Publishable Summary Report

EU 7th Framework Programme Collaborative Project

SUCCESS

-

Industrial steam generation with 100% carbon capture and insignificant efficiency penalty - scale-up of oxygen carrier for chemical-looping combustion using environmentally sustainable materials

http://www.clc-success.eu

Theme:
FP7 – ENERGY.2013.5.1.1

Grant agreement no:
608571

June 2017

Executive Summary

Chemical looping combustion (CLC) is an alternative combustion technology with inherent CO_2 capture capabilities and nearly no energy penalty. This is achieved by avoiding the energy intense gas-separation step typical for CO_2 capture technologies. CLC is thus seen as a potential break-through CO_2 capture technology. CLC of gaseous fuels is well understood being demonstrated at 140 kW and in long term experiments (1 000 hours at 10 kW) using a nickel based oxygen carriers. Further, several promising nickel free oxygen carriers have been identified reaching full fuel conversion and being economic and environmentally attractive. Interest in the technology is not only limited to European Countries shown by extensive research activities in the United States and China which are mostly focused on coal as fuel.

Despite its large potential, demonstration of the technology at large scale did not start due to unsolved technological questions. The main goal of the SUCCESS project was providing the last missing R&D input information for demonstration of Chemical Looping Combustion (CLC) of gaseous fuels at next scale in the range of 10 MW_{th}. This included research on scale-up of the most important aspects of Chemical Looping Combustion: Oxygen carrier material production and reactor system design. A scale-up to 10 MW is seen as appropriate given the complexity of the process.

Two complementary oxygen carrier materials for different applications developed in previous EU funded projects have been selected for scale-up to production at tonne scale. A copper based oxygen carrier was produced via impregnation on an alumina support. A manganese based oxygen carrier was produced via spray-drying. After successful scale-up, 3.5 t of oxygen carrier material have been produced using industrially relevant production facilities and raw materials. The oxygen carrier materials were extensively tested in several continuously operating pilot units up to 150 kW_{th}. This allowed investigation under a broad range of operating conditions. Investigations showed satisfactory fuel conversion performance levels and influence of important process parameters like temperature, solid inventories and solid circulation rates. The oxygen carriers worked well in all pilot units achieving CO_2-yields above 90%. In addition to pilot testing up to 150 kW, CLC of gaseous fuels has also been demonstrated in the size of 1 MW_{th} using the Ca-Mn based OC.

System design optimization was performed on several levels. After identification of reaction mechanisms behind fuel conversion, mathematical modelling tools in the form of parametric and 3D-models were adapted and used. Further, a design for a next scale demonstration unit was proposed and evaluated using a scaled physical flow model of the fluidized bed system.

End-user evaluation of the technology for industrial steam generation evaluated techno-economic performance as well as issues related to health and safety, potential recycling routes and the whole life cycle impact. Recommendations for handling oxygen carrier material were drawn based on a health, safety and environmental impact analysis. The analysis included human and environmental effects. Different options for reuse and recycling of spent oxygen carrier materials have been investigated. The techno economic evaluation showed that CLC for gaseous fuels is competitive compared to best available technologies for industrial steam generation.

All project objectives and technical goals have been achieved. The technology can be seen as ready for next scale demonstration having large scale oxygen carrier material production technologies at hand. Further, detailed design recommendation for such a demonstration unit were made. However, the project consortium still sees potential for further optimization of oxygen carrier material production. This further optimization should lead to reduction of production costs and increase of particle life time which will further improve the economic performance of the technology.

Table of contents

1 Project context and objectives

1.1 Introduction to Chemical Looping Combustion

Chemical looping combustion (CLC) is an innovative combustion technology with inherent CO_2 capture and nearly no energy penalty. This is achieved by avoiding the energy intense gas-separation step typical for CO_2 capture technologies. Chemical looping combustion is thus seen as a potential breakthrough CO_2 capture technology.

To avoid gas-separation, the combustion process is separated into two different reaction zones, air reactor (AR) and fuel reactor (FR) in a way that fuel and combustion air are never mixed. A solid oxygen carrier (OC), a metal oxide, is circulating between AR and FR and transporting oxygen from combustion air to fuel (see ***Figure 1***). The oxygen carrier is oxidized in the AR by combustion air end reduced in the fuel reactor by the fuel. The process yields two different exhaust gas streams. AR exhaust gas contains N_2 and excess O_2, exhaust gas from the FR contains the combustion products CO_2 and H_2O. After condensation, a highly concentrated CO2 stream can be obtained. The total heat release in the two reactors is exactly the same as in normal combustion processes.

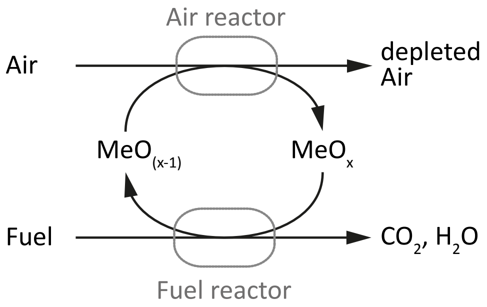


Figure 1: Concept of Chemical Looping Combustion.

AR and FR are designed as interconnected circulating fluidized bed reactors and the oxygen carrier is the bed material circulating between these reactors. The process temperature for CLC is comparable to conventional combustion processes, i.e. 800-1 000 °C depending on fuel and oxygen carrier material. Thus, a CLC reactor system can be used in the same way as a conventional circulating fluidized bed reactor (CFB) in a steam cycle process (heat recovery steam generator plus steam turbine) to produce power, heat and/or process steam.

Early deployment is seen in natural gas steam generation, where gas-to-steam efficiency penalty with CLC is below 1%-point compared to 15%-points with amine scrubbing and 8%-points with oxyfuel combustion, all for 95% capture rate. Reduction of the CO_2 avoidance cost of 60% compared to amine scrubbing post combustion capture results from higher efficiency.

Research in CLC has to focus on two different aspects of the technology: the oxygen carrier material and the reactor system. However, it is of great importance that the reactor system meets the demands of the oxygen carrier and vice versa. For successful up-scaling of the technology both need to be accomplished in parallel. CLC of gaseous fuels is well understood being demonstrated at pilot scale 140 kW and in long term experiments (1 000 hours at 10 kW) using a nickel based oxygen carrier. Further, several promising nickel free oxygen carriers have been identified reaching full fuel conversion and being economic and environmentally attractive.

1.2 Project objectives

The main objective of SUCCESS is to perform the necessary research to close the last gap between the state-of-the-art and demonstration of the CLC technology for gaseous fuels at the 10 MW scale. This will include scale-up of OC production to the 100 tonne scale, as well as demonstration of the technology at 1 MW fuel power input. Industrially available raw materials are used to produce environmentally sound oxygen carriers based on two highly successful materials developed in previous EU funded projects. This can be translated into the following project objectives:

1. Production of two large batches (≥500 kg) of scale-up ready material using industrially available raw materials and large scale production techniques.
2. Proof of performance of these materials in pilot plants up to 150 kW fuel power input
3. Demonstration of the CLC technology for gaseous fuels at 1 MW.
4. Presentation of an optimized system design for next scale (10 MW).
5. Quantification of the techno-economic potential of the CLC technology for gaseous fuels.

To reach these goals, the following work is performed within the project:

1. Applying oxygen carrier production methods at industrially required scale and assuring the adequate performance.
2. Development of a standard for determination of mechanical stability of OC particles.
3. Operation in four smaller pilots up to 150 kW of significantly different design.
4. Operation with gaseous fuels in a 1 MW pilot plant, representing a scale up of the state of art by one order of magnitude.
5. Detailed studies of reaction mechanisms and fluid-dynamics.
6. Use of results in optimization of a previous design for a 10 MW demonstration plant and techno-economic study of full-scale plant.
7. Assessment of health, safety and environmental issues associated with OC handling including life cycle analysis.

Overall techno-economic evaluation of the CLC steam generation technology.

1.3 Project structure

The project is structured in eight technical work packages, where six (WP1-WP6) are related to technology scale-up and testing of OC material up to 1 MW and two (WP7 and WP8) are related to end-user evaluation of the technology. The structure is summarized in Figure 2.

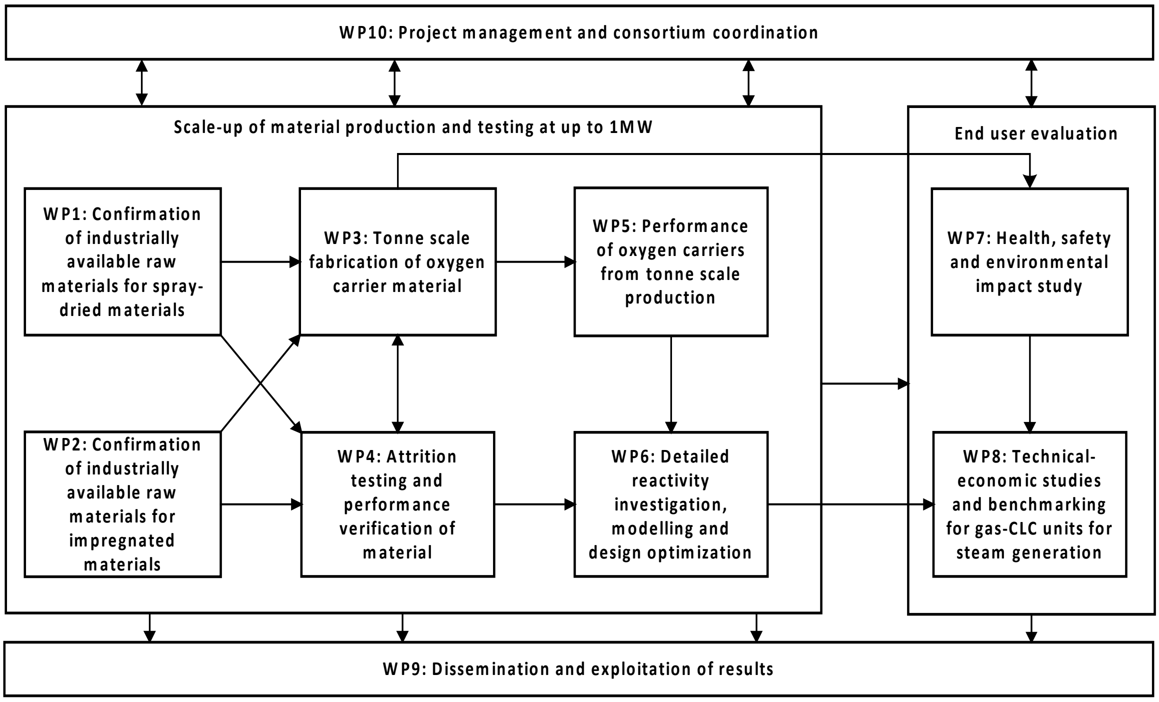


Figure 2: Structure of the SUCCESS project

Combined efforts of key European developers of the CLC technology assure the continued European leadership in this development and bring the technology a major step towards commercialization. The SUCCESS consortium consists of 16 partners from 9 countries including research institutions, technology providers and end-users. The project partners and their activities are summarized in Table 1.

Table 1: SUCCESS project consortium.

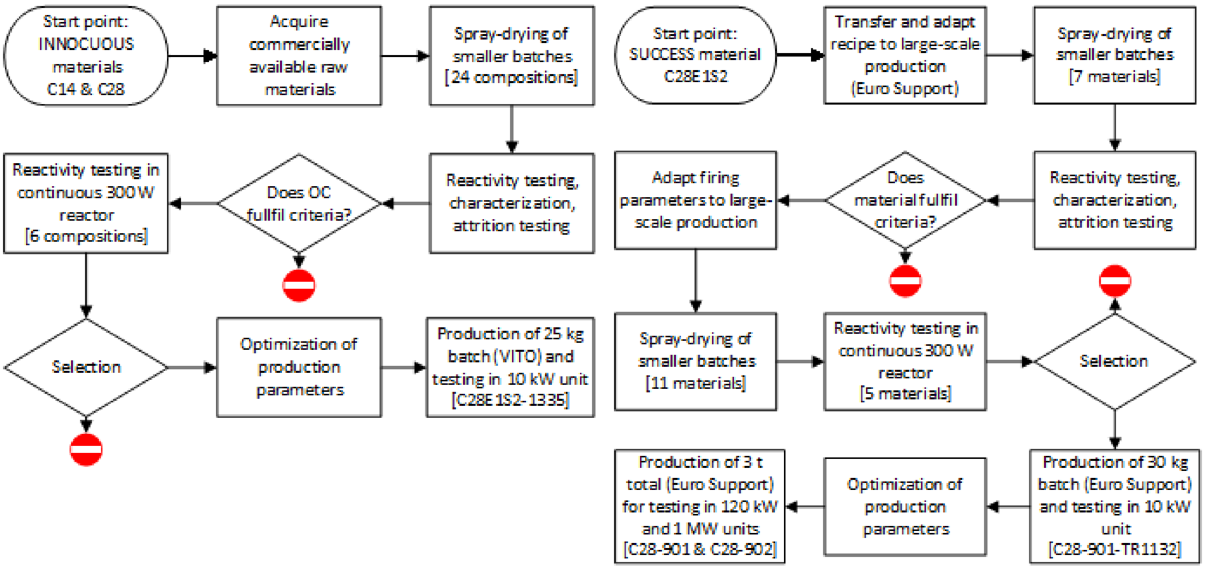
Partner name	Country	Type	Activities
Vienna University Technology	AUT	University	Project coordination, pilot plant OC testing, 10 MW system design
Chalmers University of Technology	SWE	University	OC development, pilot plant OC testing, attrition testing
CSIC	ESP	Research Institute	OC development, OC testing, attrition testing, reactivity investigations, modelling, OC recycling investigation
IFP Energies Nouvelles	FRA	Research Institute	Pilot plant OC testing, attrition testing, reactivity investigations
Institut National Politéchnique de Toulouse	FRA	University	3D-Modelling
SINTEF Materials	NOR	Research Institute	Reactivity investigations, attrition testing
SINTEF Energy	NOR	Research Institute	Pilot plant OC testing
Darmstadt University of Technology	GER	University	Pilot plant OC testing (1 MW)
VITO	BEL	Research Institute	OC development, life cycle analysis, health safety and environmental impact evaluation
Euro Support Advanced Materials	NED	Material producer	Large scale production of OC material
Johnson Matthey	UK	Material producer	Large scale production of OC material

Partner name	Country	Type	Activities
Bertsch Energy	AUT	Boiler manufacturer	Sizing and design of equipment for economic evaluation
Électricité de France (EDF)	FRA	Power	Techno-economic evaluation
Shell Global Solutions	NED	Oil&Gas	Health safety and environmental impact evaluation
TOTAL Raffinage Chimie	FRA	Oil&Gas	Pilot plant OC testing, attrition testing, techno-economic evaluation
University of Natural Resources and Life Sciences	AUT	University	Mass and energy balance calculations for techno-economic evaluation

2 Oxygen carrier production scale up

2.1 Oxygen carrier material produced by spray-drying

In the previous 7th FP INNOCUOUS project, a spray-dried calcium-manganese-based material of general composition $CaMn_{1-x-y}Mg_xTi_yO_{3-\delta}$ was found to have excellent properties as oxygen carrier for natural gas combustion. One key objective of the SUCCESS project was to confirm adequate properties of this oxygen carrier when production of this material uses commercial raw materials, i.e. materials available in quantities sufficient for large-scale production (>100 t) and at a suitable price. Several work packages have been devoted to sourcing raw materials and production of particles by spray-drying to tonnage scale. The main tasks in this endeavor are outlined in Figure 3. The process of obtaining suitable raw materials for production was mainly conducted in WP1, and main tasks described in Figure 3a, while the following up-scaling procedure is outlined in Figure 3b. These figures should be seen as a general guide as how the development work proceeded.



(a) Raw material sourcing

(b) Up-scaling of production

Figure 3: General process flow diagram with main tasks involved in oxygen carrier development.

Early on in the project it was decided to focus on sourcing of different manganese and titanium oxides as raw powders in the spray-drying procedure, and a vast number of materials were obtained from various suppliers, outlined in detail in D1.1 and D1.2. From selected raw powders a total of 24 compositions of calcium manganite were spray-dried in smaller 1 kg batches, and were tested with respect to parameters important for CLC, i.e. reactivity and attrition behavior. All materials were compared to the reference materials, produced in the earlier INNOCUOUS project. An important finding here was that all spray-dried materials evidently became calcium manganites of the perovskite structure, with rather similar oxygen release patterns. The fact that widely different Mn-oxides were used, from rather pure oxides to natural ores, amplifies the flexibility and viability of the system. Several materials had a combination of higher reactivity and lower or similar attrition index compared to the reference materials. Six compositions were tested in continuous operation using a bench-scale reactor with a nominal fuel input of 300 W_{th}, and, again, rather promising results were obtained: almost complete methane conversion could be achieved for all six investigated materials. For scale-up materials using Mn_3O_4 from Elkem (Colormax P) and TiO2 from Sachtleben (TP Hombikat M211) were chosen. The $Ca(OH)_2$ and MgO were kept the same as in the reference particles. The calcination temperature was found to have a significant effect on reactivity, and this was optimized to 1335°C for 4 h, using this oxygen carrier. This oxygen carrier, here denoted C28E1S2-1335, showed high reactivity in the Chalmers 10 kW pilot, as is seen in Figure 8.

