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Negative CO_2 Emissions with Chemical-Looping Combustion of Biomass – a Nordic Energy Research Flagship Project

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Abstract

The Nordic countries constitute a natural location for the development and deployment of Bio-Energy with Carbon Capture and Storage (BECCS). Finland, Sweden and Denmark are world-leading with respect to heat and power generation from sustainable biomass. Norway is world-leading with respect to Carbon Capture and Storage (CCS). The Nordic countries also have ambitious targets for reductions of their CO_2 emissions, host leading technology providers, and have large biomass potential per capita. System studies suggest that bioenergy could be the single largest energy carrier in the Nordic countries by 2050. *Negative CO_2 Emissions with Chemical Looping Combustion of Biomass* is a multi-partner project with the goal to develop new technology that: i) enables CO_2 capture and negative CO_2 emissions at the lowest possible cost, ii) is able to produce power and steam for industrial and other applications, iii) utilizes Nordic expertise in fluidized bed technology and iv) has potential to achieve improved fuel utilization. The technology capable of achieving these goals is Chemical-Looping Combustion of biomass (Bio-CLC). The article presents the project and features some early results from its implementation.

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Keywords: Chemical-looping combustion; Biomass; Negative emissions; Carbon capture and storage; CLC; BECCS

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1. Introduction

The Paris Agreement adopted by consensus on 12 December 2015 within the United Nations Framework Convention on Climate Change stipulates that the increase in the global average temperature should be limited to well below 2°C above pre-industrial levels, and pursue efforts to limit the temperature increase to 1.5 °C. In order to meet this target, the emissions of greenhouse gases likely will have to be reduced to zero in less than twenty years. This is an extremely difficult task to achieve only by cutting emissions. Therefore it is very likely that active removal of CO_2 from the atmosphere will become necessary. A recent compilation of climate scenarios consistent with limiting surface warming to 2°C show that out of 116 available scenarios no less than 101 require global net negative CO_2 emissions in the second half of this century [1].

The Nordic countries (Denmark, Finland, Iceland, Norway and Sweden) have ambitious targets for reductions of their CO_2 emissions by 80-100% in 2050 [2]. This is a technical, economic and political challenge because heavy industries (metallurgical, cement, chemicals, mining, oil, natural gas, pulp and paper) constitutes an important share of the Nordic economy. Therefore, to make Nordic (and global) emission targets realistically attainable, it is clear that affordable technology for achieving negative CO_2 emissions will be necessary.

One method to realize substantial negative CO_2 emissions is Bio-Energy with Carbon Capture and Storage (BECCS). The IPCC [3] states that "…technologies such as BECCS (Bio-Energy CCS) are fundamental to many scenarios that achieve low-CO_2eq concentrations". Moreover, "BECCS features prominently in long-run mitigation scenarios for two reasons: (1) The potential for negative emissions may allow shifting emissions in time; and (2) in scenarios, negative emissions from BECCS compensate for residual emissions in other sectors (most importantly transport) in the second half of the 21st century". A recent major study also identifies CCS and bioenergy as the two most valuable technologies for achieving climate policy objectives (more important than energy efficiency improvements, new renewables, nuclear power), motivated by their combined ability to produce negative emissions via BECCS [4].

The Nordic countries constitute an excellent location for the development and deployment of this kind of technology. Finland, Sweden and Denmark are world-leading with respect to heat and power generation from sustainable biomass. Norway is world-leading with respect to Carbon Capture and Storage (CCS) and operates two of the largest CCS operations in the world (Utsira, Snøhvit). The Nordic countries also have quite strong economies, is home to leading technology providers in the energy sector, and have huge forests and large areas of arable land available per capita. Results from recent system studies suggest that bioenergy could very well be the single largest energy carrier in the Nordic countries by 2050 [5]. Together these factors suggest that the Nordic countries is a natural location for large-scale deployment of BECCS.

Nordic Energy Research (NER) is a platform for cooperative energy research and policy development under the auspices of Nordic Council of Ministers. In this capacity, it funds research of special interest to the Nordic region. BECCS has been identified as one such subject and this paper presents one of three so called flagship projects funded by this organization.

The project is called '*Negative CO_2 Emissions with Chemical Looping Combustion of Biomass*' and is a multi-partner and cross-disciplinary project running 2015-2019. The ultimate goal of the project is the development of new competitive technology that: i) enables CO_2 capture and negative CO_2 emissions with the lowest possible cost and energy penalty, ii) is able to produce power and steam for industrial and other applications, iii) utilizes Nordic expertise and competence in fluidized bed technology, iv) makes thermal formation of NO_x impossible and v) has potential to achieve more efficient fuel utilization compared to ordinary biomass combustion. The technology capable of achieving these goals is Chemical-Looping Combustion of biomass (Bio-CLC).

