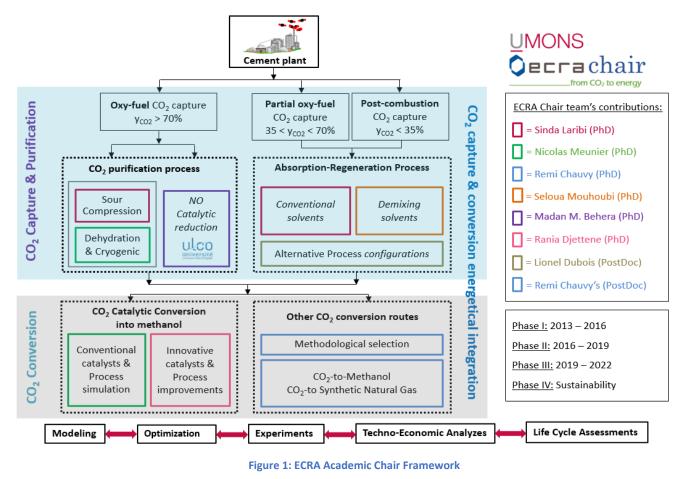
ECRA Academic Chair at the University of Mons: Nine years of achievements to support the implementation of Carbon Capture & Utilization in the cement industry

UMONS Cecrachair

Global overview of the ECRA Academic Chair studies and activities since its creation in 2013

Nine years ago, in April 2013, the University of Mons (UMONS) and the European Cement Research Academv signed (ECRA), an agreement to create an Academic Chair within the UMONS Research Institute for Energy. The main objective of this Chair, coordinated scientifically by Prof. D. Thomas and Prof. G. De Weireld, was to build a Centre of scientific expertise in the specific field of "carbon capture in cement production and its reuse" and promote research and innovation through the works of PhD students, post-doctoral

researchers, but also undergraduate students, supervised by professors with different field of expertise and with a Senior Researcher, namely Dr L. Dubois, as Research Coordinator. The present paper summarizes the main achievements of the ECRA Chair since 2013, whose framework is illustrated on Figure 1. Indeed, Carbon Capture & Utilization (CCU) process chain applied in the cement industry consists in capturing the CO₂ emitted at the outlet of the cement kiln, to purify it (if needed) and then to valorize it by converting it into different products that could be fuels, chemicals, minerals or other products. In this context, different studies were achieved in the framework of the ECRA Chair including process modeling and optimization works, experiments, techno-economic analyzes and life cycle assessments. All these works have generated data allowing to evaluate both the economic and the environmental performances of the investigated carbon capture and utilization processes, specifically for the application to cement plant flue gases.



« (...) The main objective of the ECRA Academic Chair was to build a Centre of scientific expertise in carbon capture and reuse for the cement industry (...) »

How capturing the CO₂?

Globally three pathways exist for capturing industrial CO₂ emissions, pre-combustion, namely: postcombustion and (partial or total) combustion. The first oxy-fuel possibility (pre-combustion) is not adequate solution an to be implemented in the cement industry and was therefore not investigated in the ECRA Chair. Indeed, even if the carbon source of the fuel is eliminated before the combustion itself, that will not have an impact on the two-third of the CO₂ emissions which are coming from the limestone decarbonation (CaCO₃ \rightarrow $CaO + CO_2$). The second process (post-combustion) is the most developed one as it does not imply any modification of the cement production process itself. The flue gas containing around 20 mol.% of CO₂ is sent to an "end-of-pipe" process which separates the CO₂ from the other gaseous components using absorption into a solvent (e.g. amine-based), adsorption onto porous materials, membrane process, cryogenic process or other technologies. The third possibility consists performing in the combustion using pure oxygen instead of air (oxyfuel mode). The CO₂ concentration of the flue gas is higher than with conventional mode (>70 mol.%) which then allows to use a CO₂ Purification Unit (CPU) in order to get a pure CO_2 flow (>99 mol.%). An hybrid process called "partial oxyfuel" is also possible. In such case, oxygen is added to air in order to increase the CO₂ content of the flue gas (>30 mol.%) and to facilitate the CO₂ separation from the other gaseous components using a similar process as for the post-combustion pathway.

