



Biomass direct chemical looping for hydrogen and power co-production: Process configuration, simulation, thermal integration and techno-economic assessment

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ABSTRACT

Large scale biomass utilisation in energy-related applications is of paramount importance to reduce the fossil CO₂ emissions. At European level, about a third of energy consumption is expected to be covered by renewables in the next 15 years. In addition, the CO₂ emissions need to be reduced by 40% compared to the 1990 level. Within this context, innovative energy-efficient low carbon technologies have to be developed. Chemical looping is a promising conversion option to deliver reduced energy and cost penalties for CO₂ capture.

This paper assesses biomass direct chemical looping (BDCL) concept for hydrogen and power co-production. The concept is illustrated using an ilmenite-based system to produce 400–500 MW net power with flexible hydrogen output (up to 200 MW_{th}). The performances are assessed through computational methods, with the mass and energy balances being used for in-depth techno-economic analysis. The biomass direct chemical looping delivers both high energy efficiencies (~42% net efficiency) with almost total carbon capture rate (>99%) compared to other CO₂ capture options (e.g. gas–liquid absorption). The economic parameters show also a reduced CO₂ capture cost penalty for biomass direct chemical looping technology compared to gas–liquid absorption (e.g. 7% reduction of specific capital investment).

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1. Introduction

The security of primary energy supplies and the environmental protection are objectives of great concern for the global energy sector. In terms of primary energy supply, the fossil fuels are predicted to remain the backbone of energy system in the short to medium period but the renewable energy sources (e.g. solar, wind, and biomass) become increasingly important. Within this context, the large scale biomass utilisation in energy-related applications is very important to reduce the fossil fuel dependence, to enhance the security of energy supply as well as to reduce the greenhouse gas emissions. Political measures are being put in place to foster the utilisation of renewable energy sources. At European Union (EU) level, about a third of energy consumption is to be covered by renewables by 2030 [1].

In terms of environmental protection and climate change prevention, the reduction of CO₂ emissions from fossil fuel-based industries (heat and power sector as well as other energy-intensive industrial applications) is a key aspect to address. Carbon capture, utilisation and storage (CCUS) technologies are promising options to further use the fossil fuels without increasing the CO₂ emissions [2]. The development of combined biomass-based low carbon energy conversion technologies

is significantly important considering the need to reduce both the fossil fuel dependence and the fossil CO₂ emissions.

Chemical looping is a promising energy conversion method to deliver both reduced energy and cost penalties for CO₂ capture [3]. One of the main advantages of chemical looping conversion lies in the fact that a solid oxygen carrier is used to oxidise the fuel subsequently reducing the nitrogen contamination of captured CO₂ stream compared with the case when air is used for fuel combustion. In this way CO₂ is inherently separated from the flue gas and no significant energy duty is required for the gas separation [4]. Another key aspect represents the process conditions; the looping reactors are running at elevated temperatures (500–1000 °C) which enhance the high temperature heat recovery potential [5]. In contrast, gas–liquid applications for CO₂ capture are running at around atmospheric temperatures (30–60 °C) which will end up in low temperature heat recovery with a negative effect on overall plant energy efficiency [6]. In addition, the chemical solvents (e.g. alkanolamines) require significant heat duty (around 3 MJ/kg captured CO₂) for regeneration. On the other hand, it has to be recognised that gas–liquid absorption technologies for CO₂ capture are far more technological and commercial mature than chemical looping or other emerging carbon capture technologies [7].

The current state of the art in chemical looping development is considering mostly the usage of gaseous fuels (not only mainly natural gas but also syngas) for heat and power generation [8]. The utilisation

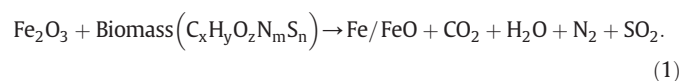
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of solid fuels in chemical looping conversion is complicated by various operational aspects such as ash removal together with some oxygen carrier, longer residence time for high fuel conversion, and oxygen carrier deactivation [9,10]. But the perspectives on solid fuel conversion in chemical looping systems are particularly appealing in delivering high energy efficiencies coupled with an almost total fuel decarbonisation rate. The carbon capture rate is more important for high CO₂ emission fuels such as solid fuels than for natural gas. From an operational point of view, the biomass conversion in chemical looping systems looks favourable due to lower ash content and higher reactivity than for coal [11].

This work evaluates the techno-economic performances of biomass direct chemical looping (BDCL) conversion for hydrogen and power co-production [12]. The BDCL concept was presented using ilmenite (FeTiO₃) as oxygen carrier [9]. As evaluated plant size, a net power output of about 400–500 MW was considered with a flexible hydrogen output in the range of 0 to 200 MW_{th} (LHV). Computational methods (using ChemCAD software) were used to assess the BDCL and the benchmark power plant concepts. The following IGCC-based benchmark cases (with and without carbon capture) were considered: IGCC without carbon capture [13]; IGCC with pre-combustion carbon capture using Selexol® (physical gas–liquid absorption) and IGCC with pre-combustion carbon capture using syngas-based chemical looping design [14]. All IGCC-based cases were considering coal and biomass co-processing using a dry fed gas quench entrained flow gasifier (Shell gasifier). The reason to use coal and biomass co-processing for the benchmark cases is the fact that currently there are no industrial size gasifiers able to process only biomass. For these gasifiers, the biomass ratio can be up to 30% from total fuel input.