In parallel with the last part of the optimization of the choice of raw materials and process conditions in WP1, the process steps for large-scale production were adapted and tested. One of the important goals of the SUCCESS project has been to produce sufficiently large batches (>100 kg) of oxygen carrier for pilot scale testing at 100kW to 10MW Chemical Looping plants. In order to prepare for fabrication of tonne-sized batches, the requirements and characteristics of new and existing infrastructure for performing the necessary process steps in the spray-drying route have been assessed. The process steps in the spray-drying route are suspension preparation, including milling of raw material, spray drying, classification, rework, and calcining or sintering, see Figure 4.

Soon after the start of the project EuroSupport Advanced Materials (ESAM) has defined the specification of a new spray dryer for the Uden location. This dryer produces spheres by means of a pressurised fountain nozzle, enabling the fabrication of spheres of the desired size and with a narrower size distribution. The main tasks in the upscaling procedure is outlined in Figure 3b and are described in more detail below.

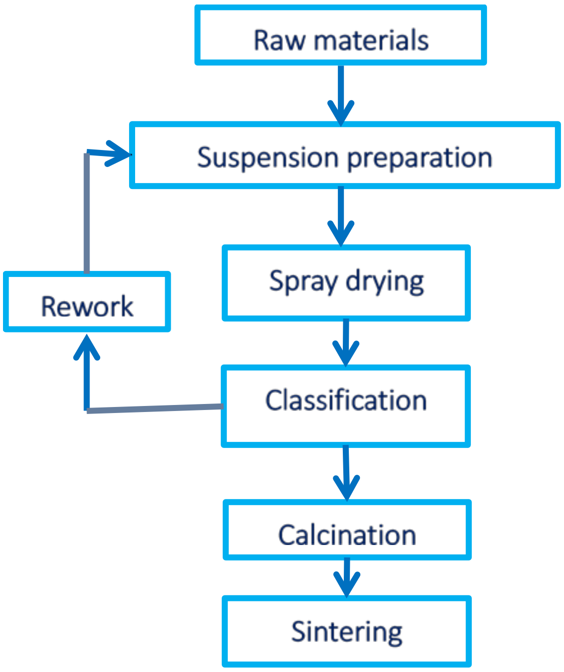


Figure 4: General process flow chart showing the steps in the spray-drying route for oxygen carrier manufacturing.

Raw materials, suspension stability and spray drying of up-scaled material

A fit-for-use spray-drying suspension was developed by Euro Support in close cooperation with VITO. A slurry that is stable for long enough time, preferably several days, is a requirement for a robust industrial process. Taking base in the synthesis procedure and recipe from WP1, i.e. C28E1S2, the upscaling procedure followed a path similar to that shown in Figure 3b. Early on, it became apparent that the slurry system used for the small- and medium-scale process at VITO was not suitable for the industrial process at Euro Support. In fact, it was found that an adaption in the Ca and Mg sources was necessary, as Ca- and Mg-oxides and -hydroxides were not viable during slurry preparation due to gel-formation. Finally, a slurry composition could be identified that showed a stable viscosity for over more than 4 weeks. Also, investigations showed that another binder system had to be adopted in order to be able to produce the desired spheres in a sufficiently wide process window. The produced spheres showed the desired round shape and a sufficiently dense internal structure, as shown in Figure 5 for a sample (center of lower crucible, furnace back-row) taken from sinter-batch 901.

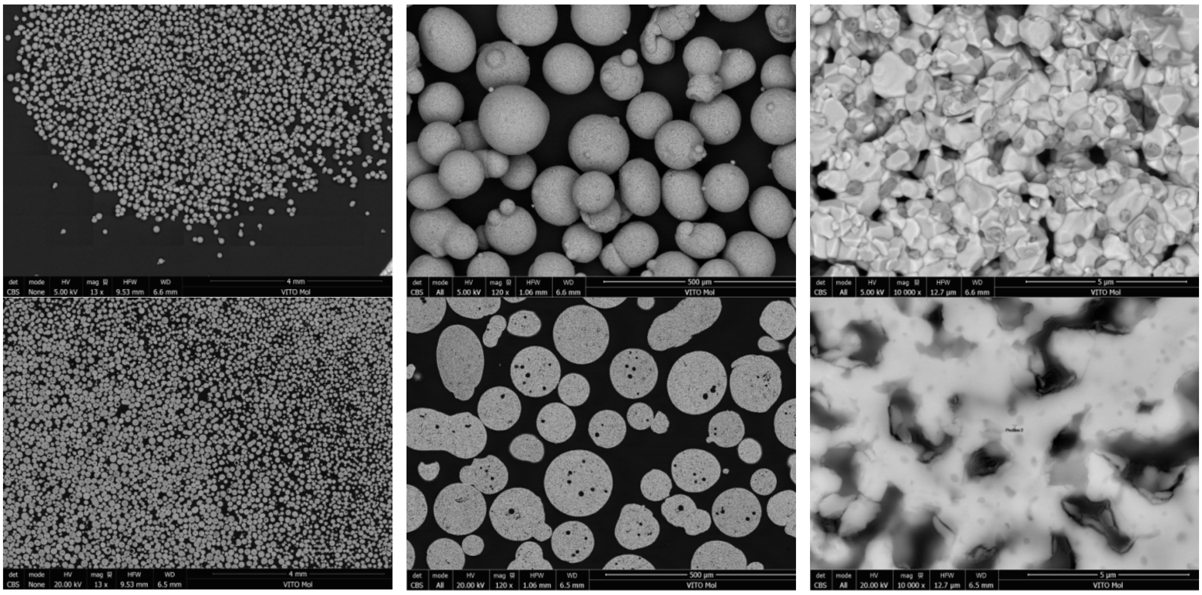


Figure 5: Calcined C28 oxygen carrier spheres from large scale batch at magnification 13x (left), 120x (middle) and 10000x (right); top row: surface view, bottom row: cross-sectioned.

Calcining/sintering

Assessment of the sintering process by VITO showed the risk for inhomogeneous calcination and resulting inhomogeneous crushing strength. Therefore, the calcining process of large quantities in the large furnace of Euro Support (saggar type and loading) was investigated in detail. Iteratively a number of C28 calcium manganite samples with a sufficient CS from preliminary ESAM batches were tested in the Chalmers bench-scale reactor. The resulting range in CO_2-yield was found to be between 81 – 97 % for materials and the C28 material that did not react with the showed a CO_2-yield of 96.3 % at 900 °C possesses both targeted low attrition (0.42 wt.%/h) and high crushing strength (2.4 N). Increased saggar loading showed a lower CO_2- yield of 81.1 – 91.1 % (246 – 331 kg/MW at 900 °C), an equally high crushing strength (2.5 N) and an attrition index of 1.02 wt.%/h, sufficiently adequate for continued upscaling. The material performed well in the runs at Chalmers and also showed good attrition behavior in the 10 kW reactor. The final material selected, C28-901-TR1132, was evaluated in the Chalmers 10 kW pilot for over 100 h with fuel, see Figure 8.

Due to unexpected side reactions and calcination atmosphere influences on the calcination process in the production environment, it was necessary to re-optimize the calcination conditions close to the project end. Since one full-scale production run requires 600 kg, the scale effects caused that the optimum calcining conditions could not be applied. As such, it was decided to proceed with production parameters that yielded a higher reactivity at a potential sacrifice of mechanical stability. Thus, the significant risk of over-calcining was avoided which would have yielded stronger but inactive spheres. The selected oxygen-carrier batch size of 1 t is of technically relevant for future multi-tonne scale production. Eventually nearly 3 t of C28 oxygen-carrier material, i.e., spray-drying batches 901 and 902, were produced by Euro Support and delivered to the project partners. Thus, it was successfully shown that oxygen-carrier production at multi-tonne-scale is available at the point where a commercial order is placed. The material was used in five pilot units of up to 1 MW, as is further explained in Section 3.

Characterisation of up-scaled material

In addition to the reactivity and attrition investigations carried out in several CLC pilots, see Section 3, physical and chemical characterizations were performed. Over 40 samples were taken of the final tonne batch product and a selection has been characterised by determination of relevant physiochemical properties at VITO. It can be concluded that there is some inhomogeneity (crushing strength, density and tap density, % of perovskite phase) throughout the large batch, mainly caused by variations in the calcination process, i.e., location in the saggar, calcination temperature and saggar material. Optimization of these parameters is expected to reduce the batch inhomogeneity and, consequently, should get attention in a next step of development.

Further, a detail analysis of sulphur tolerance was performed at Chalmers, CSIC and SINTEF using several S-precursors and the C28E1S2 oxygen carrier. It was found that the chemistry is very complex, but that the material has an affinity for sulphur at reducing conditions, thus resulting in a deactivation. This effect is very dependent on the level of oxygen-carrier conversion and temperature: for fuels with high S the C28 material is unable to perform, but for low concentrations of S the oxygen carrier could be used.

2.2 Oxygen carrier material produced by impregnation

The premise behind the choice of oxygen carrier in this current work builds on from the formulations of the impregnated iron- and copper- based oxygen carriers identified within the FP7 INNOCUOUS project. For SUCCESS the stipulation was that the raw materials selected for oxygen carrier scale-up in this project were industrially-relevant, that is they are or could potentially be available for supply at multi-tonnage scale. Currently, limited options exist for such industrially-relevant fluidizable supports within the particle size range relevant for this project (nominally 100 - 300 micron). Nevertheless, several supports were sourced from commercial suppliers for performance screening and comprised of "pure" alumina, silica/alumina and zirconia. (The exact product details of the supports and their suppliers is deemed proprietary information within the SUCCESS Consortium, so is not disclosed in this report).

Oxygen carrier formulations were produced by the incipient wetness impregnation technique using industrially-relevant protocols and equipment. For iron-based materials, a target active metal oxide loading of 20 wt.% Fe_2O_3 was chosen based on prior knowledge from ICB-CSIC and the INNOCUOUS project, whilst for copper-based this was 15 wt.% CuO. Initially, small samples (up to 150 g) of the materials were produced by JM for screening at ICB-CSIC using such techniques as TGA, XRD, mechanical crush strength and ASTM (D5757) air-jet attrition. Four iron- and one copper- based materials were ultimately selected for scale-up to 5 kg and tested by ICB-CSIC in their 500 W_{th} continuous-CLC reactor. These were compared with impregnated benchmarks derived from ICB-CSIC and the INNOCUOUS project (produced using a research grade γ-alumina that wouldn't be available at multi-tonnage scale). The materials were mainly made by JM in their Manufacturing Science Centre (MSC) located in Billingham, UK.

During the selection process approximately 307 h of hot operation was achieved from the 500 W_{th} unit using methane as the fuel gas. Based on the testing, a selected iron-based formulation using a commercial γ-alumina support was deemed suitable for an initial scale-up to 25 kg for small-pilot scale evaluation in Chalmers' 10 kW_{th} CLC reactor unit. However, the 10 kW_{th} evaluation experienced a high degree of particle elutriation and did not achieve even close to full fuel conversion and meanwhile a copper-based formulation on a commercial γ-alumina support had proven more promising upon testing in the 500 W_{th} unit. Compared to the aforementioned iron-based material, the copper-based one exhibited a lower oxygen carrier-to-fuel ratio to achieve full methane combustion and more significantly a much longer projected lifetime, based on its observed attrition rate in the 500 W_{th} unit (5000 h versus 1000 h). In fact, the copper-based oxygen carrier compared favorably with the impregnated copper benchmark material (Cu14γAl ICB, developed previously by ICB-CSIC) in terms of combustion efficiency and, moreover, exceeded it by displaying double the projected lifetime. For this reason, the copper material was ultimately selected for the main material scale-up activity in the project.

Figure 6 shows the methane combustion efficiency and attrition behavior of the selected copper-based oxygen carrier (15 wt.% CuO/Al_2O_3), designated in the chart as Cu_Imp_SUCCESS, in the 500 W_{th} unit.

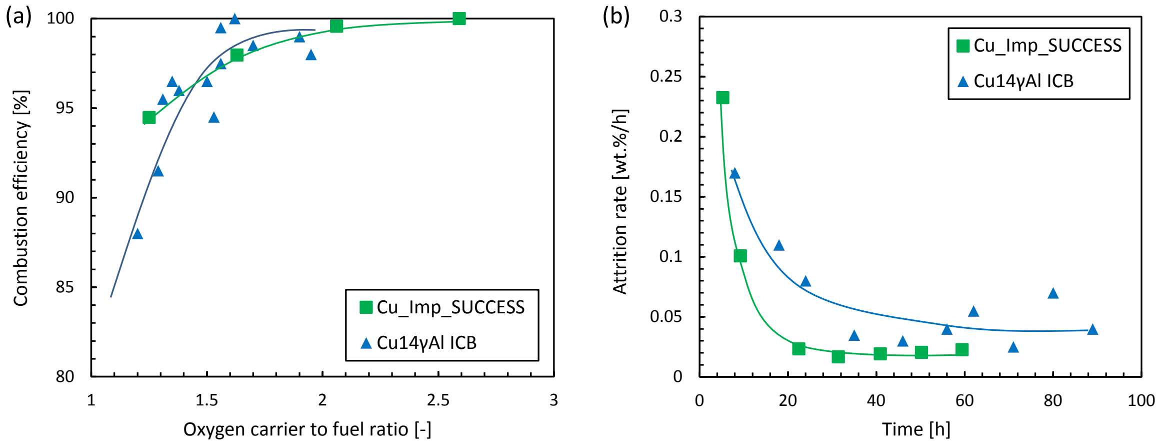


Figure 6: (a) Effect of the oxygen carrier-to-fuel ratio on the methane combustion efficiency in the 500 W_{th} CLC unit with the Cu_Imp_SUCCESS oxygen carrier. Cu14-γAl ICB oxygen carrier is taken as reference material. TFR= 800°C, TAR= 800°C. (b) Attrition behavior of the Cu_Imp_SUCCESS oxygen carrier tested in the 500 W_{th} CLC unit.

Table 2 depicts supporting characterisation and assessment data for the fresh and used (reactor discharged) material.

Table 2: Testing of the Cu_Imp_SUCCESS oxygen carrier in the 500 W_{th} CLC unit at 800°C.

Parameter	**Unit**	**Fresh**	**Discharged (60 h hot)**
Active CuO content (a)	wt.%	13.6	10.1
Oxygen transport capability	(-)	2.7	2.0
Porosity	%	53.0	49.1
Crushing strength	N	1.2	1.2
Air jet attrition index	%	3.1	2.6
XRD major phases	n/a	CuO, γ-Al_2O_3	CuO, $CuAl_2O_4$, α-Al_2O_3, θ-Al_2O_3

(a) Determined by TGA with H_2 as reducing gas

In terms of sulphur tolerance then both the selected iron- and copper- based oxygen carriers were shown to be resistant, based on testing in the 500 W_{th} unit. Up to 3000 ppm(v) of H_2S was used as the model sulphur species in the methane fuel gas, with 33 and 38 hours of continuous operation achieved for the iron- and copper- based materials respectively. The presence of sulphur had insignificant impact on the combustion efficiency for both materials and all of the sulphur was emitted as SO_2 from the fuel reactor exhaust only, with no observed accumulation within the particles.

The 15 wt.% CuO on a commercial γ-alumina (Cu_IMP_SUCCESS) was produced by JM in their MSC at a total quantity of 500 kg and supplied to Vienna, SINTEF ER and IFPEN for evaluation in their pilot-scale CLC units. The methodology for producing the material was identical to that employed for the smaller batches using industrially-relevant equipment for a scalable manufacturing process.

2.3 Oxygen carrier material produced by granulation

With the impetus to reduce oxygen carrier costs the potential of making oxygen carriers via a granulation route, utilizing industrially-relevant, low cost raw materials has been explored within SUCCESS. Granulation is a well-established industrial manufacturing process for producing materials at multi-tonnage scale.

The technique of (wet) granulation employed for these oxygen carriers typically involves combination of the powder precursors, which are then mixed under high shear whilst water is incrementally added until the desired particle size is achieved.

In total 74 granulated oxygen carrier samples with a particle size range 100-300 micron were prepared by JM at >100 g, using a high shear granulator of relevance for gaining knowledge about potentially scaling the manufacturing process. This was performed in JM's MSC. All materials prepared either targeted a 15 wt.% CuO content or 20 wt.% Fe_2O_3 content akin to those impregnated materials previously made within SUCCESS for comparison. With the selected impregnated materials, the only other main species present was alumina (the support) at 85 wt.% or 80 wt.% respectively whilst for the granulated materials the approach taken was to aim for a similar main active metal content but allow for variation in the support composition, not just to use an alumina.

Granulated samples that were deemed suitable (from a basic visual inspection after being made and then sieved) were preliminarily evaluated in JM's batch fluidized quartz tube reactor (15 g sample) with 200 high temperature (800-950°C) redox cycles using a methane-based fuel gas. Subsequent downselected samples were assessed by ICB-CSIC using TGA, mechanical crush strength and ASTM (D5757) air-jet attrition tests.

As a result of the preliminary screening at JM and CSIC, two candidate 15 wt.% CuO granulated oxygen carriers were selected for scale-up to 5 kg. One has an Al_2O_3-based support, labelled Cu_GR_03, which exhibited a similar reactivity to the SUCCESS impregnated material, and has the lowest AJI of the different alumina-based granulated samples. The other is labelled Cu_GR_04 and has a Ca-based support, which has the lowest AJI of all the granulated samples analyzed. These samples were subsequently produced by wet granulation each at 5 kg total quantity, using high shear granulation equipment in JM's MSC.

