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The EU-FP7 project SUCCESS – Scale-up of oxygen carrier for chemical looping combustion using environmentally sustainable materials

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Abstract

The paper gives a high level overview of the work performed in the EU-FP7 funded project SUCCESS (Scale-up of oxygen carrier for chemical looping combustion using environmentally sustainable materials). The project is the most recent one in a series of successful EU-funded research projects on the chemical looping combustion (CLC) technology. Its main objective is to perform the necessary research in order to demonstrate the CLC technology in the range of 10 MW fuel power input. The main focus is on scale-up of production of two different oxygen carrier materials using large scale equipment and industrially available raw materials. This will guarantee availability of oxygen carrier material at tonne scale. The scale-up of the two materials, a Cu and a Mn based, was successful and first tests with the Cu material have already been performed in four different pilot units up to 150 kW where the material showed excellent performance regarding fuel conversion. In addition to technology scale-up, extensive end-user evaluation is performed. This evaluation includes investigations on health, security and environmental impacts (HSE), a life cycle analysis and a techno-economic analysis to compare the CLC technology for steam generation against the current state-of-the-art technologies.

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Keywords: Chemical looping combustion, carbon capture, carbon capture and storgae, oxygen carrier, fluidized bed, natural gas

1. Introduction

1.1. Chemical looping combustion

Chemical looping combustion (CLC) is an innovative combustion technology with inherent CO_2 capture and nearly no energy penalty [1]. 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 break-through CO_2 capture technology [2].

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 Fig. 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 CO_2 stream can be obtained. The total heat release in the two reactors is exactly the same as in normal combustion processes.

Nomenclature

AR	Air reactor
CFB	Circulating fluidized bed
CLC	Chemical looping combustion
EU	European Union
FR	Fuel reactor
LCO_E	Levelized costs of energy
OC	Oxygen carrier
TLV	Occupational exposure limit values
X_{CH4}	Methane conversion
γ_{CO2}	CO_2-yield

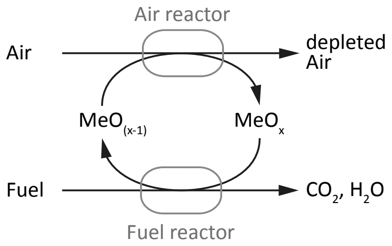

Fig. 1. Concept of chemical looping combustion.

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 [3].

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 [4, 5]. Further, several promising nickel free oxygen carriers have been identified reaching full fuel conversion and being economic and environmentally attractive [6]. From a technology point of view, chemical looping combustion of gaseous fuels can be seen close to demonstration at next scale (10 MW).

1.2. Project objectives and concept

The main objective of the EU-FP7 funded project SUCCESS (Logo in Fig. 2) 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 [6]. 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.

Fig. 2. Logo of the SUCCESS project

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

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 Fig. 3.