2. Chemical-Looping Combustion of Biomass (Bio-CLC)

The fundamental principle of Chemical-Looping Combustion (CLC) is that hydrocarbon fuel is oxidized using two separate reactor vessels, referred to as the Air Reactor (AR) and the Fuel Reactor (FR) [6, 7]. A solid oxygen carrier in form of metal oxide particles (Me_xO_y) performs the task of transporting oxygen between the two reactors, as shown in Fig. 1.

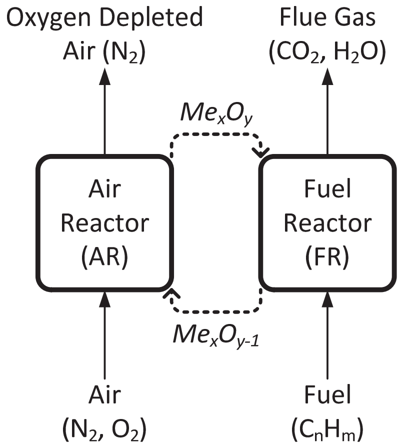

Fig. 1. Schematic description of Chemical-Looping Combustion (CLC).

In the fuel reactor, the oxygen carrier particles are reduced by the fuel (C_nH_m), which in turn is oxidized to CO_2 and H_2O. In the air reactor, the particles are oxidized with O_2 from air. The reactor temperature is in the range 800-1050°C and the net energy released in the system as a whole is the same as in ordinary combustion. An example of the reactions in each reactor and for the whole system can be found in reactions (1-3) below, which describe CLC of methane (CH_4) with manganese(II,III) oxide as oxygen carrier.

[FR]:	$CH_4(g) + 4Mn_3O_4(s) \rightarrow 12MnO(s) + CO_2(g) + 2H_2O(g)$	$\Delta H^o_{298} = 125$ kJ/mol$_{CH4}$	(1)
[AR]:	$2O_2(g) + 12MnO(s) \rightarrow 4Mn_3O_4(s)$	$\Delta H^o_{298} = -927$ kJ/mol$_{CH4}$	(2)
[Sum]:	$CH_4(g) + 2O_2(g) \rightarrow CO_2(g) + 2H_2O(g)$	$\Delta H^o_{298} = -802$ kJ/mol$_{CH4}$	(3)

Direct contact between fuel and air is avoided and therefore the combustion products are not diluted with N_2 from air. Pure CO_2 for sequestration can be obtained simply by cooling down the flue gas from the fuel reactor and condensing steam to liquid water. The reactions in both reactor vessels are flameless and takes place at moderate temperatures so there is no possibility for formation of thermal NO_x. Heat is generated and thus needs to be extracted mainly in connection to the exothermic air reactor, where the gas flow also is the largest. This characteristic could be of great advantage compared to ordinary boilers since the heat transferring surfaces could be designed with less concern about the fouling and corrosive effect of ash than normally is the case in biomass combustion. Thus less costly materials could be feasible and improved operability can be expected.

The most commonly proposed way to realize CLC is to utilize Circulating Fluidized Bed (CFB) technology with oxygen carrier particles as bed material. The fuel reactor would be a separate fluidized vessel located in between the particle return leg of the cyclone and the main body of the boiler. This could be arranged by dividing a conventional boiler into two sections by means if an isolated wall or by addition of a second vessel. The design changes would be rather substantial but seems unlikely to add significant costs for the plant as a whole. The particle size would be the range 0.1-0.4 mm, i.e. comparable to fine sand. Cheap and readily available minerals such as iron ore, manganese ore or ilmenite (iron-titanium ore) will likely suffice as oxygen carrier.

As both the high energy penalty and the high capital cost associated with gas separation can be avoided it is evident that CLC is a potential breakthrough technology for CO_2 capture. In a detailed study concerning the design of a large CLC plant for combustion of coal the CO_2 avoidance cost was estimated to 16-26 €/tonne CO_2, including costs of CO_2 treatment and compression [8]. Other studies are even more optimistic, suggesting that costs of about 10 €/tonne CO_2 could be possible [9]. In any case, CLC is expected to have at least 50% lower energy penalty and cost than any other CO_2 capture technology for solid fuels applications. Biomass should not be different from coal in this respect.