The two oxygen carriers were both evaluated in ICB-CSIC's 500 W_{th} continuous-CLC unit using a methane fuel gas, under test conditions similar as for the impregnated oxygen carriers. They were compared with the SUCCESS impregnated copper-based oxygen carrier, Cu_Imp_SUCCESS. An example of performance in the 500 W_{th} unit is depicted in Figure 7 for the Cu-GR-04 oxygen carrier.

The oxygen carriers, both fresh and used (reactor discharged) also underwent supporting characterization and assessment at ICB-CSIC using TGA, mechanical crush strength, ASTM air-jet attrition, XRD, SEM-EDX and CO chemisorption techniques. Some of this information is displayed in Table 3 for the Cu-GR-04 oxygen carrier.

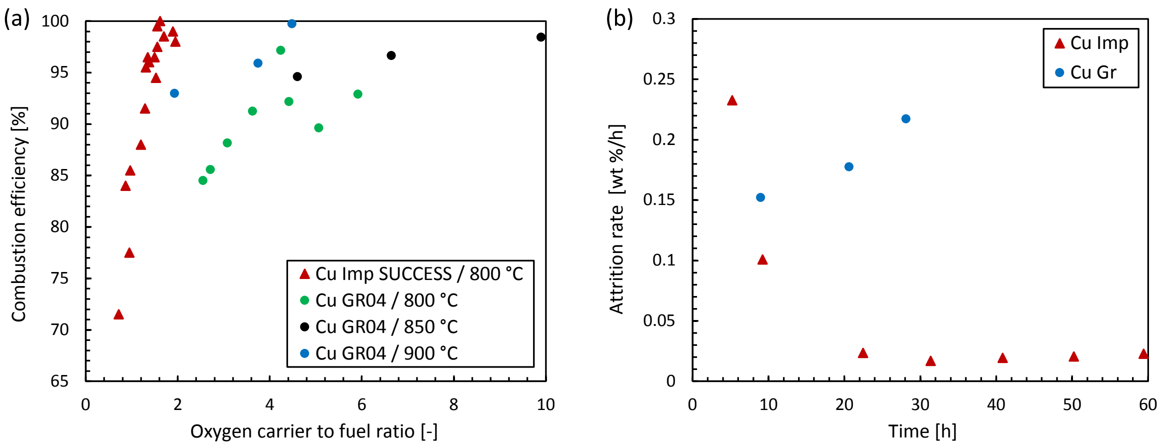


Figure 7: (a) Effect of the oxygen carrier-to-fuel ratio on the methane combustion efficiency in the 500 Wth CLC unit with the Cu_GR_04 oxygen carrier at different Fuel Reactor temperatures. Cu_Imp_SUCCESS oxygen carrier is taken as reference material. (b) Attrition behavior of the Cu_GR_04 oxygen carrier tested in the 500 W_{th} CLC unit.

In summary the Cu-GR-03 oxygen carrier exhibited good performance for methane combustion, similar to that observed with the copper-based impregnated material, although higher oxygen carrier-to-fuel ratios are needed for full methane combustion ($\phi > 3$). However, unlike for the impregnated material, copper loss was not apparent over time. Regarding the attrition behavior, the attrition rate of the Cu_GR_03 oxygen carrier was very high throughout 14 hours of continuous operation (~0.55 wt.%/h). The corresponding projected particle lifetime was less than 200 hours. This value is significantly lower than the particle lifetime for the reference impregnated material (5000 h).

Table 3: Testing of the Cu-GR-04 oxygen carrier in the 500 Wth CLC unit.

Parameter	Unit	Fresh	Discharged (27 h hot)
Active CuO content (a)	wt.%	14.7	15.2
Porosity	%	26.4	26.6
Crushing strength	N	3.1	1.9
Air jet attrition index	%	4.3	4.9
Mean particle size	µm	212	206

(a) Determined by TGA with H_2 as reducing gas

The Cu-GR-04 needed a higher oxygen carrier to fuel ratio ($\phi > 5$) and FR temperature (900°C) to reach full methane combustion. Again, copper loss was not experienced over time and observed deactivation was suggested to be due to a decrease in copper dispersion and active surface area. Although exhibiting a suitable AJI for use as a CLC oxygen carrier, which interestingly remained constant, the inferred particle lifetime of the granulated material (500 h) from CLC testing is ten times lower than the impregnated one (5000 h), evaluated after 28 hours of continuous operation.

Overall, the granulated copper-based oxygen carriers have shown sufficient promise to merit further investigation aiming to improve their technical performance. With a main drive for a granulated material being a lower price, an indicative reduction would be a market selling price of approximately half that of the equivalent copper-based impregnated material. This is mainly due to the lower raw materials cost for the granulated oxygen carrier.

3 Oxygen carrier testing

3.1 Testing in pilot units up to 150 kW

3.1.1 Testing at 10 kW_{th} (Chalmers)

Chalmers has tested three oxygen carrier materials in their 10 kW_{th} circulating CLC pilot. Two materials were based on the spray-dried calcium manganite-based oxygen carriers C28E1S2-1335, produced by VITO, and C28-901-TR1136, produced by Euro Support. In addition, an impregnated iron-based oxygen carrier produced by Johnson Matthew was investigated in the unit. An overview over the different materials and experimental parameters is shown in Table 4.

Table 4: Experimental parameters for the three oxygen-carrier materials investigated

Parameter	C28-901-TR1136	C28E1S2-1335	Fe-based
Nominal composition of oxygen carrier	$CaMn_{0.775}Mg_{0.1}Ti_{0.125}O_{3-\delta}$	$CaMn_{0.775}Mg_{0.1}Ti_{0.125}O_{3-\delta}$	20 wt% Fe_2O_3 80 wt% γ- Al_2O_3
Bulk density of fresh oxygen carrier	1240 kg/m^3 (poured)	1240 kg/m^3 (poured)	1090 kg/m^3 (tapped)
Mean size of oxygen carrier particles	143 µm	137 – 151 µm	≈ 90 – 150 µm
Fuel input (methane)	4.8 – 8.9 kW_{th} (8 – 15 L_n/min)	1.2 – 6.0 kW_{th} (2 – 10 L_n/min)	3.2 – 5.8 kW_{th} (5 – 9 L_n/min)
Fuel-reactor temperature	912 – 975°C	850 – 980°C	900 – 970°C
Superficial gas velocity in riser (air)	1.9 – 2.2 m/s (130 – 150 L_n/min)	2.0 – 2.9 m/s (160 – 190 L_n/min)	≈ 2 – 3 m/s (160 – 180 L_n/min)
Solids inventory	≈ 16 kg	≈ 8 – 15 kg	≈ 9 – 12 kg
Bed mass in fuel reactor (theoretic)	≈ 3.5 kg	≈ 2.8 kg	≈ 3.2 kg
Specific fuel-reactor bed mass	≈ 320 – 990 kg/MW_{th}*	≈ 470 – 2300 kg/MW_{th}†	≈ 620 – 1650 kg/MW_{th}†
Total time fluidized under hot conditions (here: > 600°C)	603 h	193 h	40 h
Total fuel operation time (methane)	110 h	24 h	30 h
Reference	Deliverable 5.5	Deliverable 4.2	Deliverable 4.5

* … based on estimated bed mass in fuel reactor, which is based on a pressure measurement

† … based on theoretic bed mass in fuel reactor

In each campaign, fuel conversion properties as well as attrition resistance were assessed. Based on the attrition resistance the lifetime of the particles was estimated. Figure 8 shows fuel conversion of the different materials at varied fuel input, i.e., specific fuel-reactor bed mass, and varied temperature. As is seen from the figure, the material C28E1S2-1335 was able to achieve comparable fuel conversion as the reference materials, produced in the previous INNOCUOUS and CLC Gas Power projects. For the up-scaled material, the gas yield was lower, as is seen in the figure. The impregnated Fe-based material as well as the up-scaled perovskite-based material achieved significantly lower fuel conversion, i.e., usually between 70% and 85%. This was one of the reasons why it was decided to focus on Cu-based materials.

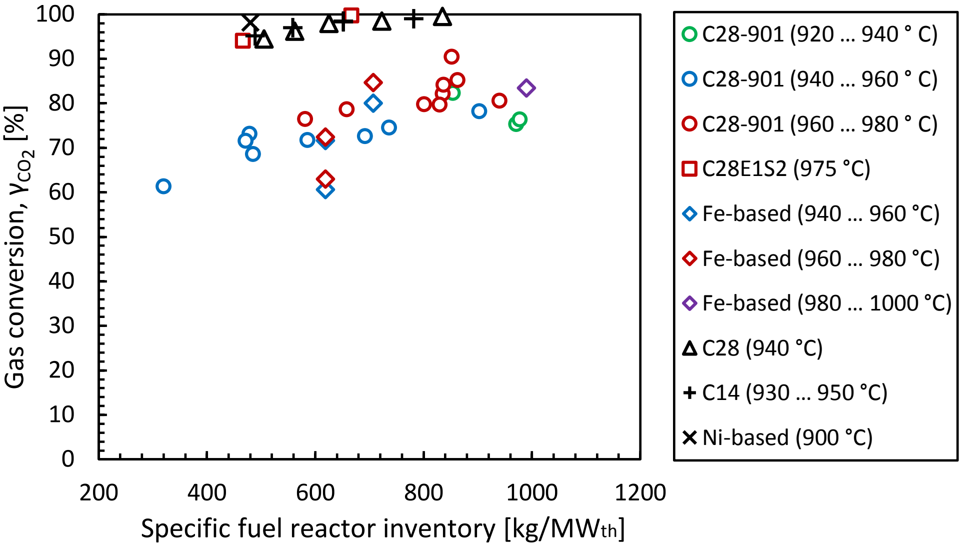


Figure 8: Gas conversion, γ_{CO_2}, as a function of specific fuel-reactor inventory (bed mass FR/fuel input) and fuel-reactor temperature. Different symbols stand for different materials and different colours for different temperature intervals. The three materials investigated in the SUCCESS project are shown for different temperature intervals, whereas the reference materials from the projects INNOCUOUS and CLCGP are shown for one temperature interval.

Prior to fuel operation, when fuel reactor and loop-seals were in inert conditions, a clear release of gas-phase oxygen was observed, i.e., the CLOU effect. The amount of oxygen that is released is believed to depend on temperatures, residence times and oxygen partial pressures in both the air and fuel reactors. For the conditions assessed with the materials C28-901 and C28E1S2 the oxygen partial pressures in the fuel reactor were usually between 1 mol% and 6 mol%.

The attrition rates during fuel operation were 0.15 wt%/h, 0.02 wt%/h and 0.14 wt%/h for the materials C28-901-TR1132, C28E1S2-1335 and Fe-based, respectively, which correspond to the extrapolated lifetimes of 700 h, 5000 h and 700 h.

3.1.2 Testing at 10 kW$_{th}$ (IFPEN)

Chemical looping combustion process performances of the copper based material (CuO/Al_2O_3) and the C28 perovskite material ($CaMn_{0.775}Mg_{0.1}Ti_{0.125}O_{3-\delta}$) from WP3 have been studied in IFPEN's 10 kW pilot plant for methane combustion. The impact on the process performance of the fuel reactor temperature, solid flow rate, fuel reactor inventory, material ageing and methane concentration has been investigated. A good circulation was obtained with both materials even though the copper based material properties was close to the A class of Geldart's classification.

For the copper based material, complete conversion of methane was observed at 900 and 850°C. At 900°C, a specific solid flow rate of 3.4 kg/MJ with a specific fuel reactor inventory of 2799 kg/MW were necessary to reach full conversion. The corresponding $R_0\Delta X$ value of the material was around 2.25 wt%. At 850°C, a $R_0\Delta X$ value of 1.61 wt% was required for total methane conversion, using a solid flow rate of 5 kg/MJ and a fuel reactor inventory of 2799 kg/MW. Other test results show that the copper based material has a strong catalytic activity when its degree of reduction is too high, which favors the production of carbon monoxide and hydrogen, decreasing the performance of the process. The operating conditions have to be optimized in order to ensure a sufficiently high ratio between the quantity of oxygen released by the material and the quantity of oxygen necessary for full methane combustion. The operating conditions' impact on process performance is summarized in the Table 5. Besides, the ageing test has shown a quick degradation of the material over the 160 h of material cycling, which could lead to an increase of the loss of fines for longer test duration and decrease the performance of the process and the material lifespan estimated in this work (9000h based on stabilized loss of fines).

Table 5: Summary of the operating conditions impact on the process efficiency for both oxygen carrier

Augmentation of :	**Impact in this way the performance of the process with :**		
	Copper base material		**Perovskite material**
Methane concentration	$[CH_4]<22\%$	$[CH_4]>22\%$	$R_0\Delta X \nearrow$ $X_{CH4} \searrow$ $CO/CO_2=0$
	$R_0\Delta X \nearrow$ $CO/CO_2 \searrow$ $X_{CH4} \nearrow$	$R_0\Delta X$ cste $CO/CO_2 \nearrow$ $X_{CH4} \searrow$	
Solid flow rate	$R_0\Delta X \nearrow$ $CO/CO_2 \searrow$ $X_{CH4} \nearrow$		$R_0\Delta X \searrow$ $X_{CH4} \nearrow$ $CO/CO_2=0$
Temperature	T<900 °C	T>900°C	$R_0\Delta X \nearrow$ $X_{CH4} \nearrow$ $CO/CO_2=0$
	$R_0\Delta X \nearrow$ $CO/CO_2 \searrow$ $X_{CH4} \nearrow$	$R_0\Delta X \searrow$ $CO/CO_2 \nearrow$ $X_{CH4} \searrow$	
OC mass in FR	No effect under our conditions		$R_0\Delta X \nearrow$ $X_{CH4} \nearrow$ $CO/CO_2=0$

For the perovskite material, the tests were performed with a higher gas flow rate in the fuel reactor inlet than for the copper based material in order to take into account the difference of density between the two materials (a gas velocity of three times the minimal fluidization rate is required for each material). It induces a shorter contact time of the gas with the oxygen carrier than for the copper based material. As a consequence, methane conversion was limited by the contact time and complete methane conversion could not be reached. Thus, changes to operating conditions leading to an increase of the contact time between gas and solid also increase the performance of the process (See table above). The 47h of material cycling of the ageing campaign has not shown any process performance loss and a particle lifetime are estimated from the loss of fines. This lifetime is equivalent to the copper based material. However, particles analysis shown formation of cracks which would certainly lead to a fractionation of the particles at longer operation times. Finally, the two campaigns with perovskite material had to be stopped due to agglomeration issues. These issues seem to be due to an interaction between the perovskite material and a small proportion of copper based material, which was still present despite cleaning of the unit, possibly favoring the melting of the perovskite in the fuel reactor (in reductive atmosphere).

According to the results presented in this study, the copper based material is more reactive than the perovskite material, which confirms the results presented in the kinetic study presented in SUCCESS' deliverable D6.1. This difference in reactivity leads to lower methane conversion with the perovskite material which may be emphasize by the difference in gas residence times in the fuel reactor for both materials. However, the copper based material degradation over time and its high catalytic activity must be avoided. With the perovskite material, no decrease of performance over time was observed over the 47 hours of operation, but the particles began to split-up and complete conversion of methane was not reached during the tests. Except for the agglomeration issues, the circulation of both materials was stable and they have shown interesting performances for the methane chemical looping combustion application.

As a conclusion, both material should be suitable for Chemical looping process application but an optimization of the operating conditions should be carried out to reduce their degradation and avoid the process performance decrease over time. Nevertheless, the ratio solid/gas flow rate must not be increased too much in order to avoid process efficiency loss at industrial scale.

3.1.3 Testing at 120 kW_{th} (Vienna)

At the very beginning of SUCCESS the 120 kW_{th} pilot plant has been adapted. The data basis for this modification has been generated during the FP7 project INNOCUOUS. A sufficiently high global solids circulation rate and high enough active inventories in the reactors, especially in the fuel reactor, were the target of the adaptation. The new fuel reactor design consists of a wider bottom area, to increase the volume of the dense zone of the fluidized bed for high solids inventories. The upper part of the fuel reactor is narrower than the old one to increase the internal circulation in the fuel reactor and good gas solid contact over the full height of the reactor. To allow high global solids circulation rates the diameter of the air reactor has been slightly decreased to reach higher gas velocities and correspondingly higher solids entrainment.

The new reactor design has been tested with a well known oxygen carrier produced in INNOCUOUS (a mix of C14 and C28). The data obtained was used to benchmark the new reactor design against the old one. This should help to compare the oxygen carriers produced in SUCCESS with other materials tested in the old 120kW unit. The design demands have been reached during the testing. The global solids circulation rate is in the range of or slightly higher than the old design. The specific inventory in the fuel reactor can be raised to larger amounts of solids in the reactor. The results of the inventory variation during the benchmark testing and a comparison with the results from the INNOCUOUS project, are depicted in Figure 9.