What can be done with the captured CO₂?

Once the CO₂ captured, different possibilities exist for its valorization. Historically, the first way envisaged for using large amounts of captured CO₂ is its geological storage (Carbon Capture & Storage - CCS) in coal seams, saline aguifers, depleted oil and gas reservoirs, or for performing Enhanced Oil Recovery (EOR) or Enhanced Coal Bed Methane (ECBM). Since several years now, with a circular economy vision, the possibility of really reusing the CO₂ for producing usable products has emerged. Indeed, a lot of different products can be obtained using CO₂ as building blocks. Some of these products use commercialized processes (e.g. urea production) or at demonstration scale (TRL -Technology Readiness Level – of 7 to 9, e.g. methane, methanol, etc.), and others still need some developments (TRL 4 to 6, e.g. formic acid, ethylene glycol) or are still at the R&D level (TRL 1 to 3, e.g. dimethylether, acetic acid, isocyanates, etc.).

What has been performed within the ECRA Academic Chair regarding CO₂ capture?

Regarding the oxy-fuel CO₂ capture process, the focus was put on the CO₂ purification process. Indeed, the concentrated flow of CO₂ at the outlet of an oxy-fuel cement kiln needs to be purified and especially, residual NOx and SO₂ species must be removed in a CO₂ Purification Unit, see Figure 2. For such purpose, in the framework of the PhD Thesis of S. Laribi, a process called "Sour Compression Unit" (SCU) using pressurized water has been investigated and optimized through modeling works using Aspen Plus[™] software. In addition to thermodynamic properties, а complete set of chemical reactions (Laribi et al., 2019a) had to be implemented to reach the obiectives. One of the main achievements of this research was the adaptation of the SCU from a 2column process to a singe-column one (see Figure 3) and the optimization of the process (in terms

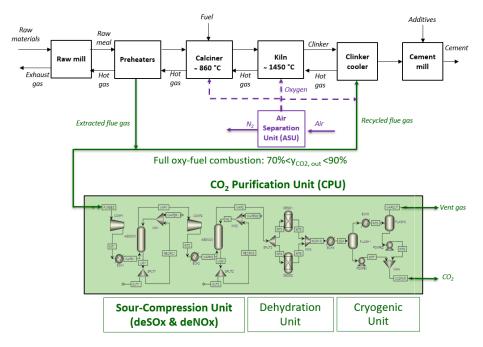


Figure 2: Illustration of the total oxy-fuel process chain in a cement plant

« (...) a lot of different products can be obtained using CO₂ as building blocks. Some of these products use commercialized processes (e.g. urea production) or at demonstration scale (e.g. methane, methanol, etc.), and others still need some developments (TRL 4 to 6, e.g. formic acid, ethylene glycol) or are still at the R&D level (dimethylether, acetic acid, etc.). (...) »

of pressure level, water flow rate, recycle flow rate, etc.) which allowed to decrease both the CAPEX (CAPital Expenditures) and the OPEX (OPerational Expenditures) by 10% and 6% respectively while keeping excellent NOx and SO₂ removal performances (abatement rates higher than 90%).

The works on the CPU (illustrated on **Figure 2**) were also completed in collaboration with N. Meunier (Meunier et al., 2014) for the implementation of a dehydration unit using a Temperature Swing Adsorption (TSA) unit and a final cryogenic unit to reach a final CO₂ purity higher than 98 mol.%.