The development of CLC has been quite rapid. As of 2016, it can be estimated that CLC has been demonstrated for coal and gas in >24 pilot units during >7500 h of operation. The largest pilot unit operated has a thermal power of 3 MW [10]. In another notable instance, 1000 h of operation was recorded using the same batch of oxygen carrier with excellent results [11]. Essentially 100% fuel conversion and 100% CO_2 capture has been demonstrated [12]. A lot of attention have been paid to developing solid materials that can be used as oxygen carrier in the process. In total >900 different oxygen-carrier materials have been investigated in laboratory scale. The most commonly used oxygen carriers include metal oxides of the d-block transition metals Fe_2O_3, Mn_3O_4, CuO and NiO. Considerable work has been done also on mixed metal oxides, in particular manganese based mixed oxides are of interest due to their low price and ability to release gas phase O_2 in the fuel reactor [13]. Ores, minerals and industrial waste materials have also been examined as oxygen carriers. This option has considerable advantages with respect to price and availability and may very well be the best option for solid fuel applications.

Up to this point research about CLC has focused on coal and natural gas. In contrast, very limited research effort has been dedicated to biomass applications. Low-volatile biomass such as wood char has been examined to a certain extent and a few experiments involving saw dust are also reported. This project is intended to fill this research gap. The envisioned flow sheet for a complete Bio-CLC boiler is described in Fig. 2.

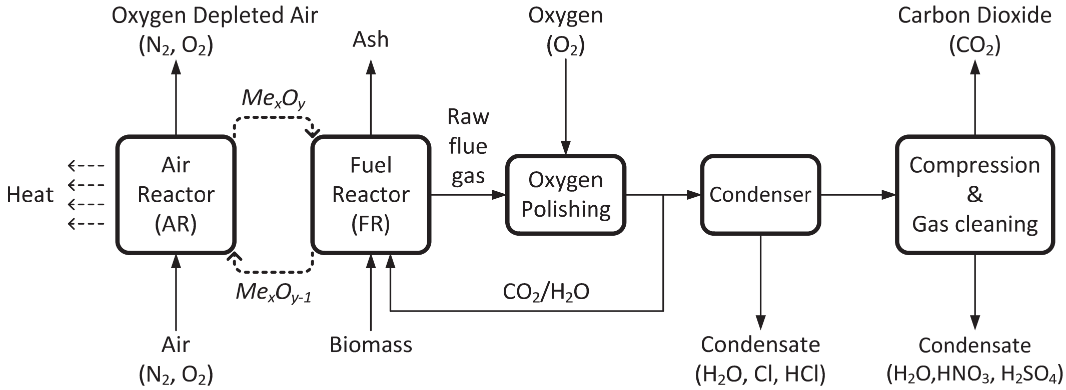

Fig. 2. Schematic description of the whole process chain for CO_2 capture by Bio-CLC.

It can be seen that the complete flow sheet involves several operations that needs to be considered. The fundamental performance of chemical-looping combustion of biomass and the influence of factors such as reactor design, choice of oxygen-carrier particles and choice of fuel is obviously of prime importance. None of these factors have previously been examined in detail for Bio-CLC. Other technical issues include ash and corrosion, which in turn will affect heat extraction and process integration. Further, oxygen polishing and flue gas cleaning will need to be considered if CO_2 of adequate quality is to be obtained.

3. Project description

3.1. Project goals

The main objective of the project is to bring Bio-CLC to the next level of development, which would enable the erection of a demonstration unit of at least semi-industrial scale (e.g. 10-100 MW_{th}) using fuels such as wood chips or forestry waste. This would enable the construction of a demonstration unit, i.e. the final step before commercial deployment. In order to enable this development a number of key research questions have been identified which will be answered during the project:

• Are there any key parts of the process chain for which current knowledge is insufficient? Three such areas have been identified: i) ash and corrosion issues in Bio-CLC ii), flue gas treatment in Bio-CLC, and iii) the specific requirements on oxygen-carrier materials for Bio-CLC. These three issues are addressed in separate work packages of the project.

• How does the performance of the core concept depend on factors such as reactor design, operational conditions, fuel properties and choice of oxygen carrier? This will be examined by pilot plant operation using experimental facilities of up to 150 kW_{th} size, located in three different Nordic countries.

• What are the key issues for up-scaling of oxygen-carrier handling and use in an industrial environment? This will be examined by demonstration of Bio-CLC at MW-scale in a semi-commercial research boiler.