The Cu15 material is an impregnated copper based oxygen carrier produced in SUCCESS by JM. As a first step of particle characterisation, the Cu15 has been tested with focus on the overall CLC performance. In a second experimental campaign its stability against sulfur in the form of H_2S in the fuel feed has been evaluated. Natural gas was the fuel during the CLC performance testing, the fuel reactor temperature was 800°C. The experiments highlighted a significant influence of the air reactor residence time on fuel conversion performance. This parameter can be influenced either with high solids inventories or low air/fuel ratios. A variation of the fuel power specific inventory highlighted the need for sufficient solids inventories in both reactors. A comparison with a similar copper based oxygen carrier (CCP3) tested in the old 120kW pilot unit, showed better fuel conversion with the Cu15 material. The dependency of the specific inventory and the comparison with the other copper based oxygen carrier is shown in Figure 9. As shown in Figure 9, 95% fuel conversion have been reached during this test. Operating conditions with nearly full conversion (>98% methane conversion) were either at elevated temperatures (880°C) or with high specific fuel reactor inventory (214 kg/MW) and low air/fuel ratio of 1.1 at 800°C. Apart from the basic performance testing, the effect of sulfur in the fuel feed has been evaluated. Two test runs have been performed with varying H_2S levels in the fuel feed. Gathering information of the development over time of the oxygen carrier performance was a major goal of the testing. The H_2S levels in the fuel feed were in the range of 500 – 2000 ppmv. The fuel power was 70 kW and the fuel reactor temperature 800°C. For all the different sulfur feedings, a drop in fuel conversion performance at the beginning was noticeable. After this first drop, the conversion remained stable for the rest of the test. During the sulfur feeding no H_2S slip has been detected in the off gas streams. Only SO_2 has been detected in the fuel reactor off gas stream. The fuel conversion has been validated after the H_2S feeding to identify a potential ageing effect of the oxygen carrier. Fuel conversion increased after the stop of H_2S and along with an operating temperature increase to 850°C fuel conversion was at about 95%. The conclusion of this testing is a certain suitability of the Cu15 material for sulfur containing fuels with some remarks. First, the fuel performance drops if sulfur is present and second, higher operating temperatures should be considered for this type of fuel.

The C28 material tested at Vienna has been produced by ESAM. Vienna received two batches, one based on production in a smaller size and the second one was part of the large batch, also tested in the 1MW unit at TUD.

The testing was divided into general CLC performance and sulfur tolerance, like for the Cu15. In contrast to the Cu15, the air reactor residence time has no influence on the performance of the material. The effect of increasing oxygen partial pressure in the air reactor at higher air/fuel ratios was strong, though. The fuel conversion increased correspondingly. The specific fuel reactor inventory has a significant influence on fuel conversion performance (compare Figure 9). The C28 material has been tested mainly at a fuel reactor temperature of 950°C. Operating conditions with nearly full fuel conversion (>98%) were at high specific fuel reactor inventories (245 – 285 kg/MW) and high air/fuel ratios (>1.8).

The fate of sulfur has been examined during two test runs. In the first run, H_2S has been added to the fuel up to a level of 100ppmv and a drop in fuel conversion was noticeable. In the second run the concentration has been raised to nearly 2900ppmv and the duration of the sulfur feeding was more than 4 hours. This was done to identify a potential accumulation of sulfur on the particles. The performance of the oxygen carrier steadily decreased from nearly full fuel conversion to below 90% during the 4 hours of H_2S feeding. During the whole test only SO_2 was detected from the fuel reactor exhaust gas stream. No other sulfur emission has been detected. After the sulfur feeding no regeneration of the oxygen carrier was noticeable.

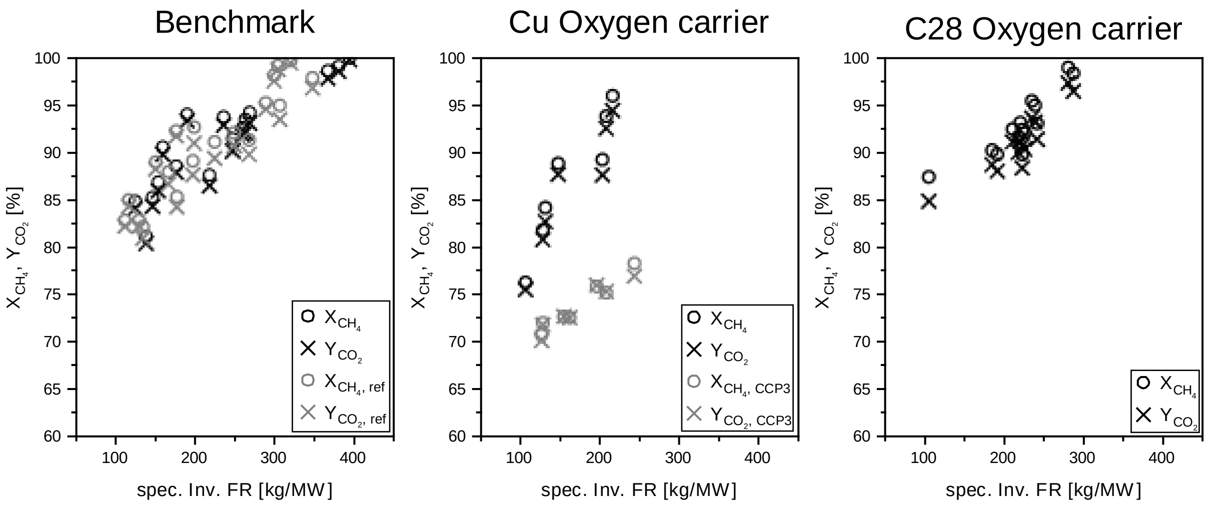


Figure 9: Results of the Benchmark testing (black: SUCCESS, grey: INNOCUOUS results), the Cu15 testing (black: SUCCESS, grey: other copper based OC) and the testing of the C28 material.

3.1.4 Testing at 150 kW$_{th}$ (SINTEF ER)

The 150 kW CLC reactor system at SINTEF Energy Research in Trondheim, Norway, has been tested with two different types of oxygen carrier particles developed and fabricated within SUCCESS; a copper oxide based material and a calcium-manganese-titanium (CMT) type material. The testing was performed as one-day tests and since the heat-up sequence takes some time the duration in CLC mode was up to 4-5 hours. The tests have been analyzed by exhaust gas analyzers and recordings of reactor pressures and gas flow controllers. Test data were recorded at different reactor inventories, temperatures and gas flows, but the main focus has been to identify the ideal condition for maximum fuel conversion.

The main findings were that the copper oxide particles performed very well in the reactor. A methane conversion of above 98% and an oxygen demand (the ratio of oxygen lacking to achieve complete combustion to the stoichiometric amount of oxygen needed) of 1 – 2% was achieved. The specific fuel reactor inventory was then about 120 kg/MW, which can be considered low. The tests with the CMT particles were less successful, as the methane conversion was poor and the temperature could not be maintained in CLC mode. The maximum methane conversion was about 75% for a period, but it dropped as the temperature in the fuel reactor (about 940°C) could not be maintained. It should be noted that the 150 kW CLC reactor at SINTEF is not equipped with external heating and a self-sustained auto-thermal operation is required. In the tests with the CMT, the fuel power and conversion was not enough to supply the needed heat to keep the temperature at appropriate level.

A better fuel conversion could be expected if it was possible to maintain a higher temperature, by e.g. using some external heating of the fuel reactor. Also, the CMT experiments should have been performed with higher gas velocities and/or smaller particle size in order to achieve CFB mode of operation and associated improvements in solids transport and gas-solid mixing. The CMT oxygen carrier particles fabricated in SUCCESS have a size and density that is somewhat outside the design values for the 150kW CLC unit.

Several improvements have been made to the rig. The particle collector system below the exhaust heat exchangers was replaced with a type that made it possible to empty them during operation and refill lost particles. In addition, water scrubber units were added to each exhaust pipe for increased exhaust cooling performance. During initial tests, it became clear that the heat-up sequence had to be improved and provisions for feeding hydrogen to the fuel reactor have been implemented to the system. The costs for these improvements have mostly been covered by a national project whereas all operational costs for the above described testing have been covered by the SUCCESS project.

3.2 Scale-up of Chemical Looping Combustion to 1 MW_{th} (TUD)

The objective of tests in 1 MW_{th} scale was the demonstration of CLC with gaseous fuels in semi-industrial scale. The 1 MW_{th} CLC pilot plant at TUD was originally designed to utilize solids fuels [1]. To enable the pilot plant for gaseous fuel operation, some adaptations of the design were made (cf. Figure 10). A flue gas recirculation was installed to simulate real conditions of a full scale CLC plant and to simplify operation. Natural gas was compressed to a pressure level of 2.5 bar(g) by a new natural gas compressor (not shown in Figure 10) and was introduced to the fuel reactor at two different heights. The secondary fluidization of the fuel reactor should enable both a high solids inventory of the fuel reactor and a high solids circulation between fuel and air reactor. Additional measurement equipment was installed to determine dry gas concentrations and moisture.

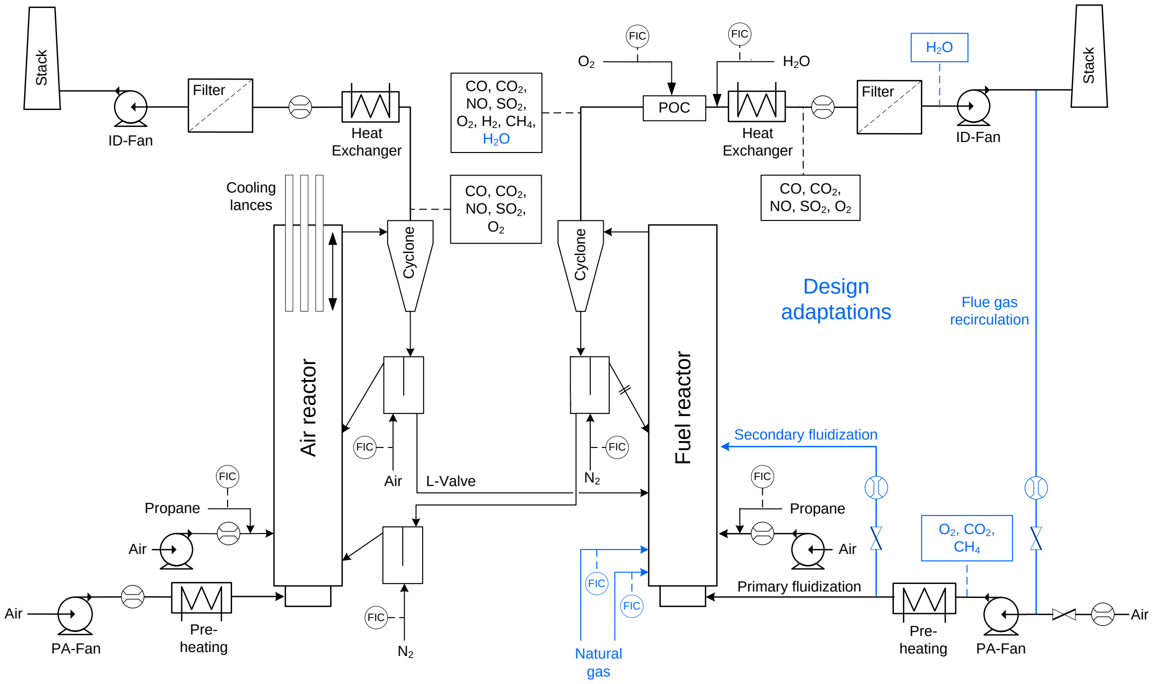


Figure 10: Simplified scheme of the 1 MW$_{th}$ chemical looping pilot plant (modifications for test campaigns with natural gas as fuel shown in blue).

Results were obtained during two test campaigns. The flue gas recirculation worked excellently, and the specific inventory was slightly increased by the utilization of the secondary fluidization. Additional measurements enabled the determination of different mass flows by means of venture nozzles. Stable CLC conditions were reached for more than 50 h during the first test campaign with the oxygen carrier C28 (LOT #902-1 and #902-2). These results present the first results of gaseous fuel CLC at MW$_{th}$ scale. During a “6 h reference point” (cf. Table **6)** only incremental adjustments were made. Hence, this point was used for further evaluation. The pilot was operated autothermally for half an hour. This means that no fuel/propane was added to the air reactor and the heat for the process was provided exclusively by CLC (except for preheating of gases, cf. Figure 10).

The fuel conversion in the fuel reactor was in the range of 80 %. The conversion was positively influenced by a higher air/fuel-ratio in the air reactor and a lower fuel power/feed to the fuel reactor. Major changes of the operation conditions were not possible due to the design of the pilot plant. Many samples were taken from fuel and air reactors to investigate the evolution of the material and the solids circulation. The circulation of solids was derived from the ratio of provided oxygen in the fuel reactor and the difference of the corresponding degrees of oxidation. This difference was between 1.0 and 1.4 % and the solids circulation was in the range of 17 tons h^{-1} MW^{-1} which is significantly higher than in former tests [2]. This was enabled mainly by a larger cross sectional area of the L-valve, whose refractory lining was renewed before CLC tests (thereby the area was increased by 70 %). Pure oxygen was introduced to the post oxidation chamber (POC), and the hot flue gas was cooled directly downstream by a water injection. Almost full conversion of gases like CH_4, CO and H_2 was achieved in the POC for the whole test duration. The performance of the POC was important for the process because recycled flue gas was used for fluidization of the fuel reactor, and high amounts of unburned gases should be avoided. Attrition was determined from the fine fraction of samples from the fabric filters and in a cold attrition test rig. The determined attrition of the material was in the range of 0.2 wt.-% h^{-1}. This corresponds to a particle lifetime of ~ 500 h. It can be reasonably assumed that the refractory lining of the 1 MW$_{th}$ unit had a great impact of the attrition rate because attrition of this material was lower in other units made from stainless steel. No agglomeration or blockage of particles was detected during the whole test. The mean diameter of all samples from the fluidized bed reactors was rather constant (dp_{50} ~ 125 µm).

The most important data that were collected during a "6 h reference point" are summarized in Table **6**. Data from the 120 kW_{th} unit from Vienna are shown for comparison reason. In general, data from the 1 MW_{th} pilot are in accordance with data from the other unit. The higher conversion of natural gas of the 120 kW_{th} unit (almost full conversion instead of 75 %) can be explained by different reasons. Higher specific inventory of the fuel reactor and higher air/fuel ratio are the main reasons. Further positive factors are the high methane concentration (100 instead of 13 Vol.-% (wet)) used for fluidization of the fuel reactor or the higher solids circulation. Higher solids circulation lead to a higher oxygen carrier-to-fuel ration and the CLOU effect can be used to a greater extent.

Table 6: Performance results for high grade steam reference case.

Description	Unit	1 MW_{th} (TUD) 6 h reference point	120 kW_{th} (TUV) Full conversion
FR temperature	°C	970	955
FR fuel power	kW	780	60
FR specific inventory	kg MW^{-1}	65	278
FR gas velocity	m s^{-1}	6.1	2.3
AR temperature	°C	1030	955
AR specific inventory	kg MW^{-1}	63	107
AR gas velocity	m s^{-1}	4.9	8.6
Conversion of natural gas in the FR	%	75	98
λ (air/fuel ratio)	-	1.1	1.8
Solids circulation	kg h^{-1} MW^{-1}	17300	64200
CH_4 concentration in FR feed	Vol.-% (wet)	13	100
POC CH_4 conversion	%	99	-
Attrition (particles < 40 µm)	wt.-% h^{-1}	0.2	-

The main objective of the second test campaign was the investigation of operation with an oxygen carrier mixture of C28 and ilmenite. Used C28 was used for start-up of the pilot and stable CLC conditions were reached. In the following, ilmenite was used as make-up material because no further C28 oxygen carrier was available. The performance with a mixture of ilmenite and C28 was significantly lower compared to operation with only C28. This can be explained by the lower reactivity of ilmenite with methane and the negligible CLOU effect of this material.

3.3 Oxygen carrier lifetime and attrition behavior

The mechanical strength of particles is an essential criterion in the process design since it has a direct impact on the oxygen carrier lifetime and thus the OPEX of the CLC plant. Therefore, several tasks of the project were dedicated to study the attrition behavior of the two chosen oxygen carriers using different testing procedures at room temperature and in hot operating conditions, with or without fuel injection.

SINTEF has compared cold and hot attrition of the materials produced in SUCCESS, reported in D4.4 in more details. Results show that cold attrition gives good lifetimes for both materials produced, in the range of 6200h for C28 and 11000h for Cu/Al_2O_3. This is good numbers for attrition especially for C28 from Euro Support where lower sintering temperature had to be used for the commercial batch compared to optimal conditions given by VITO. In comparison, a lifetime of 33000h was estimated with the

$Ni/NiAl_2O_4$ material which is used as standard since a lot of testing has been performed on this material and a relatively good lifetime prediction was made in fluidized rigs. The hot attrition results show a different trend for the materials, with a longer lifetime for C28 than for Cu/Al_2O_3, 22000h and 7000h, respectively. This can be related to Cu being more vulnerable for surface attrition of the Cu segregated on the surface, while C28 can possibly get some stress relief during the hot redox making it stronger. It is important to stress that this short experiments tries to predict long-term behaviour (lifetime) and that critical failure mechanism that might occur after some time might not be seen in such experiments.

At IFPEN, ageing campaigns have been performed on the 10 kW_{th} CLC pilot unit, following fines production over time under combustion of methane (more details in D5.3). The results based on stabilized fines production rate predict a good lifetime for both materials in the range of 9000 h for the CuO/Al_2O_3 material and 12 000h for the C28 material. However, SEM and mercury porosity measurements indicate a progressive degradation of both materials with the formation of cracks inside the particles. Additional attrition measurements were then conducted on fresh and used materials using a jet cup rig (more details in D4.6). The results indicate that the attrition index of the fresh C28 material is several times smaller than that of the CuO/Al_2O_3 material which means that it is initially more resistant to attrition, probably because of the high sintering temperature applied for its production. Samples taken after test campaigns show a dramatic increase of the attrition index for the CuO/Al_2O_3 material which indicates that the cracks inside the particles strongly lowered its attrition resistance. On the contrary, a limited evolution is observed for the C28 resistance to attrition, but this result must be tempered as the ageing conditions were less severe than for the CuO/Al_2O_3 material.