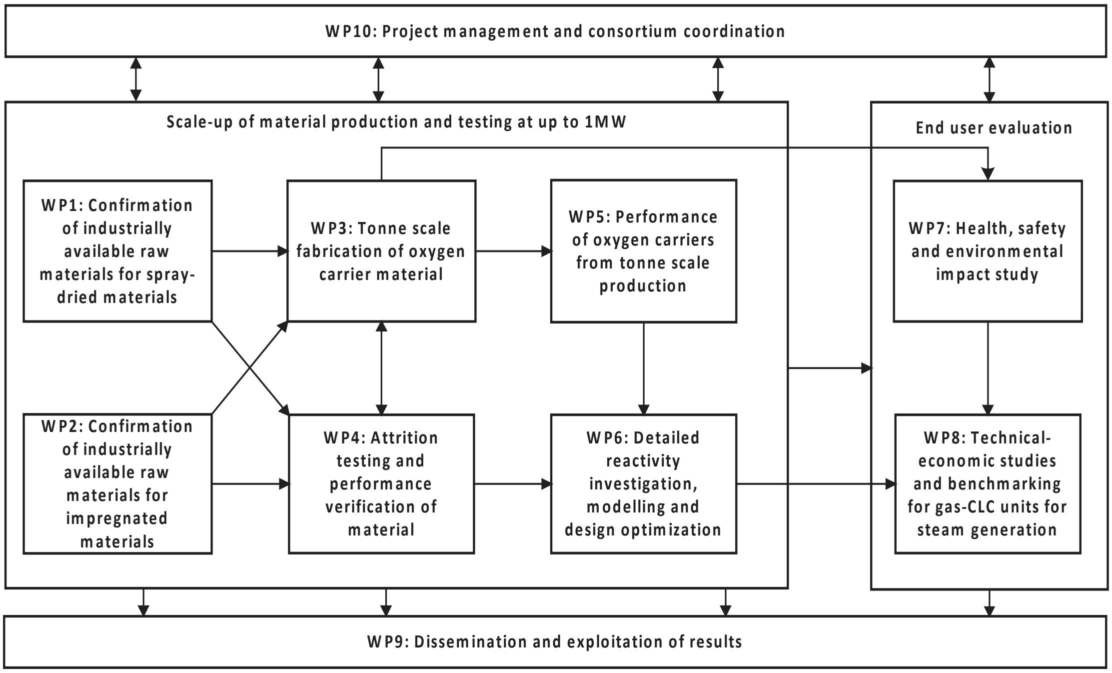

Fig. 3. Project structure

1.3. Project consortium

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. The SUCCESS project partners.

Partner name	Country	Type	Activities
Vienna University Technology	Austria	University	Project coordination, pilot plant OC testing, 10 MW system design
Chalmers University of Technology	Sweden	University	OC development, pilot plant OC testing, attrition testing
CSIC	Spain	Research Institute	OC development, OC testing, attrition testing, reactivity investigations, modelling, OC recycling investigation
IFP Energies Nouvelles	France	Research Institute	Pilot plant OC testing, attrition testing, reactivity investigations
Institut National Politéchnique de Toulouse	France	University	3D-Modelling
SINTEF Materials	Norway	Research Institute	Reactivity investigations, attrition testing
SINTEF Energy	Norway	Research Institute	Pilot plant OC testing
Darmstadt University of Technology	Germany	University	Pilot plant OC testing (1 MW)
VITO	Belgium	Research Institute	OC development, life cycle analysis, health safety and environmental impact evaluation
Euro Support Advanced Materials	Netherlands	Material producer	Large scale production of OC material
Johnson Matthey	United Kingdom	Material producer	Large scale production of OC material
Bertsch Energy	Austria	Boiler manufacturer	Sizing and design of equipment for economic evaluation
Électricité de France (EDF)	France	Power	Techno-economic evaluation
Shell Global Solutions	Netherlands	Oil&Gas	Health safety and environmental impact evaluation
TOTAL Raffinage Chimie	France	Oil&Gas	Pilot plant OC testing, attrition testing, techno-economic evaluation
University of Natural Resources and Life Sciences	Austria	University	Mass and energy balance calculations for techno-economic evaluation

2. Oxygen carrier development

Production of two different OC materials will scaled-up to multi tonne scale in SUCCESS. This scale-up does not only include scale-up of production equipment but also use of raw materials available in large quantities. The two materials have different active metal oxides (Mn and Cu) and are made by two different production methods, spray-drying and impregnation where the recipes were developed in earlier research projects.

2.1. Spray-dried CaMn particles

The spray-dried CaMn perovskite materials were developed within the EU-FP7 funded project INNOCUOUS [6-8]. The material is a so-called CLOU material (Chemical Looping with Oxygen Uncoupling [9]) and able to release gaseous oxygen in the fuel reactor to improve fuel conversion. For the scale-up of material production, different Mn sources in the form of Mn ores and Mn oxides were examined and investigated. The key question here is, how the impurities (see Table 2) and the different parameters of the spray-drying process affect the final particle properties like reactivity and lifetime. In order to find the best raw materials and production methods, 24 small batches of materials have been spray-dried and evaluated in small batch reactors. More detailed information about the scale-up process and influence of raw materials can be found in Jacobs et al. [10]. A summary of the used Mn sources is listed in Table 2.