• How can Bio-CLC be realistically deployed in the Nordic countries? This will be examined by developing a fundamental design of a Bio-CLC plant of relevant size and a detailed economic analysis of the concept. A detailed investigation of potential sites and available economic incentives will also be undertaken.

3.2. Project partners

The project consortium consists of the following parties:

- Chalmers University of Technology is located in Göteborg, Sweden. A research group dedicated to the development of CLC was started in 1998 working with the development of oxygen-carrier material. The group was the first to successfully demonstrate CLC in sustained operation in 2003. Later the group also was first to demonstrate CLC of both solid and liquid fuels.
- The Bellona Foundation is a non-governmental organization based in Oslo, Norway. Bellona works with environmental activists, scientific experts, governments, NGOs and industries to address the world’s most pressing environmental problems, find sustainable solutions and move towards a greener society. Bellona has worked on carbon negative solutions since 2008 and is a strong advocate for such technology.
- Sibelco Nordic AB is a Swedish subsidy of the global company Sibelco, a leading provider of industrial materials. It is one of the largest providesr of silica sand for fluidized bed combustion in the Nordic countries and has several potential oxygen carriers in its material portfolio such as manganese ores, rock ilmenite and sand ilmenite.
- SINTEF is a Norwegian research institute. SINTEF Energy Research is located in Trondheim while SINTEF Materials and Chemistry is a located in Oslo. SINTEF is a leading institution in the development of CCS technologies in general and also has long experience in the development of CLC, both with respect to oxygen carriers and prototype reactors.
- VTT Technical Research Centre of Finland Ltd is a Finish research institute. It is a leading institution in the development of biomass utilization and fluidized-bed technology and has been involved in upscaling and commercialization of such technologies, for example production of bio-oil by fast pyrolysis.
- Åbo Akademi University is located in Turku/Åbo in Finland. It is a leading institution in the development of biomass combustion and fluidized bed technology with long experience in ash and corrosion chemistry and combustion modelling.

3.3. Project activities

The project activities are organized in eight work packages, see Fig. 3 on the following page.

- WP1 concerns the overall management and coordination of the project.
- WP2 involves operation of three pilot plant reactors (150 kW_{th} at SINTEF, 100 kW_{th} at Chalmers, 50 kW_{th} at VTT) which will be adapted for experiments with biomass. The main output will be how the gas conversion and char burn-out depends on the factors such as reactor design, operational parameters, biomass properties and choice of oxygen carrier. The work package also includes the demonstration of Bio-CLC at MW-scale in a semi-commercial CFB research boiler located at Chalmers. The goal is to perform a test campaign of about 1-2 weeks using an interconnected reactor vessel for biomass gasification as fuel reactor and the boiler as air reactor. This should allow for the demonstration of Bio-CLC at large scale (1-4 MW_{th}) and demonstrate large scale (>10 tonnes) preparation and logistics of oxygen carrier particles.

- WP3 concerns oxygen carrier materials for Bio-CLC. In order to be feasible for commercial applications oxygen carrier material will need to i) have high reactivity with fuel and air, ii) have adequate oxygen transfer capacity, iii) have sufficient physical integrity for operation in fluidized beds, iv) be reasonably inert towards fuel impurities, v) be cheap, available and environmentally benign. The aim is to identify the most suitable materials for Bio-CLC by means of lab-scale experiments. Methods that will be used include Thermo Gravimetric Analysis (TGA) and Differential Thermal Analysis (DTA), redox experiments in Fluidized Bed (FB) and attrition testing. Materials will be further analysed by use of X-Ray Diffractometry (XRD), Scanning Electron Microscopy with Energy Dispersive analysis by X-rays (SEM/EDX). Another activity in the work package involves making a market assessment in order to ensure adequate supply of affordable oxygen carrier materials for a demonstration plant and beyond. The focus of the project will be on the use of cheap and readily available minerals such as ilmenite or manganese ore.