Chalmers has tested the oxygen carrier material C28 (spray drying batch 901, average calcination ring temperature 1136) for more than 100 h with continuous addition of methane, see Figure 11. Due to an unforeseen leak upstream of the fuel reactor, the actual flow in the fuel reactor was decreased to about 1/3 during a significant period of operation, from 9-79 h. The total flow still exceeded minimum fluidization, and there was clear circulation of material. However, there was clear agglomerations in the bed, likely formed during this period, and hence it is possible that the particles were affected in a negative way during this period of time. After 79 h, oxygen-carrier lifetime decreased by two orders of magnitude. At comparable conditions and after 1000 h of operation, the reference Ni-based oxygen carrier had an estimated lifetime of 33 000 h, whereas the lifetime of the C28-901 material was only 700 h.

Additional attrition tests of C28-901 and the Ni-based reference material were carried out in Chalmers customized jet-cup test rig. The attrition rates determined here are usually much higher than those determined under hot CLC condition. However, they follow the same trends and mirror attrition in the CLC unit well.

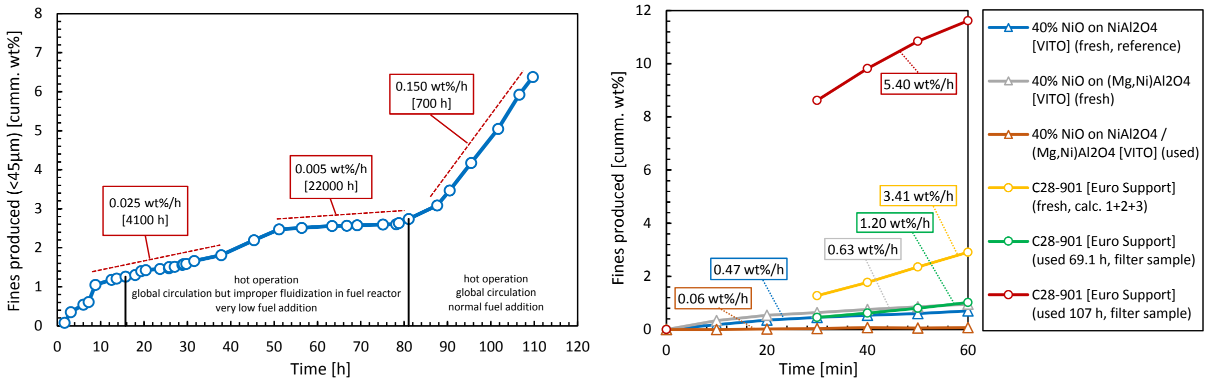


Figure 11: Attrition testing in 10 kW CLC unit (left) and in customized jet-cup test rig (right) of C28-901 oxygen carrier

At CSIC, the attrition behaviour of the developed oxygen carriers was determined both at ambient temperature and during CLC operation. AJI index was measured using a three-hole air jet attrition tester, ATT-100M, configured according to ASTM-D-5757-95. Moreover during continuous CLC operation in the 500Wth unit the attrition rate of the oxygen carrier was measured recovering fines generated in both fuel and air reactors. After sieving, particles with sizes lower than 40 μm are considered as attrited particles. Different oxygen carriers showed different evolution of the attrition rate with time. Initially the attrition rate was high and then showed a decrease and stabilization. Using the stabilized attrition rates, the expected lifetime was 5000h for Cu/Al_2O_3 material and 2000h for C28 material.

In conclusion, the expected lifetime based on attrition testing and CLC operation looks promising for both materials and an assessment of 5000h lifetime has been considered in the benchmarking study. Nevertheless, it should be noticed that prediction of oxygen carrier lifetime must be considered with caution since critical failure mechanism may occur independently of the long-term trend expected by the short experiments performed on pilot plants. Testing with fuel injection shows more severe degradation of materials than cold or hot attrition testing, which indicates that redox cycling is probably predominant in the ageing mechanism of materials.

4 System design and optimization

4.1 Determination of reaction mechanisms

SINTEF has performed pulsed and continues micro- packed bed reactor experiments, in order to study the CLOU performance of different oxygen carrier material (OCM) samples developed and upscaled in SUCCESS projects. The CLOU capacity of the OCM materials vs temperature for oxygen release during 30 min is summarised in Figure 12. This CLOU capacity gives the expected amount of oxygen released that can give full combustion and it is released faster when fuel is present. The C28 material which is the stoichiometric composition has the highest CLOU capacity at all the temperatures of study. This is due to the higher amount of CMT perovskite structure in this sample. On the other hand, the C28–EuroSupport sample exhibits much lower CLOU capacity compared to its nominal value, due to its lack of complete intermixing and unreactive CaO left over in the structure. $Cu\text{-}Al_2O_3$ exhibit low CLOU capacity but in the range expected compared to the used Cu content, and accordingly it shows the lowest value compared to others.

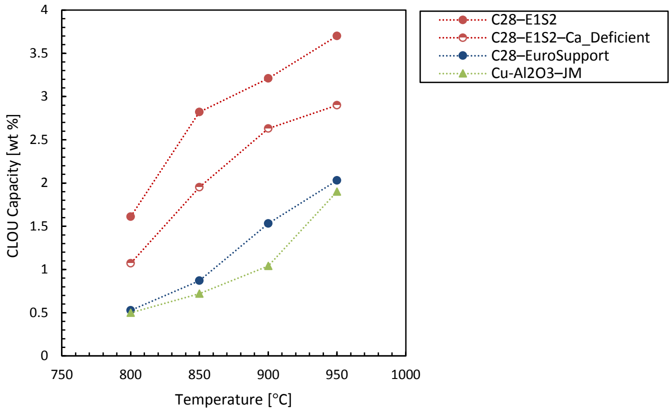
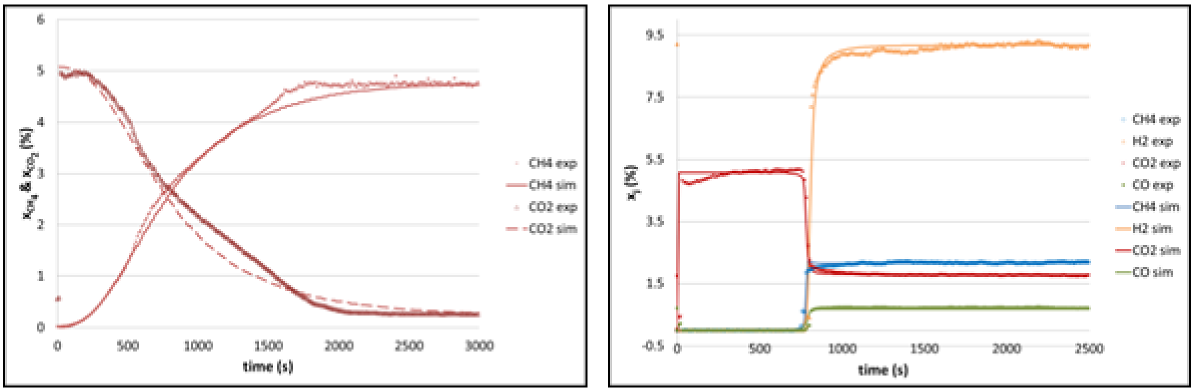


Figure 12: The CLOU capacity of the selected OCMs at different temperatures obtained from the micro-packed bed reactor experiments

A fixed bed reactor has been used by IFPEN for the determination of copper based and C28 perovskite materials reduction kinetic parameters. A coupled hydrodynamic and reaction model has been developed for the experimental data interpretation (information on reaction kinetics) and mechanism determination. The Shrinking Core model and the nucleation and nuclei growth model have been selected to represent the reduction reactions and typical kinetic laws were used to represent the catalytic reactions taking into account the equilibrium constants. The kinetic parameters of the various kinetic laws have been determined by optimization and accurate results have been obtained with the model for both materials (Figure 13). Based on these results, operation at low temperature (800°C) seems well adapted to copper based material. At this temperature, its reactivity is high enough for an application in a CLC process while the catalytic effect and deactivation will be minimized. On the contrary, reactor operation at high temperature will be recommended for perovskite (> 900°C) in order to maximize its reactivity.

At CSIC the reaction kinetics of the three developed oxygen carriers at industrial scale, was determined using a thermogravimetric analyzer system (TGA) operating in conditions of chemical reaction control. The Shrinking Core Model (SCM) in the grains with plate-like geometry was used to determine the kinetic parameters for impregnated Cu14-γAl-JM and Fe20αAl-SG, while spherical geometry was used for the spray dried C28 material. Chemical reaction control and diffusion through the product layer were considered in the model. Detailed kinetic parameters are presented in D6.1. The reaction models were able to predict the experimental results under different gas concentration, temperature or reaction type. The kinetic data obtained in this work will be useful to be included in mathematical models of CLC reactors with these materials as oxygen carrier. Moreover, oxygen uncoupling process can be relevant for C28 and this mechanism should be considered in fuel reactor modelling.



a) CH_4 variation over time for perovskite material at 925°C

(b) CH4, H2, CO2 and CO variation over time for copper based material at 800°C

Figure 13: Variation of experimental and simulated results for methane combustion (a) at 925°C for the perovskite material and (b) 800°C for the copper based material

4.2 Parametric modelling

A mathematical model for a Dual Circulating Fluidized Bed (DCFB) system was used to simulate the behavior of Cu14γAl-SCCa oxygen carrier developed in this project in the 120 kW_{th} CLC unit at Vienna University of Technology. The model consists on the coupling of individual fuel and air reactor models to simulate steady state of the CLC unit. Individual models consider both the fluid dynamic of the fluidized beds and the corresponding kinetics of oxygen carrier reactions, i.e. reduction in the fuel reactor and oxidation in the air reactor. Reaction kinetics was previously determined from TGA experiments (see D6.1). The model was validated using results obtained in the CLC unit at TUV, in which operating conditions such as solids circulation flow rate, temperature and pressure drop in the reactors were varied. Figure 14 shows the comparison of methane conversion both predicted by the model and experimentally achieved. The analysis of the results provided by the model highlighted that the main reason for the uncomplete fuel conversion was a deficit in the oxygen uptake by the oxygen carrier in the air reactor.

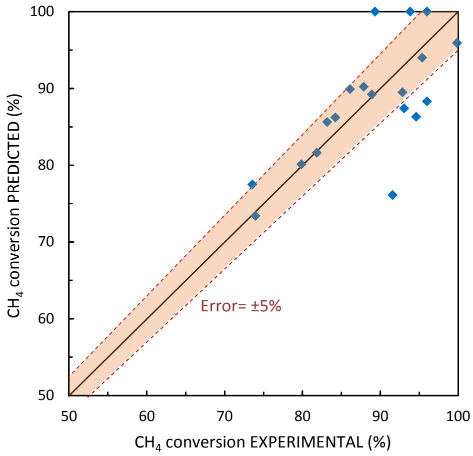


Figure 14: Model validation with experimental results using Cu based carrier in the 120 kW_{th} TUV unit

The theoretical model, after its validation, was used to identify the main factors affecting the methane conversion. The methane conversion in the fuel reactor was mainly performed in the dilute region.

Thus, poor conversion of methane is achieved in the bottom bed because a high fraction of gas is bypassed through the bubbles. As a consequence, it is not deserved to increase the solids inventory in the fuel reactor to improve the methane conversion because the additional solids will be mainly accumulated in the low-efficient dense bed.

On the contrary, it was identified that methane conversion can be highly improved by increasing the solids circulation flow rate and the solids inventory in the air reactor. In this way, it was possible to improve the fuel conversion by increasing the oxygen transferred both in air and fuel reactors. Optimized conditions to achieve complete fuel combustion includes an oxygen carrier to fuel ratio $\phi=4$ and solids inventories in fuel and air reactors of 130 and 120 kg/MW$_{th}$, respectively.

4.3 Next scale reactor design

A physical flow model or cold flow model (CFM) was used to give conclusions for suitable reactor concepts using the OC developed in SUCCESS, with the focus on an optimized fuel conversion. Therefor an existing CFM, originally designed for nickel-based OCs, with a scaling factor of 1:11 to a 10 MW$_{th}$ reactor design was adapted. This adaptations concerned the lower part of the fuel reactor (FR) which was replaced by several ones with a significantly wider diameter.

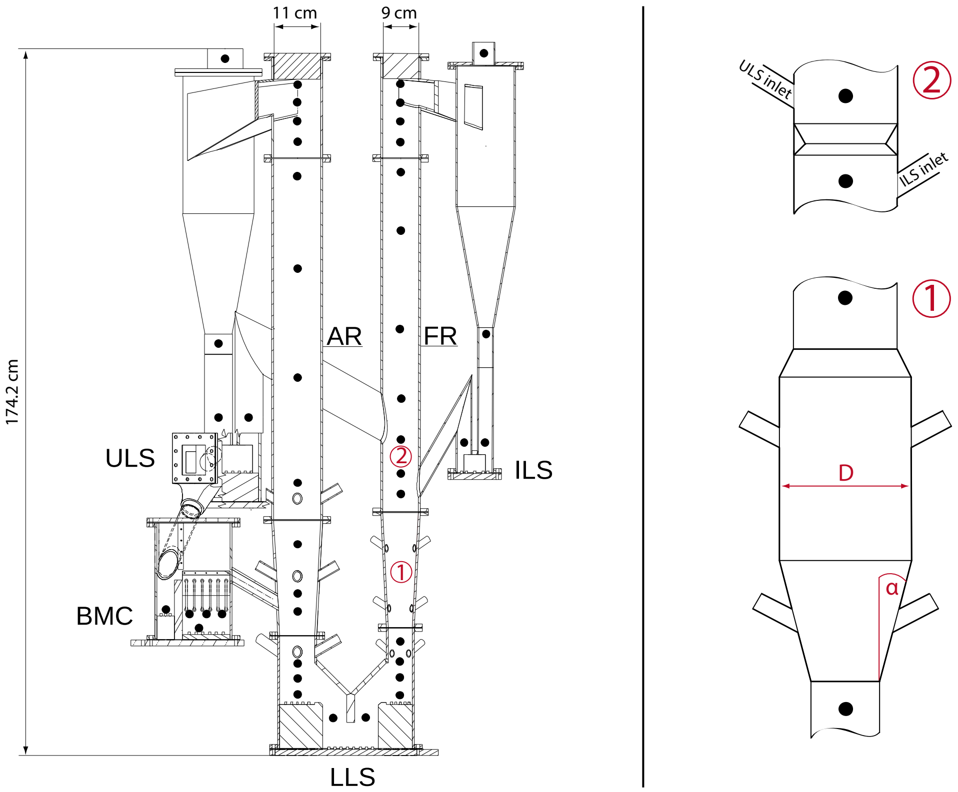


Figure 15: 10 MW$_{th}$ CFM and adapted sections

In total, two new geometries of the lower part of the FR (position labelled with 1 in Figure 15) were investigated. Those new lower parts (LP1, LP2) have the same maximum diameter (see D in Figure 15) but different spread angles (see α in Figure 15). The different spread angles were chosen to investigate their influence on the solid distribution in the FR and on the operation behavior of the fluidized bed system. Further, for one design an internal was placed between upper loop seal (ULS) inlet and internal loop seal (ILS) return leg (position labelled with 2 in Figure 15)

For all reactor designs, the pressure profiles of the AR show only slightly differences for different FR designs. Thus, it can be assumed that the AR is not affected by the changes of the FR design. In contrast, the pressure profile of the FR shows significant differences for the different reactor designs. These differences occur mainly at the lower part of the FR. Further, the almost identical solid distributions of the LP1 and LP2 designs it can be assumed that the spread angle of the lower part has only a minor influence. With integration of the internal, it was possible to significantly increase the solid fraction in the upper part of the FR.

The experiments showed that for two FR designs (LP1, and LP2 with internal) the operating range of the fluidized bed system is limited due to unstable operating conditions caused by fluid dynamic effects. This does not principally mean that these FR designs are inappropriate for the application in a DCFB. Rather, their suitability depends on the operating parameters of the CFM.

5 End-user evaluation

5.1 Health, safety and environmental impact analysis

The analysis of the health, safety and environmental impact investigates the potential human and ecological recommendations for safe use of oxygen carriers (OCs) in closed loop combustion (CLC).

First the potential **intrinsic hazardous properties** of the complex chemical structures in OCs were investigated and information for further human and ecological risk assessment for the use of these OCs in CLC was provided. Initially three different OC materials were used as model compounds: spray-dried Mn-ore based C28-OC, and Fe and Cu impregnated Al_2O_3. During the course of the project C28-OC and Cu impregnated Al_2O_3 (Cu-OC) were selected for upscaling.

To evaluate the potential **environmental health effects** the intrinsic hazardous ecotoxic properties of the fresh OC materials were assessed in: 1) acute and chronic aquatic toxicity tests on the leachate (water soluble fraction, WSF) in a standard ecotoxicity test battery of fish larvae, daphnia and algae 2) acute and chronic terrestrial ecotoxicity using earthworm and plant tests, and 3) potential toxic effects on bacteria of the waste water treatment plants. No effects were seen with the latter. Also effects of spent C28-OC were measured to evaluate possible changes in ecotoxicity during use.