Table 2. Overview of different Mn sources (data from [10]).

Material	Supplier	Mn oxide	Mn content	Main impurities	BET [m^2/g]
Hausmannite LM type	Erachem-Comilog	Mn_3O_4	68%	1-2% Fe, <1% Al	2.7
Elkem Colormax P	Elkem, Norway	Mn_3O_4	69	1% Al, 1% Fe	1.4
BassTech	BassTech	MnO	>76-78	-	23.3
CDMA	Erachem-Comilog	MnO_2	51	2-3% Fe, 1-2% Si, 4% Al	28.3
Battery grade N60	Autlan	MnO_2	45	5-6% Fe, 5-6% Si, 2-3% Al, 2-3% Ca	33.3

Based on the investigations, the best candidate was selected for production of up to 2 tons of material by Euro Support Advanced Materials using large scale equipment. This material will be tested in four different pilot units up to 150 kW fuel power input and also in the 1 MW unit at Darmstadt University of Technology.

2.2. Impregnated CuO particles

The recipe for the impregnated material, CuO on Al_2O_3, has also been developed before the start of the project by CSIC and was already tested before in different pilot units up to 120 kW [11-16]. The OC material has an active Cu content of 14 wt% and is impregnated on a γ-Al_2O_3 support material. The main task in scale-up of this material was identification of an industrially available support material and keep the excellent properties of the original benchmark material. Several smaller batches have been produced and tested in a continuous 500 W unit. The best and scaled-up OC material using industrially available support materials and relevant protocols was manufactured by Johnson Matthey using the incipient wet impregnation method. It is here referred to as *Cu14γAl_Commercial*.

The properties of fresh and used particles and the comparison to the original benchmark material are shown in Table 3.

Table 3. Properties of scaled up material (fresh and used) in comparison with benchmark material (data from [17, 18]).

Oxygen Carrier	Cu14γAl_Commercial		Benchmark Material
	Fresh	Used	Fresh
Total CuO content [wt%]	13.8	10.1	69
Particle size [µm]	153	151	
Porosity [%]	53.0	49.1	
Mechanical strength [N]	1.2	1.2	
AJI	3.1	2.6	
XRD phases	CuO, $CuAl_2O_4$, δ-Al_2O_3	CuO, $CuAl_2O_4$, $CuAlO_2$, δ-Al_2O_3, α-Al_2O_3	CuO, $CuAl_2O_4$, δ-Al_2O_3

The material has been extensively tested in a 500 W continuously operation unit (information about unit in [17]) where fuel conversion above 95% has been reached.

3. Oxygen carrier testing

The oxygen carrier material produced using large scale equipment is tested in four different pilot units up to 150 kW fuel power input with different designs to allow an unprecedented characterisation of the oxygen carrier material. Further, each unit has special features for very detailed investigation of parameters like solids circulation, solids inventory, long-term stability and sulphur tolerance.

Furthermore, the spray-dried perovskite material will be used to demonstrate the CLC technology at 1 MW fuel power input. The mechanical stability of the oxygen carrier materials is tested under hot and cold conditions in three different units.

3.1. Available pilot plants

Continuous pilot unit testing is performed in five pilot units up to 1 MW fuel power input. Each of these units shows specific advantages and potential disadvantages regarding specific inventories, solid circulation rates and possible range of operating parameters. However, the differences in designs give great opportunity for extensive material testing and identification of important fuel conversion mechanisms. A summary of the different pilot units and their special features is shown in Table 4. Sketches and flow sheets of the pilot units are shown in Fig. 4- Fig. 6.

Table 4. Pilot units up to 1 MW used in SUCCESS.