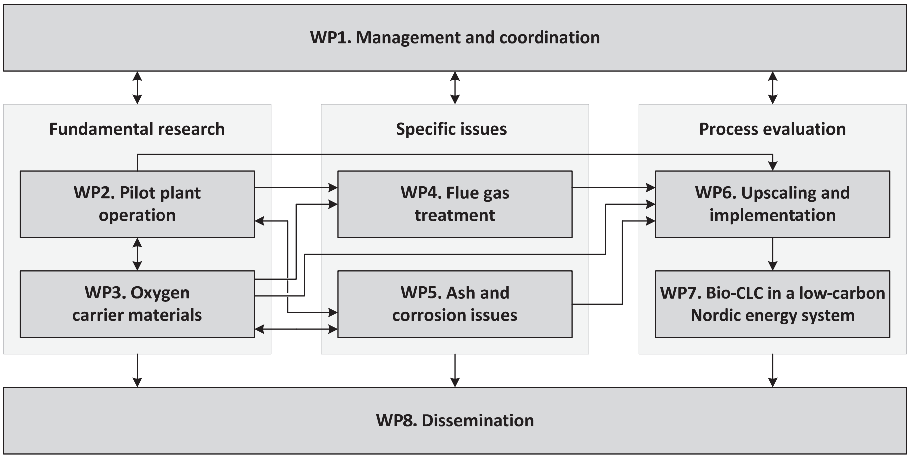

Fig. 3. Overview of project activities and interaction between work packages.

- WP4 deals with the design of an efficient flue gas cleaning method for Bio-CLC that is capable of converting raw flue gas from the fuel reactor to compressed CO_2 suitable for transport and storage. Bio-CLC provides possibilities with respect to flue gas cleaning compared to conventional combustion. The concentration of combustion products will be much higher due to the elimination of nitrogen dilution. Further, during the subsequent CO_2 compression step the temperature will be lower and the gas pressure elevated. Previous work with respect to the chemistry related to CO_2 gas cleaning strongly indicates that both nitrogen and sulfur oxides (NO_x and SO_x) can be captured in the form of acids in the liquid condensate during CO_2 compression, at these altered conditions. The concept will be studied by means of a combined modelling and experimental effort. Moreover, several studies indicate that a small fraction of combustibles will remain in the gas leaving the fuel reactor when solid fuels are used in CLC. Therefore an O_2 polishing step is needed to reach full conversion to CO_2 and H_2O prior to condensation and CO_2 compression. As a part of the WP, the design and performance of an O_2 polishing device will be experimentally examined.
- WP5 deals with ash and associated corrosion related issues which are critical aspects in biomass combustion. Bio-CLC will provide both opportunities and challenges in this area. The following phenomena will be studied in laboratory and in pilot-scale experiments: i) the interaction between oxygen carrier materials and biomass ash, ii) ash chemistry at reducing conditions, and iii) and corrosion behavior. The interactions between oxygen carrier materials and ash components will be studied in laboratory environment using methods such as Thermo Gravimetric Analysis (TGA), Differential Thermal Analysis (DTA) and Scanning Electron Microscopy with Energy Dispersive analysis by X-rays (SEM/EDX). Further, SEM/EDX will also be utilized to study the interaction between oxygen carriers and biomass ash during pilot plant experiments performed in WP2. The ash chemistry in the fuel reactor will be studied experimentally during the pilot plant runs. These studies will focus on

alkali release. Laboratory scale corrosion tests with simulated ash or deposit chemistry will be performed according to best possible knowledge of ashes from different biofuels.

- WP6 is about upscaling and implementation. A comprehensive plan showing how Bio-CLC could be implemented in the Nordic countries will be devised. This includes identification of potential sites for a demonstration plant, fundamental plant design and a techno-economic analysis. The prospects for providing funding for a demonstration plant will also be examined with the possibility of co-funding between industrial end-user and funding agencies being an option.
- WP7 deals with the economic performance and emission reduction (and removal) potential of Bio-CLC in the future Nordic energy system will be studied using energy system models, cost-benefit analysis and cost-effectiveness analysis. The main tool for the analysis will be VTT's energy system model TIMES-VTT. Multiple scenarios will be crafted and studied in order to form a good understanding of the desirable values of key parameters. The research effort in this work package will provide insights and recommendations on the cost-efficient CO_2 reduction potential of Bio-CLC in the Nordic countries.
- WP8 will ensure that the knowledge gained in the project is disseminated, not only among the partners but also to other stakeholders such as policy makers, researchers and industry, and also to ensure that knowledge generated in the project becomes available to the interested general public. A particular focus of the work package is to make the concept of negative CO_2 emissions and BECCS more known through demonstrating the potential of Bio-CLC in the Nordic countries.

4. Early results

The project was started in November 2015. During its first year several milestones have been reached including the commissioning and operation of VTT's pilot plant reactor, introduction of biomass as fuel in Chalmers pilot plant reactor, operation of O_2 polishing reactor at Chalmers and first reports with respect to emission reduction potential in the Nordic countries. The following results can be highlighted:

4.1. Operation of semi-commercial boiler/gasifier

Pre-experiments with manganese ore as bed material have been performed in the semi-commercial CFB research boiler/gasifier at Chalmers. The main results are summarized in Fig. 4 in which they are compared to the use of ilmenite and silica sand.