The leachate fraction of fresh Cu-OC clearly had acute effects in both algae and daphnia tests and thus also has chronic impacts. In the fish larvae test also pronounced acute effects were observed with two out of three batches. Leachate fractions of Fe-OC had no adverse effects in the acute aquatic biotests. As this material was not selected for upscaling no further tests (chronic aquatic or terrestrial) were performed. The leachate fraction of fresh C28–OC had no acute or chronic hazardous effects in the aquatic biotests. Also leachate of spent C28-OC material was evaluated: no effects were seen when exposing daphnia and algae, but there were adverse effects on fish larvae. This needs further investigation on the changes responsible for the increased toxicity.

No adverse effects were seen in chronic ecotoxicity tests on worms and higher plants with C28-OC and Cu-OC. The direct risk for terrestrial organisms seems therefore limited. Direct release into soil is not expected from the OC use in CLC, but could accidentally occur e.g. during transport.

To evaluate potential **human health** risk it was explored whether inhalation could be a pathway. It was shown that indeed inhalable fractions are released from the three OC materials during operation. Extracts of the PM10 fractions (particulate material with diameter ≤ 10 µm) were tested *in vitro*. Beas2Be cells - representing the human bronchial inhalation pathway – were exposed to dilution series of the extracts. Cu-OC caused slight but significant toxicity to the cell line (20% cell death), but had no effect on the inflammation responses. When exposed to C28-OC however the cells demonstrated clear concentration dependent changes and significant effects.

Also the potency to induce malformations in human embryos was evaluated using the fish embryo model. C28-OC and Fe-OC had no adverse effects on the zebrafish embryo, but the Cu-OC showed pronounced teratogenic effects. Non-hatching of embryos was seen down to the lowest test concentration (6.25% WSF). Although the concentration of some metals were elevated, it is not clear which contaminant in the Cu-OC causes these effects. Further investigation is needed.

It can be concluded that both selected materials have intrinsic hazardous properties. Environmental exposure assessment is needed for the Cu-OC material to evaluate the potential risk for the aquatic environment. Human exposure assessment is needed to evaluate the potential risk for the inhalation pathway (C28 and Cu-OC) or other pathways (Cu-OC).

Secondly, the **exposure assessment** was performed with focus on the determination of exposure levels and recommendations of safety measures. The two OCs considered are Mn-OC and Cu-OC.

The main route for human exposure is through inhalation of dust that is generated during the manipulation of OCs (loading, unloading) and during cleaning and maintenance. By comparing the exposure concentration with a concentration that is considered to be safe, the risk is characterized.

The level of exposure via **inhalation of dust** was simulated by deliberate dust generation. Comparison with the occupational exposure limits (OELs) showed that whenever opportunities for dust generation occur, at least respiratory protection should be used. In case of exposure to the Mn-OC dust (Mn is an element with health concern) wearing an FFP2 mask reduces the risk to a low level; in combination with LEV or good mechanical ventilation the risk is reduced to a minimal level. This recommendation is confirmed in the Stoffenmanager exposure model which estimates the risk of exposure to OC dust to be of low priority when LEV or respiratory protection is used.

For the industrial scale, exposure was estimated using the Advanced Reach Tool (ART), considering 5 scenarios. From the identified scenarios, OC sampling creates the highest exposure concentration, however, still below the OEL. Nevertheless, it is recommended to wear an FFP2 respiration mask during sampling and fresh or spent OC unloading and other operations with dust formation. Nevertheless, some technical actions are recommended to minimize exposure, taking as a reference the best practices applied on fluid catalytic cracking (FCC) process: a) material delivery in pressurized silo trucks and through a pipe to the storage hopper (equipped with sintered metal filters on the vent line to avoid emissions to the pipe); b) equipment loading with special powder handling equipment hooked upon the hopper and process unit; c) monitoring OC loading for accurate dosing; d) recovery of fines in fractionated bottoms or slurry products; e) gas seals to prevent gas and fines leakage, and e) use of sintered metal filters to retain dust during spent OC removal.

Dermal exposure during OC delivery or equipment loading was also considered; the latter is relevant for pilot scale and research environment as in these facilities loading is often a manual operation; also dermal exposure during OC sieving (to separate the fines) is relevant. Dermal exposure was estimated with the occupational exposure model RiskOfDerm. Two scenarios were considered: first loading and spent OC sieving. The calculated exposure was compared with the safe concentration (Derived No Effect level or DNEL) to estimate the risk. To control the risk it is necessary to wear disposable gloves and long sleeves. Dermal exposure to OCs is mainly through deposition of dust on the skin and less by direct dermal contact. Hence in addition to gloves, it is recommended to use LEV or good mechanical ventilation to decrease the risk to a minimal level.

The main route for release to the environment is the **emission through exhaust air**. The exposure evaluation for the environment was based on the experience from the Fluid catalytic cracking (FCC) process in Oil & Gas refining industry. The Best Available Techniques (BAT) Reference Document for the Refining of Mineral Oil and Gas includes emission limits for pollutants and the following techniques to mitigate pollutant emissions: a) additional external set of cyclones in the third/fourth stage separator; b) electrostatic precipitator with electric field to charge and capture solid particles; c) other filters (ceramic stainless steel/sintered metals) to capture particulate emissions, and d) scrubbing, mainly to reduce SOx emission, but also useful for particle emission reduction.

Release to the aquatic environment is low as CLC is a water-free process. Leaching tests showed that the level of Cu leached from the Cu-OC is high enough to harm aquatic life. Hence the Cu-OC should be prevented from entering surface water or soil (e.g. transport accidents).

The **solid waste** produced in the CLC process is mainly spent OC, filters, OC fines and tank sludge slurry. Reuse or recycling options exist and should be considered.

5.2 Life cycle analysis

The LCA investigates the environmental impacts from cradle to grave of steam production in full-size CLC and amine-based post-combustion capture steam plants. Both large scale (700 MW) plants that produce electricity and medium scale (100 MW) plants that produce steam are assessed. The functional unit is defined as “the production of one kWh of power, produced by a power plant with a reference service lifetime of 25 years”. The power can be electric or thermal. In the baseline, spray-dried calcium-manganese-based (C28) oxygen carriers are assumed to be used for the CLC steam plants. The ILCD impact assessment method is selected to calculate the environmental impacts.

When comparing the large (700 MW) atmospheric CLC steam plant with the high grade steam reference plant, electricity production in the CLC plant results better for terrestrial and marine eutrophication (Figure 16). However, for most other impact categories, the reference plant has a lower environmental impact. For some impact categories, such as climate change, the results are not conclusive. The result is largely determined by the consumption of natural gas. Since the atmospheric CLC steam plant has a lower efficiency than the high grade steam reference plant (36,8% versus 51,2%, respectively), its impact is higher for many categories considered.

For the medium scale (100 MW) plants, steam production in the atmospheric CLC steam plant results better than steam production in the low grade steam reference plant for photochemical ozone formation, terrestrial and marine eutrophication. For all other impact categories, the uncertainty is too high to conclude that one plant is better than the other. Although the efficiency of the CLC plant is higher than the efficiency of the low grade steam reference plant (83,7% and 79,0%, respectively), the difference is not large enough to lead to a significant difference in environmental impact. The environmental impact categories for which the CLC plant has a better result are those for which the direct process emissions have a large influence on the results for the reference plant. It can thus be concluded that the results of the comparison of CLC steam plants and amine-based post-combustion capture steam plants depend largely on the application and type of plant.

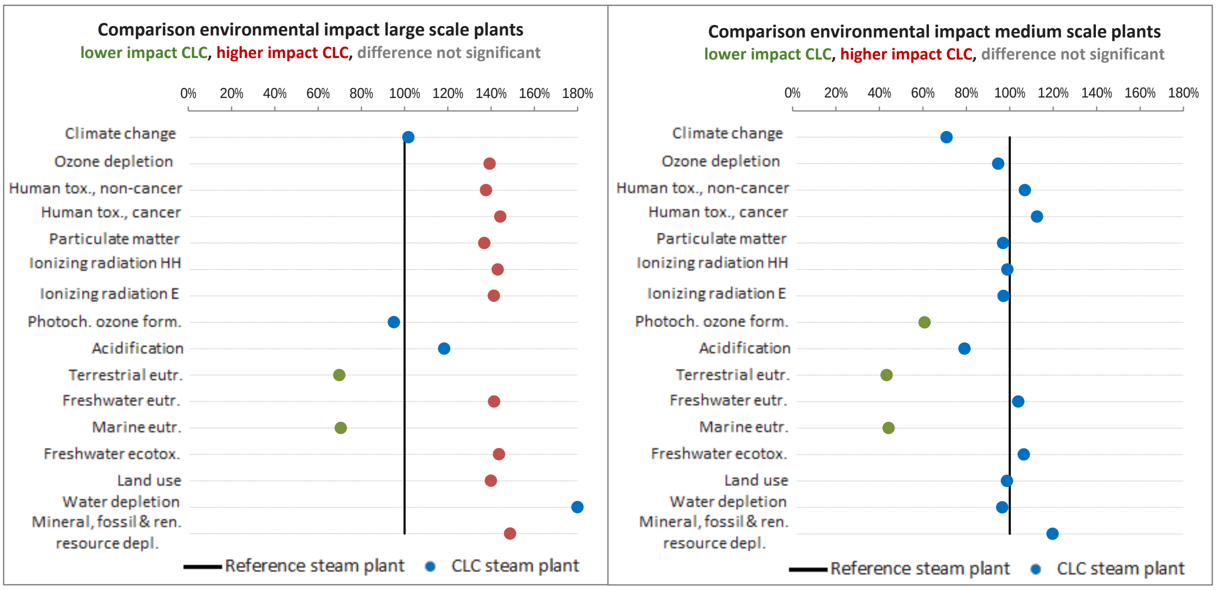


Figure 16: Comparison of one kWh of electricity produced in the large scale CLC and high steam reference plant (left) and one kWh of steam produced in the medium scale CLC and low steam reference plant (right)

5.3 Reuse and recycling options

The **significant quantities of raw materials** that will be used in **oxygen carriers** (OCs) mean that an early consideration of recovery routes is required. The recycling of OCs is not yet current practice. Therefore, **possible recycling routes** were proposed based on the physicochemical properties of the spent OCs and recycling routes of similar materials. The recycling aim of the materials in the SUCCESS project was to follow the waste hierarchy principles, whereby firstly it is tried to re-use the spent OC materials as novel OCs. According to the following step of the waste hierarchy, recycling of the materials in novel products was studied, followed by an evaluation of the material in the case it should be landfilled.

The **deactivation** of the **Cu-based impregnated OCs** in the CLC process is caused by **sintering effects** and **attrition loss** of the CuO. While, **C28 spray dried perovskite type material** suffers from deactivation with time due to the **formation of $CaSO_4$**. There is no indication that fouling-based deactivation mechanisms occur, thus in-situ regeneration processes are not feasible.

An initial literature study listed interesting recycling routes for spent impregnated and spray dried OC materials and based on techno-economic feasibility evaluations a selection of these possible routes were further developed on laboratory scale.

Reuse:

At the lab-scale, the feasibility of **metal recovery from spent impregnated OCs by acid (HNO_3) leaching** was investigated. The recovered copper was used in the production of new OCs by re-impregnation of spent OCs or by **impregnation** of new raw material (support). This recovery route proved to be successful. The tests were performed on spent OCs obtained from a 10 kW_{th} CLC prototype operated at 900 °C. The proposed recovery and recycle processes can largely decrease the amount of natural resources (Cu and Al_2O_3) employed in a CLC power plant as well as the waste generated in the process. Moreover a preliminary study into **reprocessing of spent spray dried C28 OCs into new OCs** showed promising results. Preliminary tests showed the possibility to reprocess spent spray dried (C28) OCs. SEM imaging showed a **similar structure** for the reprocessed OCs as for the fresh OCs. The produced OCs from this process (milling + re-spray drying) show **less activity** then fresh OCs, this is probably caused by a **higher density and lower pore volume** in the reprocessed OCs. An optimisation of the sintering process could strongly improve the reactivity.

Recycle:

The solid (alumina) residue obtained after the copper recovery process was used as a raw material for producing high-strength ceramic spheres. Such spheres can be used e.g. as high performance fillers, to allow liquid flow in intercavities, withstanding high pressures. The high market price of such fillers (500-1800 EUR/t) can cover the costs arising from the treatment steps in the suggested recycling process. In the optimal conditions, spheres with a **compressive strength of >100 MPa** can be produced.

Disposal:

If landfilling of certain fractions would be necessary, results of batch leaching tests indicated that the OCs could be suitable to be landfilled on **landfills for inert waste** according to Council Decision 2003/33/EC of the European Union.

5.4 Overall techno-economic evaluation

A first step has been to agree on a common assessment methodology using the EBTF (European Benchmarking Task Force) technical and economic framework as a starting point. Key criteria for benchmarking has been identified and defined.

In a second step, the production costs for two oxygen carriers (OC) materials have been detailed and extrapolated to multi-tonne production units from the know-how of the suppliers:

- for the spray-drying route (C28 composition – $CaMn_{1-x-y}Mg_xTi_yO_{3-\delta}$ – produced from relatively cheap and abundant Mn- and Ca-based raw material) on large scale infrastructure at Euro Support Advanced Materials,
- for the impregnation route (Cu-impregnated – 15 wt.% CuO – using a commercial alumina substrate) using large scale infrastructure at Johnson Matthey.

For both manufacturing routes and oxygen carrier types, an indicative cost price calculation has been done for order of magnitude production levels of 100 (demonstration plant facility) and 1 000 (commercial scale manufacturing plant capacity) metric tonnes OC per year. The generated OC indicative market prices (based on 1 000 t/yr OC capacity) from a techno-economic manufacturing study have been incorporated within the overall CLC techno-economic evaluation.

In a third step, a reference steam generation unit for natural gas has been produced. The need was to clearly define against which best available technologies the CLC technologies are benchmarked. A techno-economic study on generation of two different steam grades has been done (cases 1.1 to 2.2 in Table 7):

- The first one for power generation in a steam cycle according to practices in electrical industry (EDF) using a single compressor/gas turbine (GT) group coupled to a three-pressure-levels heat recovery steam generator (HRSG), a post-combustion CO_2 capture unit based on 30 wt% monoethanolamine (MEA) aqueous solution and a CO_2 compression unit (110 bar),
- The second one suitable for high-temperature industrial usages, e.g. steam-assisted gravity drainage according to practices in oil and gas industry (Shell and Total), that includes a single GT coupled to a two-pressure-levels HRSG, a post-combustion CO_2 capture unit based on 30 wt% MEA aqueous solution and a CO_2 compression unit (110 bar).

In a fourth step, the gas-CLC-based steam plant has been designed and scaled-up (Figure 17) to account for all necessary equipment for an industrial application:

- **Atmospheric CLC**

Two different steam grades have been considered, i.e. high grade steam (540°C – 255 bar) – suitable for power generation (electrical industry) in a steam cycle, and low grade steam (510°C – 100 bar) for industrial usage (oil and gas industry). The gas-CLC plants for the atmospheric case have been investigated by Bertsch and BOKU with C28 OC and for 100% CO_2 capture efficiency on the same basis and on a common thermal input (694 MW_{LHV}) comparable with the reference cases. 3D CFD models using a mesh of around 1 million cells have been used by INPT to better understand the hydrodynamic of reactive flows and gas-solids circulation at full scale (Figure 17b and c).
The Air Reactor (AR) is divided into two reactors and each reactor has two cyclones for separating flue gas and OC (Figure 17a). The reactors and the cyclones are quite similar for the two cases. The main differences are in the heat exchangers after the cyclones. The overall CLC and steam plant (reactors, cyclones and channels for heating surfaces) will be executed in a hanging version in an appropriate steel structure. For design of the heating surfaces, only plane tube surfaces have been used because there is not sufficient experience to use finned tubes for CLC flue gases.

- **Pressurized CLC**

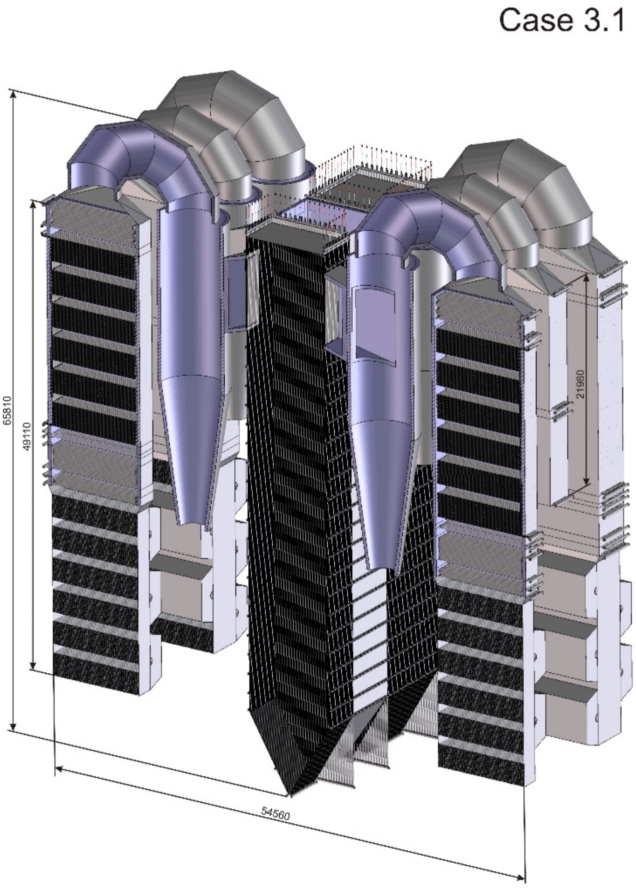
Based on the same approach as atmospheric CLC, the gas-CLC plants for the pressurized case (i.e. 5 bar at inlet of reactors) have been investigated on a common thermal input. Two plant designs have been considered with C28 OC and for 100% CO_2 capture efficiency.
The performances of pressurized gas-CLC for power generation have been studied in details by BOKU. It was found that the CLC process comes along with technological limitations that need to be resolved to make CLC technology competitive: (i) turbine inlet temperature is limited by the oxygen carrier material (ii) pressure drop of CLC AR path increases the required air compression (iii) the requirement for low pressure steam for gas-sealing between AR and FR reduces the efficiency of the steam cycle.