In October 2019, as joint-supervised PhD Thesis with ULCO (Université du Littoral Côte d'Opale, Dunkerque, France), M. Behera began his PhD Thesis on the optimization of a catalytic process for the CO₂ purification from oxy-fuel combustion. More precisely, an deNOx innovative process is investigated where NO species are reduced by oxidizing carbon monoxide (CO). CO is therefore used as a reducing agent of NOx (essentially NO) by a catalytic way. Nitrogen and water are produced while CO is converted to CO₂, enriching advantageously the flue gas in CO₂. The research works include the development and the characterization of new catalysts, such as the optimization of the catalytic material by catalytic tests in a micro-reactor (at ULCO). The shaping of the material is also realized in order to perform micropilot tests (at UMONS) such as the development of a modelling of the catalytic process at industrial scale applied to the cement industry for performing techno-economic and environmental analyses.

Figure 3: Illustration of the deSOx-deNOx Sour Compression Unit (SCU) improvement

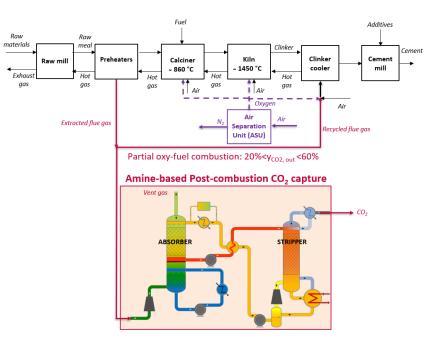


Figure 4: Illustration of the partial oxy-fuel process chain in a cement plant



Figure 5: Absorption-regeneration micro-pilot unit at UMONS

The implementation of a **partial oxyfuel combustion process** in a cement plant was also investigated in the ECRA Chair by S. Laribi (see **Figure 4**). In such configuration, the CO₂ content of the flue gas (between 20% and 60%) is higher than in conventional combustion mode which allows to apply a **conventional** **CO₂ capture technology** (especially amine-based process) with reduced operational costs (Laribi et al., 2019b). Both experiments (using an absorption-regeneration micro-pilot unit available at UMONS, see **Figure 5**) and Aspen Hysys[™] simulations were performed on this topic.

« (...) switching from a 2-column to a 1-column SCU CO_2 purification process allows to decrease both the CAPEX and the OPEX by 10% and 6% respectively while keeping excellent NOx and SO₂ removal performances. (...) »

It is important to remind the principle of the CO₂ capture absorption-regeneration process using amine-based solvent (see Figure 6). The flue gas to be treated is sent into a first column where the CO₂ is absorbed into a chemical solvent (e.g. monoethanolamine (MEA) 30 wt.%), which is then sent into a second column where it is regenerated thanks to heat. This regeneration step, releasing a quite pure CO₂ flow (98 mol.%), consumes a lot of energy (e.g. 3.4 GJ/t_{CO2} for MEA 30 wt.%) provided as steam. By increasing the CO₂ content of the flue gas from 20 mol.% to 40 mol.% using partial oxy-fuel combustion, it has been shown with different solvents (MEA, but also piperazine (PZ) and diethanolamine (DEA) and PZ blend, see Figure 7) that the solvent regeneration energy (and therefore the OPEX) can be reduced by around 25%. The increase of the CO₂ content of the flue gas to 60 mol.% did not lead to significant supplementary savings.

Focusing on the post-combustion CO₂ absorptioncapture regeneration process, two other options were also investigated in the framework of the ECRA Chair for reducing its energy consumption and therefore its cost. The first solution was investigated by S. Mouhoubi in her PhD Thesis, namely the use of demixing solvents (see Figure 8). Indeed, thanks to the use of adequate proportions of specific amines (e.g. diethylethanolamine (DEEA) and methylaminopropylamine (MAPA) blends were investigated by the ECRA Chair), at a certain level of temperature and CO₂ loading of the solvent, two phases are formed and can be separated in a decanter unit: one lean-phase in CO₂ which can be directly recycled