2. Biomass direct chemical looping (BDCL) concept

Direct solid fuel conversion via chemical looping concept can be done for power generation only or for hydrogen and power co-generation [15]. The first option implies two gas–solid reactors; in the first reactor (fuel reactor) the fuel is totally oxidised to CO₂ and water with an oxygen carrier (metallic oxides such as Fe, Ni, Mn, and Cu). The gas leaving the fuel reactor is then cooled down and after condense removal, the captured CO₂ stream is conditioned (dry and compress) for transport and storage. The chemical reaction that occurs in the fuel reactor is presented below (considering iron oxide as oxygen carrier):



The contacting method between gas and solid phases in the fuel reactor is counter-current moving bed. This reactor design assures almost full conversion (oxidation) of the fuel and the volatiles [9]. The reduced form of the oxygen carrier is sent to a separate reactor (air reactor) where the oxygen carrier is oxidised back with air and recycled to the fuel reactor. The reoxidation reaction is highly exothermic and it is presented below:



When hydrogen and power co-production is targeted, the BDCL system implies three interconnected gas–solid reactors [16]. The fuel reactor is the same as for the power only case. In the second reactor (steam reactor), the reduced form of the oxygen carrier is partially reoxidised using steam (mild exothermic process) as follows:



In the third reactor (air reactor), the oxygen carrier is fully reoxidised with air and then recycled to the fuel reactor. The reoxidation process is highly exothermic, the solid flow recycled to the fuel reactor being used to balance the heat within the scheme (fuel oxidation is

highly endothermic). The chemical reaction which occurs in the air reactor is the following:



As the fuel used in the BDCL concept was presented in this work, sawdust was considered. The sawdust characteristics (low ash content) and the different operating conditions (e.g. temperature) minimise the alkali corrosion problems in comparison to the biomass-based combustion systems. The conceptual scheme of sawdust-fuelled biomass direct chemical looping (noted Case 1) for hydrogen and power co-generation is presented in Fig. 1.

To compare the main techno-economic and environmental performances of BDCL concept, the following benchmark cases were considered: (i) Case 2: IGCC power plant without carbon capture; (ii) Case 3: IGCC power plant with pre-combustion carbon capture using Selexol® (physical gas–liquid absorption) and (iii) Case 4: IGCC with pre-combustion carbon capture using syngas-based chemical looping design. The benchmark Case 2 (IGCC without carbon capture) is a standard gasification-based power plant without carbon capture [17]. The benchmark Cases 2–4 are only used to compare the performances of BDCL concept (Case 1). For instance, Case 2 (IGCC without CCS) is used to quantify the energy and cost penalties imposed by carbon capture step. Cases 3 and 4 are used just for comparison reason to put into perspective the performances of Case 1 in comparison to other carbon capture options (gas–liquid absorption for Case 3 and syngas-based chemical looping for Case 4).

The conceptual layouts of benchmark Case 3 (IGCC with pre-combustion capture using Selexol®) and benchmark Case 4 (IGCC with pre-combustion capture using syngas-based chemical looping) are presented in Figs. 2 and 3.

All benchmark cases are suitable for hydrogen and power co-generation and they are using a Shell gasifier (dry fed gas quench entrained flow type) co-processing a mixture of coal and biomass (sawdust) in the ratio of 80:20 (wt.). The reason for selecting coal and biomass co-processing is that the industrial size gasifiers are designed for coal processing only but they are tolerating a limited amount of biomass in the fed (up to 20–30 wt.%). Considering the size of evaluated power plant concepts (hundreds of MW scale), the idea was to use an already proven coal gasification technology. A similar size gasifier able to process 100% biomass is not available yet on the market [17]. The biomass (sawdust) feedstock is dried to 10 wt.% moisture prior to processing.

The captured CO₂ has to comply with strict quality specifications due to transport and storage requirements [18]. The following quality specification (expressed in vol.%) was considered in the present analysis: >95% CO₂; <2000 ppm CO; <250 ppm H₂O; <100 ppm H₂S and <4% all non-condensable gases (H₂, N₂, Ar, etc.). The hydrogen output has purity higher than 99.95% (vol.) to be suitable for chemical and energy applications.

3. Assessment of key technical and environmental performances

The following power plant designs were assessed in this paper:

- Case 1: Biomass (sawdust) direct chemical looping;
- Case 2: IGCC power plant without carbon capture using coal and sawdust as fuel;
- Case 3: IGCC power plant with pre-combustion capture based on gas–liquid absorption (Selexol®) using coal and sawdust as fuel;
- Case 4: IGCC power plant with syngas-based chemical looping using coal and sawdust as fuel.

As an illustrative example, the main design assumptions of biomass direct chemical looping power plant (Case 1) are presented in Table 1 [16]. The biomass (sawdust) drying is based on innovative energy-

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