In a final step, the gas-CLC plants have been calculated (CAPEX, OPEX and Levelized Cost of Energy – LCOE) on a common technical-economic basis (cases 3.1 to 4.2 in Table 7) including CO_2 compression unit. The equipment costs for gas-CLC steam plants have been based on costing methods and database from technology developer (Bertsch) and end user (EDF). For steam generation, both atmospheric and pressurized gas-CLC are more competitive than the conventional best available technologies, i.e. GT-HRSG with CO_2 capture in amine based solvent (cases 3.2 and 4.2 vs 2.2). On the contrary, for power generation, both atmospheric and pressurized gas-CLC are less competitive than the conventional best available technology, i.e. combined cycle with CO_2 capture in amine based solvent (cases 3.1 and 4.1 vs 1.2). Furthermore, for combined generation of power and heat (CHP) which is important for higher total efficiency, there is no significant advantage for pressurized CLC compared to atmospheric CLC. Finally, the LCOE sensitivity on selected parameters follows the following order: natual gas price > full load operation > process equipment cost > auxiliaries consumption > oxygen carrier lifetime.

The following conclusions can be drawn:

- Gas-CLC is an intermediate step towards solid fuel CLC. Thus, demonstration of gas-CLC over long time periods is crucial to gain operational experience.
- Continuous and transient operation with a mechanically, chemically and thermally-stable and competitive oxygen carrier like spray dried C28 and impregnated Cu-based material has to be pursued.
- Steam generation for industrial purpose may be a promising potential demonstration scenario at larger scale, e.g. in the range of 10 MW_{LHV}.

Besides demonstration, gas-CLC is of interest in steam generation and in certain application such as oil and gas industry.



(b) Instantaneous O_2 molar fraction – INPT **(c) Instantaneous CO2 molar fraction – INPT**

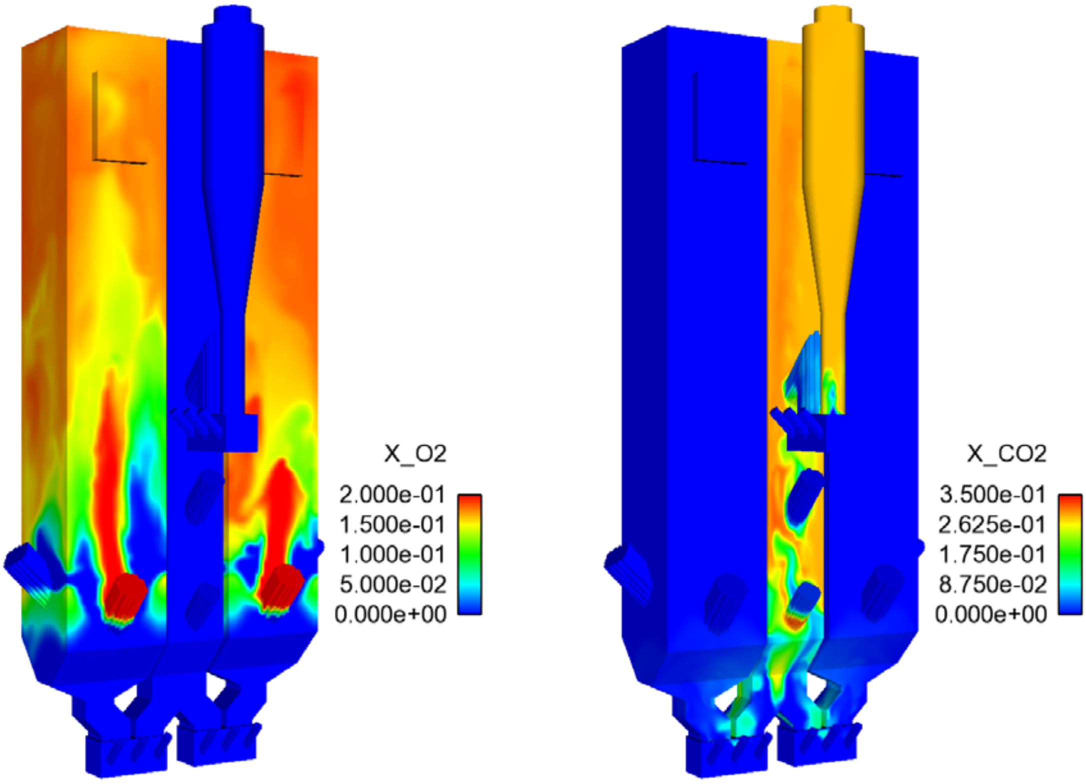


Figure 17: 3D view of industrial gas-CLC unit (694 MWLHV) at atmospheric pressure

Table 7: Results of techno-economic evaluation

Case 1.1: CC (GT + HRSG + steam turbines) – Case 1.2: CC (GT + HRSG + steam turbines) + CO_2 capture and compression unit
Case 2.1: cogeneration unit (GT + HRSG) – Case 2.2: cogeneration unit (GT + HRSG) + CO_2 capture and compression unit
Case 3.1: atmospheric CLC (AR/FR reactors + HRSG + steam turbines) + CO_2 compression unit and Case 3.2: atmospheric CLC (AR/FR reactors + HRSG) + CO_2 compression unit
Case 4.1: pressurized CLC (AR/FR reactors + GT + HRSG + steam turbines) + CO_2 compression unit and Case 4.2: pressurized CLC (AR/FR reactors + GT + HRSG) + CO_2 compression unit
* thermal + electrical

Parameter	Unit	Case 1.1	Case 1.2	Case 2.1	Case 2.2	Case 3.1	Case 3.2	Case 4.1	Case 4.2
Application		Power	Power	CHP	CHP	Power	Steam	Power	CHP
Natural gas thermal power	MW_{LHV}	749	749	694	694	694	694	694	694
Gross electric power	MW_e	447	407	262	262	285	-	281	99
Gross electric efficiency	$\%_{LHV}$	59.6	54.3	37.8	37.8	41.1	-	40.5	14.3
Net electric power	MW_e	443	382	262	243	255	-	272	90
Net electric efficiency	$\%_{LHV}$	59.2	51.2	37.8	25.0	36.8	-	39.2	13.0
Steam thermal power	MW_{th}	-	-	291	182	-	635	-	493
Steam flow rate	t/h	410	410	360	225	832	725	420	502
Steam temperature/pressure	°C/bar	580/167	580/167	510/117	510/117	540/255	510/100	580/167	510/100
Total efficiency*	$\%_{LHV}$	59.2	51.2	79.7	61.2	36.8	91.5	39.2	84.0
Specific CO_2 emissions	gCO_2/kWh	349	48	259	34	0	0	0	0
Total capital requirement	$M€_{2013}/MW_{LHV}$	0.61	0.87	0.49	0.76	0.94	0.73	0.99	0.57
LCOE	$€_{2013}/MWh$	64 electricity	83 electricity	31 steam	75 steam	119 electricity	46 steam	113 electricity	43 steam

6 Project conclusions

The main goal of the SUCCESS project was providing the last missing R&D input for demonstration of Chemical Looping Combustion (CLC) of gaseous fuels at next scale in the range of 10 MW_{th}. This included research on scale-up of the most important aspects of Chemical Looping Combustion: Oxygen carrier material production and reactor system design.

Operation of next scale CLC plants requires large amounts of oxygen carrier material in the range of several tons. The scale-up of oxygen carrier material production from 100 kg to several tons using industrially available raw materials was one big focus of the SUCCESS project. Production of two different materials (calcium manganese based and copper based), using two different production routes (spray-drying and impregnation) were scaled up to tonne scale production. This included not only scale-up of material production methods itself but also identification of suitable raw materials needed for production which are available in required quantities. In total, more than 40 samples of the two materials were produced during scale-up and tested in lab scale facilities according to their reactivity and life time. The best performing sample of each material has been selected for scale-up to tonne scale. In total, almost 3.6 t of material (600 kg Cu based material and 3 t Ca-Mn based material) were produced using industrially relevant material and production facilities.

The overall performance of produced materials has been extensively evaluated in pilot units up to 150 kW. Investigating concerned not only influence of important process parameters like temperature, solid material inventory or material circulation but also investigations on the influence of sulphur contaminants in the fuel and on long time particle performance. The oxygen carriers worked well in all pilot units achieving CO_2-yields above 90%. Comparison of the large scale material with the original benchmark material produced at small scale using pure chemicals showed, that the large scale material shows similar (Ca-Mn material) or even better performance (Cu based material). In addition to pilot testing up to 150 kW, CLC of gaseous fuels has also been demonstrated in the size of 1 MW_{th} using the Ca-Mn based OC.

The end-user evaluation of the CLC technology for industrial steam generation investigated not only techno-economic performance but also issues related to health and safety, potential recycling routes as well as the whole life cycle impact. Recommendations for handling oxygen carrier material were drawn based on a three tier health, safety and environmental impact analysis. The analysis included human and environmental effects. The available reuse and recycling options are leaching of metals (Cu based material), reprocessing of spent material to new oxygen carrier material (Ca-Mn based material) as well as production of high value ceramics out of spent Cu based material. The techno economic evaluation showed that CLC for gaseous fuels is competitive compared to best available technologies for industrial steam generation.

Based on the work performed, the following conclusions can be drawn:

- All project objectives and technical goals have been achieved. The technology can be seen as ready for next scale demonstration having large scale oxygen carrier material production technologies at hand. Further, detailed design recommendation for such a demonstration unit were made.
- Gaseous fuel is an intermediate step towards solid fuel CLC. Thus, demonstration and long term operation of gas-CLC is crucial to gain operational experience with CLC processes.
- Industrial steam generation in the range of 10 MW_{th} may be a promising scenario for next scale demonstration of gas-CLC.

Besides demonstration scenarios, gas-CLC is of interest for industrial steam generation and certain applications in oil and gas industry.

7 Project impact

7.1 Expected project impact

The main impact of SUCCESS can be seen in the contribution to reduction of the energy penalty for CO_2 capture technologies. This impact is achieved by development of the CLC technology for gaseous fuels beyond its state-of-the-art and make it ready for demonstration at next scale in the range of 10 MW_{th}. The achievements and the resulting impact happen in the areas of technology development and end-user evaluation. Technology development included scale-up of oxygen carrier material production and overall system design optimization. Availability of large quantities of OC material is ensured through scale-up of production technologies for two different oxygen carrier materials including confirmation of suitable raw materials. Further, system design has been optimized and detailed design recommendations were made for next scale demonstration. However, there are currently, different to the start of the project, no specific plans for such a demonstration project.

Further impact can be expected by development of tools and advancement of general knowledge during the project, which can be used in other areas than CLC. This includes, e.g. production of spheres via spray-drying, in the case of material production. Large impact can also be expected in the field of fluidized bed technology in both, engineering and operation. Modelling tools for fluidized bed engineering have been created and adapted and a lot of experience has been gained during operation of the CLC pilot units. Fluidized bed systems used for this work can also be used in other application and gained experience and know-how can be transferred.

The SUCCESS project concerned development of an energy production technology. Thus, socio-economic impact and wider societal implications have to be seen in the context of the impact of the technology on the fight against global warming in general and development of a greenhouse gas mitigation technology in particular. Thus, the socio-economic impact is mainly connected to the socio-economic impact of climate change and climate change mitigation respectively. A broader socio-economic impact beyond the technology itself is not seen.

7.2 Dissemination and exploitation of results

Dissemination of project results was mainly organized through publication in scientific journals and presentations at scientific conferences:

- The original plan at the start of the project was an output of 15 peer-reviewed journal publications. At the end of the project, 12 articles were either published or accepted for publication in scientific peer-reviewed journals. Additionally, 5 peer-reviewed journal publication are currently under preparation or submitted for publication. It is expected that the total output will be beyond 20 journal publications since a lot of results were obtained at the very end of the project and will be published in the future.
- The plan at the start of the project included 15 presentations at international conferences. At the end of the project, 40 presentations have been held at international conferences. Out of these 40, 6 were held at the first public workshop. One of the 40 presentation was a project overview presentation at the 13th Conference on Greenhouse Gas Technologies. Further, 10 presentations will be held during the second public workshop in the scope of the Trondheim CCS conference (June 2017).

A project website (http://www.clc-success.eu) has been set-up to present the content of the project and project partners. Results, which can be disclosed to a public audience, were made available at the project website in the form public project Deliverables. A project information flyer was created and distributed to all partners in sufficient quantities. The flyer was also distributed at the public project workshops.

8 Project team

Vienna University of Technology

Vienna University of Technology, Institute of Chemical Engineering (Vienna). The Research Platform on "Future Energy Technology" (about 50 scientific employees) unites three Research Groups on Zero Emission Technologies, Gasification and Gas Cleaning as well as on Synthetic Biofuels. During the last decade, several energy technology processes have been developed - starting from the idea via laboratory and pilot plants up to full scale demonstration units. The tools applied are of experimental and modelling nature. Vienna's special experience is with hot laboratory/pilot units in a scale of 100 kW and also with cold flow models of fluidized bed systems. During the recent years the research group has been involved in several European research projects in the field of fluidized bed systems for advanced fuel conversion technologies: GRACE (FP6), CCCC (RFCS), CLC GAS POWER (FP6), CACHET (FP6), RENEW (FP6), FLEXGAS (RFCS), BioSNG (FP6), AER GAS II (FP6), BiGPower (FP6), UNIQUE (FP7), INNOCUOUS (FP7), FECUNDUS (RFCS), ACCLAIM (RFCS).

The available hardware resources to be contributed comprise:

- 120 kW$_{th}$ dual circulating fluidized bed pilot plant for chemical looping combustion/reforming of gaseous fuels
- Fluid dynamic cold flow model of a dual circulating fluidized bed 10 MW$_{th}$ gaseous CLC demonstration plant
- Analytical equipment for measurement of concentration of gaseous species and sulphur components

Contact:
Stefan Penthor
P: +43-1-58801-166367
stefan.penthor@tuwien.ac.at
www.tuwien.ac.at/en/

Chalmers University of Technology

Chalmers University of Technology, Dept. of Energy Conversion (Chalmers) is world-leading in CLC, having worked with development of oxygen-carrier material for the process since 1998 and being first to successfully demonstrate this process in sustained operation in a 10 kW prototype unit for gaseous fuel. Further, Chalmers is also first to demonstrate operation with solid and liquid fuels. Chalmers has four CLC units for gaseous/solid/liquid (G/S/L) fuels: 300 W (G/L), 10 kW (G/L), 10 kW (S) and 100 kW (S), that have totally been in operation with fuel for 2600 h, using 27 different oxygen carrier materials. This involves the first successful demonstration of chemical-looping combustion with oxygen carriers based on nickel, iron and manganese oxides, as well as natural minerals and combined oxides (e.g. CaMnO3), using natural gas, syngas, bituminous coal and pet coke as fuels. Further, Chalmers has investigated more than 300 different oxygen carrier materials in laboratory. Chalmers has >180 publications on CLC, of which >100 are reviewed and 10 are PhD theses. Major experimental and modelling work at the department has been related to combustion, with focus on fluidized beds utilizing a unique 12 MW circulating fluidized bed (CFB) research boiler located at Chalmers. Chalmers has been initiator of chemical-looping research in the EU-projects GRACE, CLC Gas Power, Cachet, Innocuous, NoCO2, the ECSC-projects CCCC and Acclaim, and together with Alstom, the RFCS-project ECLAIR and solid fuel CLC work within EU-project ENCAP.

The available hardware resources to be contributed comprise:

- 0.3 and 10 kW_{th} fully equipped chemical looping combustors for gas
- Jet cup attrition rig.
- Four fully equipped rigs for laboratory testing of oxygen carriers, as well full access to all available laboratory analysis equipment, XRD, SEM ....

Contact:
Anders Lyngfelt
P: +46-31-772 1427
Anders.Lyngfelt@chalmers.se
www.chalmers.se

Consejo Superior de Investigaciones Científicas

Consejo Superior de Investigaciones Científicas (CSIC), Instituto de Carboquímica (ICB). (www.icb.csic.es). The “Combustion and Gasification” research group at ICB is composed by about 20 scientific researchers including staff, students and technicians. The group has a long expertise in gassolid reactions, reaction kinetics, modelling applied to fluidized bed combustion and gasification processes as well as on Chemical Looping Combustion (CLC) process. Since 2000 the Department is involved in the particle development, selection and testing of oxygen carriers for CLC, kinetic studies and reactor optimization for this process. They have participate in several EU projects related with CLC: GRACE(FP6), CCCC (RFCS), CLC GASPOWER(FP6), CACHET (FP6), UNIQUE(FP7), ACCLAIM(RFCS), and other Spanish research projects. CSIC has worked in the CLC Process to transfer the concept from laboratory to prototype scale carrying out a demonstration of the technology. Main achievements have been: 1) Development of oxygen carrier materials based on Cu and Ni suitable for the process, 2) Demonstration of the process concept at 10 kW_{th} continuous CLC for 200 h working with Cubased oxygen carriers and 1000 h in a 500 W_{th} facility with Cu, Fe and Ni based oxygen carriers, 4) Testing of the effect of sulphur and light hydrocarbons in CLC, 5) Modelling of CLC for several gaseous and solid fuels (CH4, syngas, coal, etc.), 6) First demonstration of the CLOU technology for coal combustion in a 1 kW_{th} continuous CLC plant.