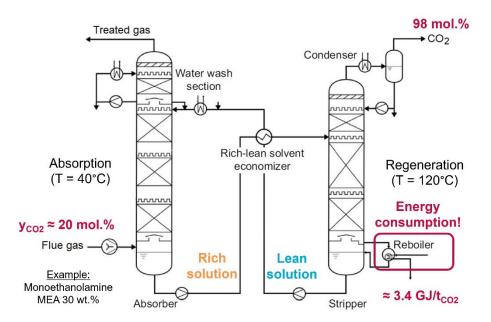
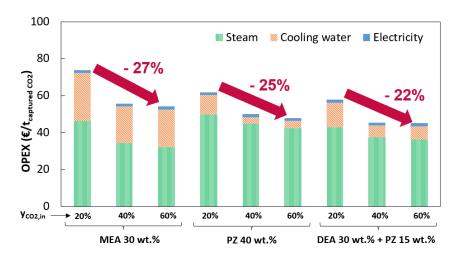


Figure 6: Conventional CO₂ capture absorption-regeneration using amine-based solvent (illustration for monoethanolamine (MEA) 30 wt.%)





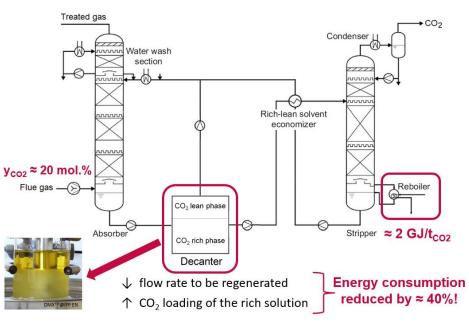
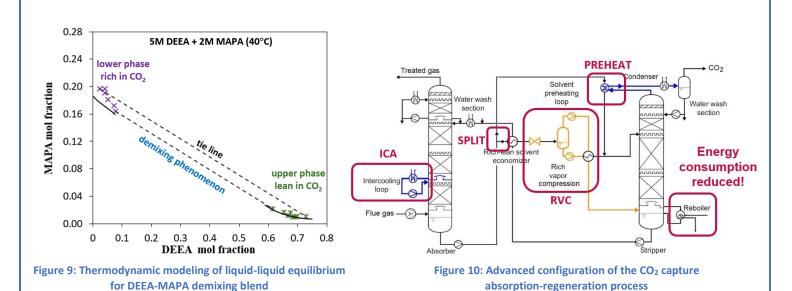


Figure 8: CO₂ capture absorption-regeneration process using a demixing solvent

« (...) increasing the CO₂ content of the flue gas to 40% thanks to partial oxy-fuel combustion decreases the CO₂ capture OPEX by 26%. (...) »



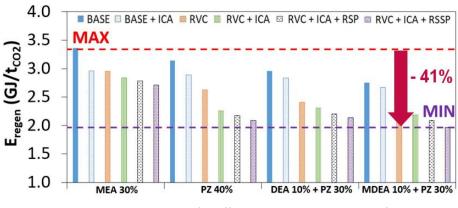
back to the absorber and one richphase in CO₂ which is sent to the stripper for its regeneration. By performing such operation, as the CO₂ loading of the rich solution is higher than with the conventional process and as the solvent flow rate to be regenerated is reduced, the process energy consumption is decreased. The purpose of the PhD Thesis of S. Mouhoubi was to develop a modeling in order to simulate such demixing process. This modeling was implemented in Aspen Plus[™] software and its development needed to take into account the different aspects: thermodynamics (see illustrations on Figure 9), kinetics, physical properties, etc. (Mouhoubi et al., 2020). Some of the modeling results in non-demixing conditions were also validated using the UMONS micro-pilot unit (Figure 5). Based on the simulations developed considering a BAT (Best Available Technology) cement plant, it was estimated that the solvent regeneration energy could be reduced by 40% (2 GJ/ t_{CO2}) with such demixing process in a comparison with a conventional MEA 30 wt.% technology. A techno-economic analysis also highlighted that the CO₂ capture costs could be reduced by 35%.