Operator	Fuel power	Fuel	Special feature
Chalmers University of Technology	10 kW	Natural gas	• Overnight operation • High gas velocities for attrition testing
IFP Energies Nouvelles	10 kW	CH_4, CO, H_2	• L-valves for control of solid flow rate • Control of gas and solids residence time • 3 reactors (2xAR, 1xFR)
Vienna University of Technology	120 kW	Natural gas, CO, H_2, Hydrocarbons, H_2S	• High solids circulation rates • Operation with higher hydrocarbons and sulphur
SINTEF Energy	150 kW	Natural gas	• Routing of particle flow • Internal recirculation of particles
Darmstadt University of Technology	1 MW	Natural gas	• Fully refractory lined reactors • Post combustion chamber using pure oxygen

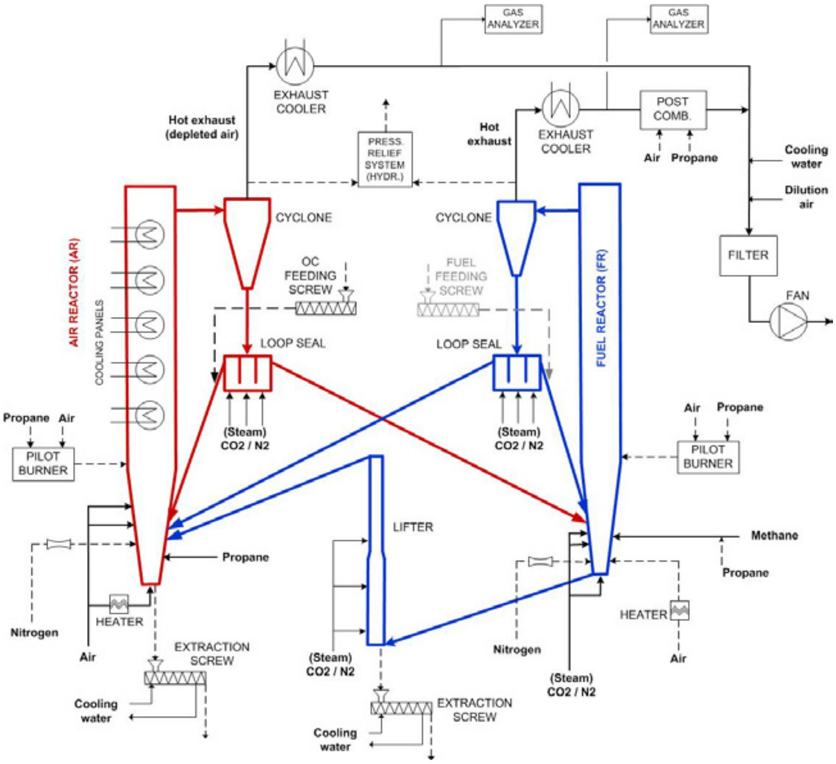

Fig. 5. SINTEF 150 kW unit.