Resources to be used in the project:

- Thermogravimetric analyzers to determine reduction/oxidation reactivity of carriers.
- Batch fluidized bed reactor setup with on line gas analysis.
- Continuous 500 W_{th} unit for CLC with solid circulation control.
- Facilities for carrier preparation by pelletizing and impregnation.
- Facilities for oxygen carrier characterization i.e. physisorption, chemisorption, Hg porosimetry, TPR, XRD and SEM-EDX.
- Facilities for leaching tests.

Contact:
Juan Adánez
P: +34-976-733977
jadanez@icb.csic.es
www.csic.es

IFP Energies Nouvelles

IFP Energies nouvelles is a public-sector research, innovation and training center active in the fields of energy, transport and the environment. Its mission is to provide public players and industry with efficient, economical, clean and sustainable technologies to take up the three major challenges facing society in the 21st century: climate change and environmental impacts, energy diversification and water resource management. It boasts world-class expertise. IFP Energies nouvelles sets out 5 complementary, inextricably-linked strategic priorities that are central to its public-interest mission. Renewable energies: producing fuels, chemical intermediates and energy from renewable sources. Eco-friendly production: producing energy while mitigating the environmental footprint. Innovative transport: developing fuel-efficient, environmentally-friendly transport. Eco-efficient processes: producing environmentally-friendly fuels and chemical intermediates from fossil resources. Sustainable resources: providing environmentally-friendly technologies and pushing back the current boundaries of oil and gas reserves As an integral part of IFP Energies nouvelles, its graduate engineering school prepares future generations to take up these challenges.

The available hardware resources to be contributed comprise:

1. Continuous 10 kW_{th} pilot plant for chemical looping combustion of gaseous fuels.

2. Fixed-bed reactor pilot plant for the determination of kinetics.
3. Attrition testing unit and other characterization techniques for CLC oxygen carriers.

Contact:
Stephane Bertholin
P: +33 437702309
stephane.bertholin@ifpen.fr
www.ifpenergiesnouvelles.com

Institut National Polytechnique de Toulouse - Institut de Mécanique des Fluides de Toulouse

L'Institut de Mécanique des Fluides de Toulouse (IMFT) is a French laboratory (UMR 5502 CNRS/INPTUPS) worldwide known for its expertise in fluid mechanics and more specifically in multiphase flows. The research group Particles, Spray and Combustion (PSC) has numerical and experimental activities focused on multispecies mixing, particulate flows, and combustion. People in charge of particulate flows are worldwide recognized for their competences in numerical simulation and modelling of particle-laden turbulent flows and have a large number of publications in highly ranked scientific journals. More specifically the PSC group skills are:

- DNS at the particle scale;
- DNS/LES of non-isothermal turbulent flows coupled with Lagrangian tracking of particles accounting for particle-particle collisions;
- Modelling of the particle-turbulence interactions in the frame of RANS or LES approaches;
- Development of statistical joint fluid-particle approach and moment method for particulate dense, and dilute, flows;
- Numerical simulations at industrial scale of reactive particulate dense and dilute flows by using the code NEPTUNE_CFD.

The PSC group has several collaborations with industrial partners: current projects are with EDF R&D, TOTAL, INEOS, ALSTOM, AREVA, PSA, and INRS.

Contact:
Pascal Fede
P: +33 534322899
pascal.fede@imft.fr
www.imft.fr

Stiftelsen SINTEF

SINTEF, Institute of Materials and Chemistry, Department of Energy Conversion and Materials. SINTEF has a main focus on materials formulation, shaping, development and testing related to technologies for CCS and power production. The group has therefore been involved in several EU projects like i.e. GRACE, ENCAP, DECARBIT, ICAP, CARENA, CHACET I and II, REAL-SOFC, EFFIPRO, RAMSES, ÉCLAIR, ACCLAIM, DEMOCLOCK, HETMOC. Several of these projects are focused on development of oxygen carriers for i.e. CLC, CLR, CAR (CLOU) technologies. The materials have been tested in small scale reactors, TG and TPX for material characterisation. The group has a fixed bed CLC unit and a double fluidised bed CLC rig designed for attrition testing under real operating conditions. It includes double changeable filter systems for both fuel and air reactor.

The available hardware resources to be contributed comprise:

- 3 kW_{th} dual circulating fluidized bed test rig for chemical looping combustion/reforming of gaseous fuels, but with an aim of testing material attrition strength.
- Analytical equipment for measurement of concentration of gaseous species and sulphur components
- TPX, HPTG, SEM, XRD, BET equipment for material characterization.

Contact:
Yngve Larring
P: +47 22067350
yngve.larring@sintef.no
www.sintef.no

SINTEF Energi AS

SINTEF ER (a legal entity affiliated to SINTEF) is a contract research institute focusing on power production and conversion and has built up a sizeable group of people working within CCS technologies. In total SINTEF ER employs about 200 people. SINTEF ER, in collaboration with NTNU, has about 30 years of experience in numerical simulation of combustion processes and experimental capabilities within combustion and thermal energy, including a laser diagnostic combustion lab. SINTEF ER is co-ordinating a Norwegian R&D project on CLC which is now a part of the BIGCCS International CCS Research centre, one of Europe’s largest R&D projects on CO_2 capture and underground storage. Experience has also been built on oxy-fuel and hydrogen combustion in CO_2 capture processes through the Norwegian research program BIGCO2, and EU projects like ENCAP, DECARBit and DYNAMIS. Within combustion and particle flows SINTEF ER has developed a high level of expertise on the use of Direct Numerical Simulations, in close collaboration with Sandia National Laboratories, and more recently with Stanford. SINTEF ER also has expertise within bio energy, gas technology and cryogenic processes, including modelling and simulation capabilities for both components and systems. In the national CLC project, a 150 kW CLC pilot plant and a 1:1 scale cold flow model has been designed. The cold flow model has been used to verify and improve the design, and the 150 kW pilot plant is now in the final stage of construction with start of commissioning in November 2012. The design is novel in the sense that it enables flexibility in the flows between the reactors, and it has possibilities for makeup feeding and extraction of particles during operation. Rig velocities and fluidization modes are representative for industrial systems and will provide a valuable step for particle and system testing.

The available hardware resources to be contributed comprise:

- 150 kW_{th} CLC pilot plant with flexible particle exchange system, online particle feeding and extraction for chemical looping combustion/reforming of gaseous fuels. Well equipped with instruments and control valves and a LabView control system.
- Online gas analysis of exhaust O2, CH4, CO and CO2, and FTIR / GC measurement of other species.
- Particle analysis at SINTEF particle lab.

Contact:
Øyvind Langørgen
P: +47 73591612
oyvind.langorgen@sintef.no
www.sintef.no/SINTEF-Energi-AS/

Darmstadt University of Technology

The Institute of Energy Systems and Technology (EST) at TUD has large experience in the field of power plant technology, particularly in solid fuel combustion processes and related CO2 capture techniques. Its key expertise lies in modeling and simulation of multiphase flows (e.g. coal-fired boilers, mills, cyclones), modeling of heat exchanger systems (e.g. HRSG), steady-state and dynamic simulation of power plant processes, and experimental investigations of solid fuel combustion processes in laboratory and pilot scale test facilities. A 1 MW_{th} pilot plant consisting of two interconnected CFB reactors has been erected and operated to investigate the carbonate and chemical looping technologies for CO2 capture from coal combustion in semi-industrial scale. Advanced process models including 1D reactor models have been developed for chemical looping combustion and other processes. A highly sophisticated in-house CFD code using the discrete element method (DEM) for particle/particle interaction has been developed and validated for detailed 3D simulations of dense gas/particle flows. Advanced reaction models for solid fuel conversion, gas phase kinetics, radiative transfer etc. have been implemented into CFD codes for efficient simulations of solid fuel combustion devices.

Resources to be used in the project:

- 1 MW_{th} CLC pilot plant consisting of two interconnected CFB reactors
- ASPEN CLC process model including 1D reactor models

Contact:
Jochen Ströhle
P: +49 6151164791
jochen.stroehle@est.tu-darmstadt.de
www.est.tu-darmstadt.de

Flemish Institute of Technological Research (VITO)

The Flemish Institute for Technological Research (VITO) implements client-driven research projects and develops innovative products and processes. The multidisciplinary skills and technological knowhow of this 750-people strong R&D community make it a crossroads of technology, where state-of-the-art technologies are successfully blended into practical applications. The Ceramic and Powder Metallurgy team of VITO is embedded in a well-equipped R&D-environment. For preparation, processing and shaping of ceramic materials, it has semi-industrial infrastructure and equipment at its disposal, including spray drying, sieving, mixing and milling equipment and the necessary furnaces for calcination and sintering. In addition, the group has a vast range of characterization facilities such as measurement of particle size distribution, specific surface area, porosity, ζ-potential, contact angle, rheology, thermo-gravimetric and differential thermal analysis. The group has strong collaboration with the Waste Recycling Technology group that addresses recycling and re-use issues in the SUCCESS project, and with the Advance team that will address the environmental benefits and burdens from a life cycle perspective by applying Life Cycle Assessment (LCA) methodology. The Exposure and Risk assessment group, together with toxicologists of the team Applied Bio & molecular Systems have long-term experience in performing health and safety assessment of chemicals and products for workers, consumers and the environment, which includes in house performance of eco-toxicity tests and non-animal alternative tests.

Contact:
Frans Snijkers
P: +32 473 34 12 16
F: +32 14335665
frans.snijkers@vito.be
www.vito.be

Eurosupport Advanced Materials

Euro Support offers manufacturing, scale-up and development services in the area of catalysts, adsorbents and similar inorganic materials. With in-house capacity spanning from a few grams to hundreds of tons, our capabilities cover a wide range of synthesis, forming and treating processes. As leading independent service provider we focus on utilizing our extended manufacturing technology, know-how and expertise to manufacture your products to your specification or recipe. Our head office is located in Amersfoort (Netherlands) with manufacturing plants manufacturing plants in Uden (Netherlands) and Litvinov (Czech Republic).

Contact:
Knuth Albertsen
P: +31 413283286
F: +31 413283369
knuth.albertsen@eurosupport.nl
www.eurosupport.nl

Johnson Matthey

Johnson Matthey is a UK based specialty chemicals company focused on its core skills in catalysis, precious metals, fine chemicals and process technology. The company employs around 10000 people worldwide located in over 30 countries. Johnson Matthey's principal activities are the manufacture of autocatalysts, heavy duty diesel catalysts and pollution control systems, catalysts and components for fuel cells, catalysts and technologies for chemical processes, fine chemicals, chemical catalysts and active pharmaceutical ingredients and the marketing, refining, and fabrication of precious metals. More than half of JM’s products have a direct environmental benefit, a figure that is set to increase as a key part of the company’s growth strategy is to focus on emerging environmental opportunities.

Role in SUCCESS

Johnson Matthey’s role in SUCCESS will be in the development of industrially ready methods for the impregnated oxygen carriers, the supply of a large batch (up to 0.5 tonnes) for pilot-scale testing, and in the support of some of the technical-economic studies. This will involve materials preparation, and formulation activities coupled with small scale testing and characterization of the materials pre- and post-testing. Research process engineering work will be carried out in support of product and process understanding.

Johnson Matthey’s participation in SUCCESS will be through the Technology Centres based in Reading and Billingham, UK. These central facilities act as focal points for the development of new technologies into emerging market applications. At these sites, there are established facilities for the preparation, advanced characterization and testing of materials, as well as access to pilot scale manufacturing assets and state-of-the art analytical and electron optic equipment.

Contact:
Suzanne Ellis
P: +44 1189242157
F: +44 1189242025
elliss@matthey.com
www.matthey.com

Bertsch Energy

Bertsch Energy is a member of the family owned Bertsch Group located in Bludenz, Austria and currently employs approximately 250 people. With more than 80 years of experience, the main activities of Bertsch Energy are designing and manufacturing of heat recovery steam generators for combined cycle plants, biomass or waste fuel combustors for heat and power production (applying stoker fired furnaces as well as fluidized bed and grate technology) and waste heat boilers for different processes downstream of syngas and sulfuric acid production plants. Additionally, one internal division focuses on the construction of pressure vessels and different apparatuses. Bertsch Energy has their own manufacturing facilities which allow the production of all major parts of modern steam boilers. In 2006/2007, Bertsch Energy was engaged to design and manufacture the 30 MW steam generator for the oxy-fired lignite coal CCS demonstration power plant at "Schwarze Pumpe" owned by Vattenfall.

Contact:
Otmar Bertsch
otmar.bertsch@bertsch.at
www.bertsch.at

Electricité de France

Electricité de France (EDF), a leader in the European energy market, has developed an integrated network of companies servicing over 36 million clients in Europe. EDF R&D provides guidance to the EDF group in its technological choices, in order to keep electricity costs competitive, prepare the generating facilities of the future, enhance the quality of supply while preserving the environment, as well as to develop innovative solutions. EDF R&D has developed a strong know-how on the 3 CO_2 Capture routes. EDF built in 2012 its own capture pilot plant based on Alstom's AAP (Advances Amines Process). The CO2 capture team of EDF R&D's research group "new generation technologies and thermochemistry" was/is embarked in various collaborative research projects: OXYCOAL 1 and 2 (UK), RELCOM, H2IGCC, OCTAVIUS (EU), DALMATIEN, AMELIE-CO2 (FR) and is also strongly involved in testing at EDF's Capture Pilot Plant (1,1 t/h CO2 captured / Alstom's Advanced Amine Process) at Le Havre Power Plant. Our main expertise lies in process optimization (e.g. capture flowsheet) and overall CCS plant assessment, but more specific studies are also on-going: solvent/degradation products characterization and mechanisms, material issues and selection, combustion, etc. Methodologies and tools to assess new technologies in terms of performance, maturity and costs, such as: Technology Readiness Level (TRL) assessment, bottom-up costing approach based on equipment sizing and Life Cycle Assessment (LCA) for electricity are commonly used.

Contact:
Myriam Louis-Lousy
myriam.louis-louisy@edf.fr
www.edf.com

Shell Global Solutions

Shell Global Solutions International is a Dutch-based company and a wholly owned member of the Royal Dutch/Shell Group. The organization carries out research and provides technical services to Shell companies and a growing number of other customers in the oil, gas and chemical processing business, and in related energy and manufacturing industries.

Contact:
Ytalo Dávila
ytalo.davila@shell.com
www.shell.com

TOTAL Refinery & Chemicals

Total is a multinational energy company committed to leveraging innovation and initiative to provide a sustainable response to humankind's energy requirements. The fourth largest publicly-traded integrated oil and gas company and a world-class chemicals manufacturer, Total operates in more than 130 countries and has 95, 000 employees. Total engages in all aspects of the petroleum industry, including Upstream Operations (oil and gas exploration, development and production, LNG) and Downstream Operations (refining, marketing and the trading and shipping of crude oil and petroleum products). Total also produces base chemicals (petrochemicals and fertilizers) and specialty chemicals for the industrial and consumer markets (rubber processing, adhesives, resins and electroplating). In addition, Total has interests in the coal mining and power generation sectors. Total is helping to secure the future of energy by progressively expanding its energy offerings and developing complementary next generation energy activities such as solar power and biomass. Over 4,000 researchers are developing and improving processes and products in research centres located in France, Belgium, UK, Norway, United States, Canada and Qatar. Total aims to pursuing research and development to develop "clean" sources of energy, contributing to the moderation of the demand for energy and participating in the effort against climate change.

Contact:
Mahdi Yazdanpanah
mahdi.yazdanpanah@total.com
www.total.com

University of Natural Resources and Life Sciences Vienna

Founded in 1872, the Universität für Bodenkultur Wien / University of Natural Resources and Life Sciences, Vienna, known by its acronym "BOKU", now comprises 15 departments and four service centres in Vienna, as well as a number of experimental centres around Vienna. The university is attended by approx. 10,000 students, provides study courses at the bachelor, masters and doctoral levels, has approx. 1,800 staff who are engaged in teaching and research, a broad range of external lecturers, and some 4,603 persons working in services and administration. The university sees itself as a teaching and research institution that focuses on renewable resources that are a prerequisite for human existence. The relationships between the individual, society and the environment form the basis of all activities at BOKU, and its foremost aim is to make decisive contributions to securing the well-being of future generations. In this endeavour, it will seek ways of ensuring a sustainable and environmentally sound management of natural resources by allying the competences of the natural, engineering, economic, and social sciences. The Department of Material Sciences and Process Engineering comprises five institutes. One of these is the Institute of Chemical and Energy Engineering. The institute has been active in the field of thermodynamic modelling and simulation of energy technology processes, and both software tools and experience are available for description of energy conversion processes including non-conventional fuel conversion. The available resources to be contributed comprise: Software licences for assessment of complex energy conversion processes.

Contact:
Tobias Pröll
P:+431476543531
tobias.proell@boku.ac.at
www.boku.ac.at