The addition of a decanter leads only to 1% increase of the CAPEX.

The second solution to reduce the energy consumption of such CO₂ capture process, that was investigated in the ECRA Chair by L. Dubois as Senior Researcher, is the implementation of advanced process configurations with amines blends (Dubois and Thomas, 2018; Dubois and Thomas, 2021). Actually, as illustrated on Figure 10, by implementing supplementary units (e.g. intercooling absorber (ICA), rich vapor compression (RVC), split and preheat, etc.) to the absorption-regeneration process it is possible to significantly reduce its energy consumption, especially if a more efficient solvent than MEA 30 wt.% (e.g. methyldiethanolamine (MDEA) and piperazine (PZ) blend) is implemented.

of different The investigation process configurations considering different solvents was performed through Aspen Hysys[™] simulations. Considering the process represented **Figure** 10 on and the implementation of MDEA+PZ blend, it was shown (see Figure 11) that 41% energy saving is possible in comparison with the conventional MEA process. In terms of economics, despite an increase of 10 to 15% of the CAPEX, a decrease of 31% of the global CO₂ capture costs was estimated.

Globally, the research works performed within the ECRA Chair have clearly highlighted different technical solutions that could help to reduce the CO₂ capture energy consumption and its cost for the application in the cement industry.





« (...) Both demixing solvents and advanced process configurations allows to decrease the solvent regeneration energy by 40% (...) »

What about CO₂ conversion?

As illustrated on Figure 1, the ECRA Academic Chair research activities also focused on different CO2 utilization options. In the framework of the PhD thesis of R. multistep Chauvy, an **original** method was developed (Chauvy et al., 2019) to classify different CO₂ conversion alternatives and to identify the best emerging options to be implemented in short- to midterm via a multi-criteria assessment including technical, economic, energetic, environmental and market considerations. The impact different factors of on the assessment results also was investigated (Chauvy et al., 2020a). Different alternatives were reviewed (Chauvy and De Weireld, 2020), involving the production of compounds that have a low unit price, but significant market volume, such as methanol, methane, calcium or sodium carbonates, microalgae, urea, syngas and ethanol, and compounds that have a high unit price but low market volume, such dimethyl carbonates. as polycarbonates, formic acid and salicylic acid (see Figure 12). Globally, methanol and methane (as Synthetic Natural Gas - SNG) were identified as two of the most promising CO₂-based products among examined cases through the analysis sensitivity and were therefore more deeply investigated in the ECRA Chair.

The **CO₂ catalytic conversion to methanol** was specifically studied within the ECRA Chair by N. Meunier. During his PhD Thesis, N. Meunier worked both on the development of a modeling of such process, but also on the establishment of a micro-pilot unit (see Figure 13).

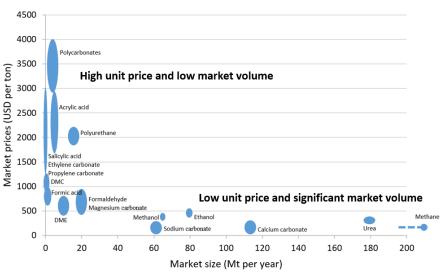


Figure 12: Market sizes and market prices for the main CO₂-based compounds (non-exhaustive) from (Chauvy et al., 2019)



Figure 13: CO₂ catalytic conversion to methanol micro-pilot unit at UMONS

experimental investigation The performed in the work of N. Meunier assessed the interest in developing innovative catalysts for the direct of conversion pure CO_2 into methanol by comparing two catalysts: a commercial CuO/ZnO/ Al_2O_3 catalyst and an innovative homemade CuO/ZnO/ZrO₂ one, synthesized with the collaboration School of the European of Chemistry, Polymers and Materials (ECPM) from Strasbourg. This experimental investigation allowed the determination of kinetic constants related to both these catalysts. From the experiments, it was noticed that the temperature