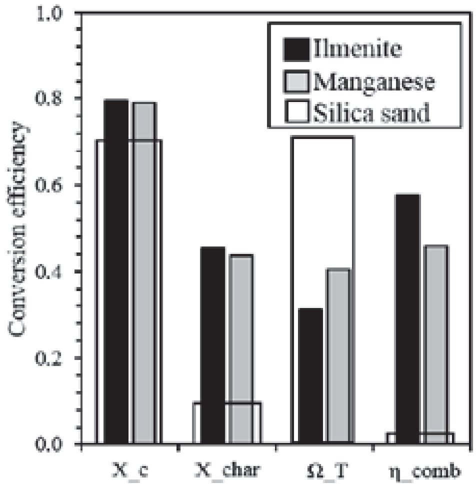

Fig. 4. Oxygen demand (Ω_T), combustion efficiency (η_comb), carbon conversion (X_c) and char conversion (X_char) for ilmenite, manganese ore and silica sand at 830°C. The figure is from Berdugo Vilches et al. [14].

It can be seen that simply by changing from silica sand to an oxygen carrying bed material in form of manganese ore or ilmenite in the semi-industrial circulating fluidized bed boiler/gasifier, the product composition in the gasification reactor completely altered. Basically, the gasification reactor turns into a 'fuel reactor' in what could be described as a 'pseudo-CLC plant'. The combustion efficiency (η_comb) is up to 60% already at 830°C, or up to 70% if char is discarded (1-Ω_T). By now, the research boiler has been operated in this mode over extended time periods, demonstrating the feasibility of Bio-CLC at MW-scale [14]. By improving fuel feeding (the gasifier now uses top-feeding), fuel reactor design and increasing temperature, the project consortium believes that fuel conversion above 90% could be achieved in future Bio-CLC plants. This remains to be demonstrated within the project, primarily by the use of the three pilot plants. A dedicated two-week campaign in the semi-commercial research boiler in which more benign operational conditions will be strived for, is scheduled within the project for 2018.

4.2. Pilot tests with wood pellets as fuel

Chemical looping combustion tests with biomass as fuel has been carried out in a new dual fluidized bed (DFB) process development unit located at VTT Bioruukki piloting center, in Espoo, Finland [15]. The DFB-CLC test rig consists of a circulating fluidized bed air reactor interconnected with a bubbling fluidized bed fuel reactor and has a nominal power of 10-50 kW_{th}. The research was performed in collaboration with the Finnish Carbon Capture and Storage Program (CCSP).

A set of tests were carried out using ilmenite as oxygen carrier. The fuel was Finnish (white) wood pellets and Norwegian steam treated (black) wood pellets. The main targets were to study the impact of operational parameters on the performance of CLC using biomass-based fuels with a high volatile content. Another aim was to assess the risk for high-temperature corrosion in the flue gas path subsequent of the air reactor. The gas compositions were monitored and fly ash was sampled from both the air reactor and the fuel reactor.

The DFB-CLC test rig was successfully in 9 tests lasting for 16 h in total. The main challenge encountered during the tests was to maintain a high enough bed temperature in the fuel reactor, which was operated at 840-860ºC. Another challenge was a short residence time of the bed material in the fuel reactor. The main reason for these challenges was that the test rig originally was designed for gasification purposes. Because of the less than optimal operational conditions the combustion efficiency was limited to approximately 60-70%. For improved performance of the ilmenite oxygen carrier a temperature of 900-950ºC would likely have been needed [16]. The test rig will be modified for the next experimental campaign within the project.

As for results, a clear dependency of the fluidization velocity on the CO_2 capture efficiency and fuel conversion was found. No accumulation of alkali components to the surface of oxygen carrier particles were found. The concentrations of potassium and sodium in the bed material remained constant and low, suggesting lower concentrations of vaporized alkaline components in the air reactor flue gas than in conventional biomass combustion applications. The results indicate that the risk of high-temperature corrosion of superheater tubes could be lower in bio-CLC than conventional biomass combustion.

5. Concluding remarks

The project will run for four years, i.e. until November 2019. Continuous updates about the progress can be found at the project homepage (http://www.nordicenergy.org/flagship/negative-co2/).

Acknowledgements

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