutions are required for that: zero-carbon fuels or carbon capture. That's where Project ANICA comes in. By combining IHCal with LEILAC technology, we can capture the process CO_2 emissions, as demonstrated with LEILAC, along with the combustion CO_2 emissions in the IHCal carbonator.

The inevitable question is: why not just use IHCal for all of the CO_2 , as shown in the main ANICA designs? At Calix, we believe that applying LEILAC's very low energy penalty and cost to the process emissions means that the IHCal plant for combustion CO_2 can be smaller and less intrusive. But most importantly, the overall cost of capture would be lower. This is a key driver for the ANICA project and we believe the Calix Technology make a meaningful contribution.



Figure 7: The LEILAC-1 pilot plant in Lixhe, Belgium.

It has the capacity of a small lime plant

Do you expect any synergies that could arise between the two projects, LEILAC and the ANICA Project ?

Of course! At the heart of the IHCaL process is an indirectly heated calciner; this is what distinguishes it from 'conventional' carbonate looping where the calciner is run in oxy-fuel mode. The Calix Technology, as used in LEILAC and further developed in ANICA, re-engineers the existing process flows of a traditional calciner, indirectly heating the limestone via a special steel vessel. I work on both, the LEILAC project and the ANICA project, and developments in one often benefit the other. ANICA provides one vision of zero-carbon cement and lime plants which integrate LEILAC technology. LEILAC, thanks to its larger scale and higher level of development, is encountering and solving many of the challenges that we can expect ANICA to face in the future; sharing solutions can help accelerate ANICA's development.

Why is it important that CALIX is involved in the ANICA Project ?

The European cement and lime sectors want to reach net zero by 2050. Both the cement and lime industries have high CO₂ emissions with the majority of their total CO₂ emissions being released directly, and unavoidably, from the processing of limestone (which is nearly 50% by weight CO₂). Carbon capture is the only means by which these industrial processes can dramatically reduce their emissions.

To reach >90% reductions in direct CO₂ emission, we need to combine LEILAC with other approaches, as discussed earlier. One option is to go for zero-emission fuels such as electricity, hydrogen or sustainable biomass; and in fact, electrification is a key aspect of the LEILAC2 project. Those fuels are expensive and likely to be in great demand in the coming decades, so may not be available in all cases. The other option is to continue burning carbon fuels and capture the CO₂. Developing this technology with our partners in ANICA offers one route for Calix to offer a complete near- or net-zero carbon solution for cement and lime plants. In fact, if the fuel contains a reasonable share of biomass, the process can become net-negative carbon!

What appealed to you personally in the ANICA project, sparking the interest to get involved in it ?

My PhD is in carbonate looping in the cement industry, so working on ANICA brings me full circle! Back then there was some passing interest but not much engagement on CCS in the cement industry. In the past five years industrial CCS has really come to the fore, and there is so much engagement from industry, government and academia to collaborate meaningfully to develop a suite of options for the cement and lime sectors. ANICA offers me the chance to bring my doctoral and industrial experiences together, which is really satisfying.

What is your role in the project and what expertise do you bring to the consortium ?

I lead Work Package 6, which focuses on the combination of the Calix Technology into the IHCaL process. Together with a team of engineers mostly from Calix, I am developing the process concepts, process simulations, reactor models and techno-economics. This builds on similar work in LEILAC-1, with the addition of the IHCaL process, of course. I keep in touch with the rest of the consortium because there is a lot of overlap, and enjoy contributing to the discussions in the other work packages. Hopefully I provide some useful insights.

In your opinion, which impact will the ANICA project have on the society?

Cement and lime are literally the foundation of our built environment, and that includes our wind turbines, solar fields and sustainable housing – modern society can't function without it. Cement production currently accounts for 8% of global CO₂ emissions, so decarbonising cement and lime with technologies like ANICA would shave several percent off our annual CO₂ emissions. That said, ANICA will never be as visible to the general public as, say, electric cars or domestic heat pumps. However, ANICA can help to accelerate the cement and lime sectors' transition to net-zero by reducing the cost of capture. In support of Sustainable Development Goal (SDG) 9, cement production is expected to grow from about 4.1 billion tonnes in 2018 at between 3.3% to 8.2% CAGR over the next decade. In order to act on SDG 13 "Climate Action" – a cost-effective, timely option for producing low carbon cement and lime is critical.

Thank you for this insightful interview, Tom. We are looking forward to the results of your work.

My pleasure. See you next time.

ANICA VIRTUAL WORKSHOP

The ANICA consortium cordially invites you to participate in the
“Workshop on the Development of Efficient CO₂ Capture Technologies for Cement and Lime Industries”
held on the
6th of October 2021,
from 9:00 to 12:45 CEST (UTC +2).

Timetable

Welcome & Introduction

9.00—9.15

Session 1: Development of the IHCaL Process for Cement and Lime Plants

9.15– 10.45

Session 2: Ongoing Projects on CO₂ Capture from Cement Production

11.00—12.30

Concluding Remarks

12.15—12.30

This workshop will be organized as an online event featuring speakers from research and industry. You will have the chance to learn about current endeavours of decarbonizing lime and cement plants.

Follow this link to register:

<https://act-anica.eu/anica-virtual-workshop>

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