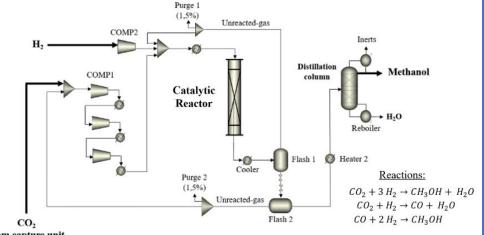
had a dramatic influence on the productivity and selectivity towards methanol. It has been pointed out that the commercial catalyst has a higher selectivity towards methanol at temperature higher than 250°C, where the zirconia supported catalyst has a higher selectivity towards methanol at temperature below 230°C.

The great interest in zirconiasupported catalyst was thus proven for the production of methanol as this kind of catalysts demonstrated higher methanol productivities than the commercial one. Together with other ECRA Chair researchers (Meunier et al., 2019), the determined kinetic parameters were then used for the development in Aspen Plus[™] of a simulation of the CO₂ conversion to methanol process thermally integrated with a CO₂ capture unit (using amine-based technology), including technoeconomic and environmental assessments. Indeed, the CO₂ conversion to methanol being an exothermal process, the possibility to recover the released heat to reduce the global energy consumption of both the CO₂ capture and the water-methanol distillation steps was investigated. From these developments, one of the main conclusions is the fact that significant energy savings are possible thanks to the implementation of thermally optimized CO₂ capture (quite similar advanced process as the one investigated by L. Dubois) and CO₂to-methanol processes. Moreover, the renewable hydrogen production cost remains one of the key elements for such CCU process chain. The works on the CO₂ conversion into methanol are still in progress in the ECRA Chair through the PhD Thesis of R. Djettene, started in October 2019. The objective is to continue the optimization of the process thanks to the exploitation of the CO₂-tomethanol micro-pilot catalytic reactor (see Figure 13), including experiments with different catalysts, but also modeling development and process simulations (see Figure 14) perform economic in order to and environmental analyses.

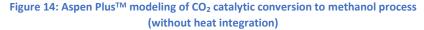
As mentioned, the environmental evaluation (Life Cycle Assessment (LCA)) of the CCU processes (including CO₂-to-methanol one)

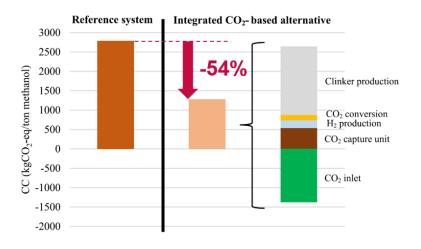
also performed the was in framework of the ECRA Chair thanks to the works of R. Chauvy during his PhD Thesis and his Post-Doc. Such analysis allows to evaluate the environmental impact in terms of Climate Change (CC) (kg_{CO2equ}/t_{CO2}based-product), but also in terms of fossil water depletion, depletion, metal/mineral depletion, etc. In the case of CO₂ conversion to methanol, as represented on Figure 15 for the CC indicator, in comparison with a production reference system (methanol production from fossilbased syngas), a reduction of 54% of the equivalent CO₂ emissions was estimated, which confirms the interest of such integrated CCU process in comparison with a fossilbased one.

In addition to the analysis of the CO₂to-methanol process, and in close collaboration with L. Dubois, R. Chauvy also investigated during his Post-Doc the entire CCUS chain, from industrial CO₂ capture from BAT cement plant flue gas, to its conversion to SNG together with renewable hydrogen, through simulation and process-to-process integration. It allowed to get relevant technical and economic parameters, and to perform LCA, identifying hotspots contribution and quantifying most significant environmental impacts. As for methanol, naturally the economic breakthrough was identified to be strongly related to high capital investments and electricity costs



from capture unit

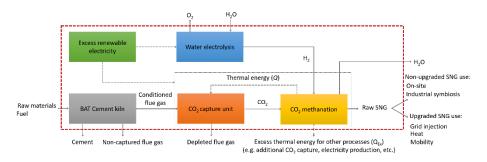






« (...) In comparison with a conventional methanol production system from fossil syngas, a reduction of 54% of the equivalent CO₂ emissions was estimated (...) »