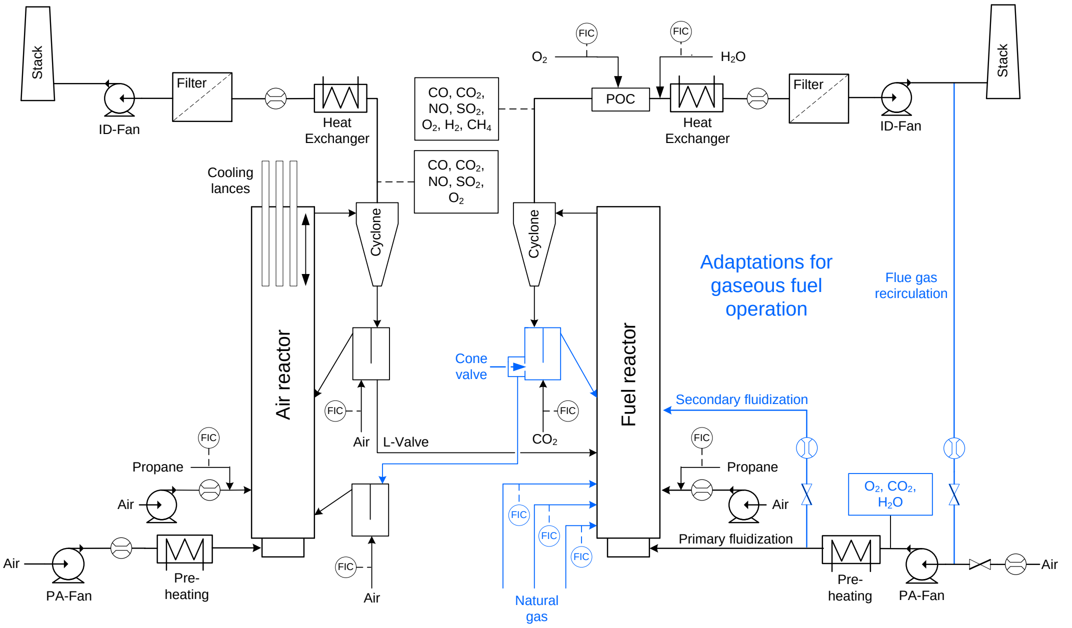

Fig. 6. Darmstadt 1 MW unit. Blue lines show adaptations for natural gas operations.

3.2. First results from material testing

So far, the impregnated *Cu14γAl_Commercial* OC has been tested in pilot units up to 150 kW where it showed very good performance. The two most important performance parameters for comparison of different operating conditions are the methane conversion written as

$$X_{CH4} = 1 - \frac{y_{CH4,FR}}{y_{CH4,FR} + y_{CO2,FR} + y_{CO,FR}} \tag{1}$$

and the CO_2-yield written as

$$\gamma_{CO2} = \frac{y_{CO2,FR}}{y_{CH4,FR} + y_{CO2,FR} + y_{CO,FR}} \tag{2}$$

The material performed outstanding in all units reaching methane conversions X_{CH4}>90% (for more detailed information see [19, 20]). When comparing these numbers, it has to be taken into account that the investigated particles were not the design particles for these pilot units. Detailed results for the pilot units are shown in Fig. 7-Fig. 9.

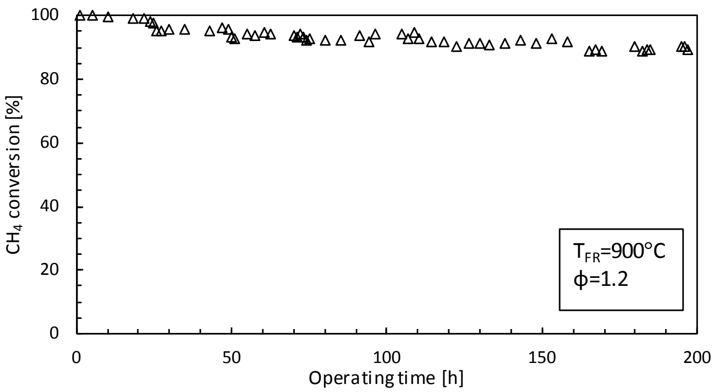

Fig. 7. Results from testing in 10 kW IFP unit. CH_4-conversion over operating time (data from [19]).

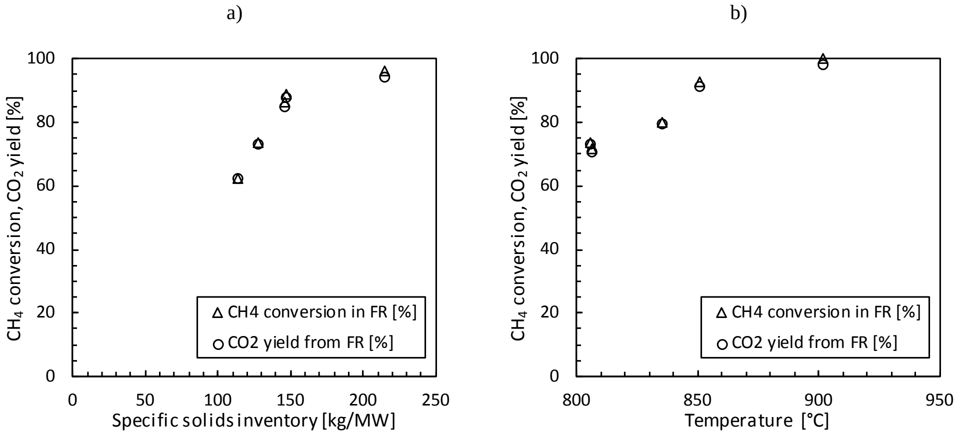

Fig. 4. Results from testing in Vienna 120 kW unit. a) Influence of solids inventory, b) influence of temperature.