required for the production of hydrogen, which is nevertheless expected to decrease in the upcoming years. As illustrated on Figure 16, the thermal integration between an advanced CO₂ capture unit and a CO₂ methanation unit was performed at a representative scale for current electrolyzers used for hydrogen production, namely 10 MW. Moreover, as the CO_2 methanation process is highly exothermic, in addition to be selfsufficient in terms of thermal energy, different options were investigated through Aspen Plus[™] simulations for the valorization of the excess heat available, namely capturing more CO₂ (Case 1, see Figure 17) leading to some extra-CO₂ compressed in view of transport for other CCUS applications (Chauvy et al., 2021a), generating electricity (Case 2) (Chauvy et al., 2020b) or providing heat as steam available for other industrial processes (Case 3) in an industrial symbiosis vision (Chauvy et al., 2021b), all the details regarding these investigated cases being available in the mentioned papers. For example in Case 1, it was pointed out that the use of the entire excess heat for capturing the CO₂ allows to recover a supplementary flow corresponding to 70% of the needed for the quantity methanation unit. In Case 2, it was shown that the electricity generated using the excess heat covers the entire electrical demand of the methanation unit, covering also 5% of the electricity demand of the CO₂ capture unit. On the other hand, in Case 3, it was highlighted that only 60% of the excess heat is used for the CO₂ capture, meaning that 40% is still available as steam for other purposes. In terms of economics, one of the main observations is the fact that the supplementary CAPEX





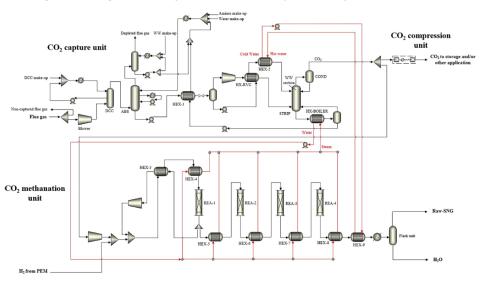


Figure 17: Aspen Plus[™] flow sheet of the integrated CO₂ capture and methanation units with extra-CO₂ captured for CCUS purposes, from (Chauvy et al., 2021a)

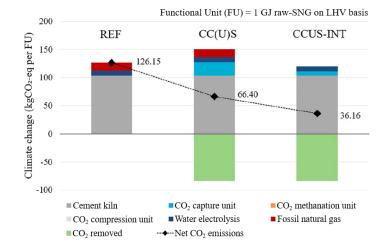


Figure 18: Climate Change impact (CC) of the CO₂ conversion process to SNG with thermal integration (CCUS-INT) in comparison with a reference cement plant (REF) and a CC(U)S process with fossil-based natural gas production, from (Chauvy et al., 2021a)

related to the heat integration (e.g. heat exchangers, pumps, etc.) represents only around 2% of the total CCU investment costs, while for example in Case 3 (total OPEX estimated to $117 \notin t_{CO2}$ without H₂ production) the OPEX would be 30% higher if external steam must be provided to the capture unit (without heat integration).

Globally, the three Cases have shown an environmental interest.

Different environmental impacts were investigated but focusing on CC) (see Figure 18 for Case 1), in comparison with their respective reference scenario, the three approaches led to a relative decrease of the net CO_2 emissions higher than 70%.

ECRA Academic Chair Activities

As illustrated on Figure 19, in parallel of the scientific research works leading to PhD theses and international publications (see P11), the ECRA Chair members are taking part to different activities. Two times a year, ECRA and UMONS members meet for the ECRA Chair Scientific Committees.