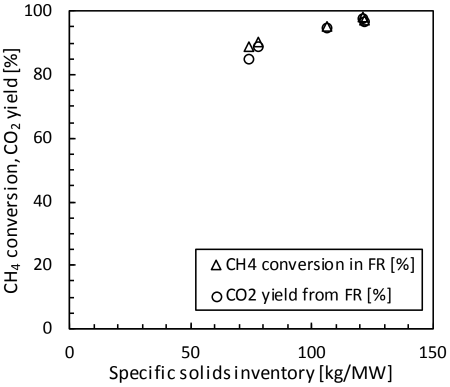

Fig. 9. Results from testing in 150 kW SINTEF unit. Influence of solids inventory (data from [20]).

4. End-user technology potential evaluation

In addition to experimental process investigations and development, a lot of effort is put into end-user evaluation of the CLC technology for industrial steam generation. These investigations includ extensive health, security and environmental impact assessment of the two scaled-up oxygen carrier materials, a life cycle analysis and techno-economic evaluation of different application scenarios.

The environmental impact assessment investigated potential risks related to humans and the environment in a three tier approach. Based on the results, detailed recommendations for required safety equipment and material handling can be made. Acute and chronic toxicity tests were performed on algae, waterflea, and zebrafish embryo to assess the human and environmental hazard of fresh and spent oxygen carriers. Acute risk on aquatic ecosystem is low except Al poisoning of algae is possible using the Cu-based OC material. For the exposure assessment a realistic exposure level for workers was established. The potential human exposure has been evaluated by simulation experiments (generating dust under controlled conditions). The risk evaluation is based on measured data (during dust generation, as worst case proxy for OC handling) and occupational exposure limit values (TLVs). It showed a potential risk for Mn exposure.

Techno-economic evaluations were performed based on methods from the European Benchmarking Task Force (EBTF). To allow the comparison of CLC with other CO_2 capture technologies, a techno-economic study will be performed. As basis for this study, a natural gas reference case with and without post combustion CO_2 capture via mono-ethanol-amines was established for several capacities and steam parameters. In total, two different cases, one with high grade steam and one with low grade steam will be evaluated. The first case is preferable for power (high grade steam) whereas the second case is for oil and gas industry (low grade process steam). All necessary plant equipment is considered in both cases, state-of-the-art penalty demonstrated in real conditions as well as up-to-date thermal integration options are integrated for CO_2 capture. In both cases the levelized cost of energy ($LCOE_E$) increases.

5. Conclusions and outlook

The objective of the EU-FP7 funded project SUCCESS is to close the gap between the state-of-the-art of chemical looping combustion for gaseous fuel and next scale demonstration in the size of 10 MW fuel power input. To achieve the goal, necessary research is performed in the fields of oxygen carrier production at large scale, extensive material testing in pilot units up to 1 MW and end-user evaluation regarding techno-economic and life-cycle performance. The production methods of two different oxygen carrier materials based on Mn and Cu have been successfully scaled-up to the tonne scale using industrially available raw materials. First tests of these materials in pilot units up to 150 kW show very good results regarding fuel conversion performance and particle lifetime.

Intensive investigations regarding health, safety and environmental impact assessment of the oxygen carrier materials give detailed instructions for material handling and necessary safety equipment. To evaluate the techno-economic potential of the CLC technology for industrial steam generation, comparison between the new technology and a reference case is performed based on methods from the European Benchmarking Task Force. Here, two different cases, high grade and low grade steam, are investigated.

Acknowledgements

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