The ECRA Chair Professors, Post-Docs and PhD Students also participate regularly to international scientific events in order to present the advancements of the ECRA Chair works. Moreover, in 2014 and 2016, the ECRA Chair organized his own Scientific Events with around 100 international attendees each time and including the visit of a cement plant. Undergraduate students are also associated to the ECRA Chair works for Master Theses, projects, internships, etc., and an ECRA Award is delivered annually to reward the best student work(s) achieved.

Final remarks & ECRA Academic Chair perspectives

The ECRA Academic Chair is contributing on different aspects (CO₂ capture, purification and conversion, both in (total/partial) oxy-fuel combustion and postcombustion) to help for the implementation of CCU in the cement industry. Different technologies and cases studies were studied. Nevertheless, it is important to highlight that each case investigated has specificities that need to be considered, such as: the production of electricity with excess heat needs either the establishment of, or to have access to, a highpressure water network, as well as the implementation of a turbine, etc., which is not necessarily easy;

the eventual extra-CO₂ captured needs to fulfill the CO₂ transport network specificities in terms of purity (and such network must be available), and a potential other utilization or geological storage capacity of this CO₂ must be ensured; and finally the valorization of excess thermal energy as steam is possible only if another industrial plant is situated not far from the cement plant itself in order to really perform an industrial symbiosis. For all cases investigated the considering methanol or methane as CO₂-based product, it has to be noted that the production of renewable H₂ remains an important factor for CCU, especially due to the high costs (both CAPEX and OPEX) of the electrolysis process, and also for the need of renewable electricity.

It is worth mentioning that the methodologies developed in the framework of the ECRA Chair could be potentially applied considering other CO₂ capture and/or CO₂ conversion processes. The works are still in progress with currently two PhD Theses, namely on CO₂ purification and on CO₂ conversion to methanol.

More globally, the ECRA Chair members at UMONS are working for the sustainability of the Centre of scientific expertise built since 2013, through the broadening of the collaborations, with cement industry actors but also with other industries and with the introduction of new research subjects (e.g. Direct Air Capture). Indeed, the ECRA Academic Chair at UMONS became a real Centre of scientific expertise for CCU application in the cement industrv but also for other applications as UMONS is now regularly contacted for performing studies (e.g. technological guidance, process simulations, TEA, LCA, etc.) in order to help for implementing CCU options in different industrial sectors. Furthermore, in 2022, ECRA and UMONS confirmed their common willingness to continue their cooperation until at least 2025 through the signing of а Memorandum of Understanding.

The ECRA Chair really put on track a lot of activities and a real way of working that will be pursued because the fight to tackle climate change is not finished, still many battles have to be won!



Figure 19: Illustrations of some ECRA Academic Chair activities

ECRA Academic Chair Research Team at UMONS

Academic & Scientific Coordinator:



Prof. Diane Thomas Chemical & Biochemical Process Engineering Unit





Prof. Guy De Weireld Thermodynamics Unit

Research Coordinator & Senior Researcher:

PE

Prof. Anne-Lise Hantson Chemical & Biochemical Process Engineering Unit



Academic & Scientific Advisors:

Prof. Paul Lybaert

Thermal Engineering

& Combustion Unit

Prof. Ward De Paepe Thermal Engineering & Combustion Unit



Dr Lionel Dubois Chemical & Biochemical Process Engineering Unit

PhD Students & Post-Doc :



Dr Nicolas Meunier (2018)



Dr Sinda Laribi (2018)



Ir Madan Behera (in progress)**



Dr Remi Chauvy (2019)*



Ir Rania Djettene (in progress)

*Post-Doctoral researcher from 2019 to 2020.

**Joint-supervised PhD Thesis with ULCO (Université du Littoral-Côte-d'Opale, Dunkerque, France).

Acknowledgments

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For more information on the ECRA Academic Chair: <u>https://hosting.umons.ac.be/html/ecrachair</u>



Dr Seloua Mouhoubi (2020)

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https://doi.org/10.1016/j.egypro.2014.11.109

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In addition to these publications, numerous scientific communications were also presented in international conferences.



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