



Research Paper

Chemical Looping with Oxygen Uncoupling (CLOU) concepts for high energy efficient power generation with near total fuel decarbonisation



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HIGHLIGHTS

- Chemical Looping with Oxygen Uncoupling (CLOU) for solid and gaseous fuels.
- High energy efficient and near-zero CO₂ emission solutions based on CLOU concept.
- Energy integration analysis for CLOU-based power plant with carbon capture.
- Integrated techno-economic and environmental assessment of CLOU technology.

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ABSTRACT

Developing high energy efficient solutions with low CO₂ emissions for heat and power generation sector is of main importance today. One innovative carbon capture method based on Chemical Looping with Oxygen Uncoupling (CLOU) concept for power generation with near-zero CO₂ emissions was evaluated in this paper. As an illustrative example, the copper-based oxygen carrier was considered for both fossil and renewable fuels conversion. The sizes of evaluated power plant concepts are 100 MW net power output. The carbon capture rate of the CLOU systems is almost total (>99%). The paper evaluates the conceptual designs of CLOU systems focusing on the assessment of mass and energy integration aspects and quantification of key techno-economic and environmental performances. The evaluated designs were simulated, the results being used to assess the overall indicators. For comparison reason, similar power plants without carbon capture and with carbon capture by gas-liquid absorption or oxy-combustion were considered as benchmarks. The assessments show that CLOU system has significant advantages (higher efficiency, lower CO₂ emissions and plant complexity, better economic performances, etc.) compared to the benchmark cases without capture but also to the carbon capture cases by gas-liquid absorption or oxy-combustion.

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1. Introduction

The continuous usage of fossil fuels and the correspondent greenhouse gas emissions are matters of great importance in the fight against climate change [1]. Accordingly, the heat and power sector as well as the other energy-intensive industrial applications need innovative solutions with high energy conversion efficiency and low CO₂ emissions [2]. Two important research and development directions are to be exploited here: one is relating to the increase of energy efficiency and the other is referred to reducing the CO₂ emissions. Both these targets are already emphasised in key EU energy and climate policies e.g. cutting the greenhouse gas emissions by at least 40% until 2030 (compared to 1990 levels)

and reducing energy consumptions by at least 27% of projected 2030 levels by improving energy efficiency [3].

Carbon capture and storage (CCS) technologies are expected to play a significant role in the attempt to reduce the global greenhouse gas emissions (at least 20% by 2050) [4]. The most technological and commercial mature carbon capture option is based on post-combustion method using chemical gas-liquid absorption [5]. The negative aspect of gas-liquid absorption technology is the high thermal duty for solvent regeneration which implies an overall energy penalty for CO₂ capture of at least 10 net electricity percentage points [6]. As consequence, the innovative energy efficient carbon capture options need to be developed for fossil fuel-intensive processes.

High temperature solid looping technology is a promising method to deliver both high energy efficiency and low CO₂ emissions. One of the key advantages of thermo-chemical looping

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cycles conversion lays in the fact that a solid oxygen carrier (metallic oxides of Fe, Ni, Cu, Co, Mn, etc.) is used to oxidize the fuel subsequently reducing the nitrogen contamination of captured CO₂ compared to the conventional case when air is used for fuel combustion [7]. Thermal integration of chemical looping reactors which are operated at high temperature is also very promising to further increase the overall plant energy efficiency [8].

A particular attractive looping system is based on Chemical Looping with Oxygen Uncoupling (CLOU) concept in which the oxygen carrier releases gaseous oxygen allowing the fuel to burn efficiently with gas phase oxygen [9]. This concept overcomes some drawbacks of Chemical Looping Combustion (CLC) technology in which a slow gasification step is happening in the fuel reactor to produce char (in case that a solid fuel is used) [10]. Copper, manganese and cobalt-based materials are being identified as promising oxygen-carriers for CLOU systems [11]. Fig. 1 presents the conceptual layout of chemical looping with oxygen uncoupling system for power generation.

The chemical reactions which take place in the CLOU system are the followings:

– Fuel reactor:



– Air reactor:

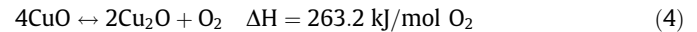


This article evaluates the copper-based CLOU system for power generation with almost total fuel decarbonisation. Various gas and solid fuels (both fossil and renewable) were considered for production of 100 MW net power with an almost total carbon capture rate (>99%). The evaluated CLOU designs were simulated using process flow modelling, the results being used to assess the mass & energy integration aspects as well as the overall techno-economic and environmental indicators (e.g. energy efficiency, ancillary consumptions, carbon capture rate, specific CO₂ emissions, cooling water consumption, specific investment cost, operation & maintenance cost, levelised cost of electricity, CO₂ capture cost, etc.). As benchmark cases, similar power plants without carbon capture and with carbon capture by chemical gas-liquid absorption or oxy-combustion were considered for comparison. The key novelty

and innovation aspects of this work are dealing with an integrated techno-economic and environmental assessment of the evaluated technology which underlines the key promising performances of CLOU method. As an illustrative example of novelty of this work compared to the present state of the art, an economic evaluation of CLOU technology was presented to emphasize the key advantages of this method and to close a knowledge gap identified in the literature.

2. Description of copper-based CLOU system for power generation

The copper-based system for CLOU concept is based on the following reaction [12]:



The CuO decomposition reaction to produce oxygen takes place in the fuel reactor. The released oxygen is then used for the fuel total oxidation (combustion). The endothermic duty of the CuO decomposition is covered by the exothermic duty of the fuel oxidation reaction. The reduced form of the oxygen carrier (Cu₂O) is then transferred into the air reactor where the reverse reaction takes place (to oxidise Cu₂O back to CuO). The two CLOU reactors are operated in a Circulated Fluidised Bed (CFB) mode.

The operating temperature has an important influence on the oxygen release reaction based on copper-based system [13]. The CuO decomposition reaction is favoured by high temperatures as can be observed from Fig. 2.

The oxygen equilibrium concentration as function of temperature is characterised by the following mathematical correlation [14]:

$$\text{C}_{\text{O}_2\text{eq}} = \frac{101325}{RT} * \exp(22 - 2.993 * 10^4 * T^{-1} - 1.048 * 10^6 * T^{-2}) \quad (5)$$

The oxygen equilibrium concentration represents a thermodynamic constraint of this system. In the fuel reactor, the oxygen concentration should be lower than the equilibrium one to favour the oxygen release for the fuel oxidation (combustion). In the air reactor, the oxygen concentration should be higher than the equilibrium one (up to the oxygen concentration in the inlet air) to favour the CuO formation.

Beside thermodynamic considerations, the kinetic factors are also important. For CuO reduction (in the fuel reactor), two reaction rate equations were proposed as follows [15]:

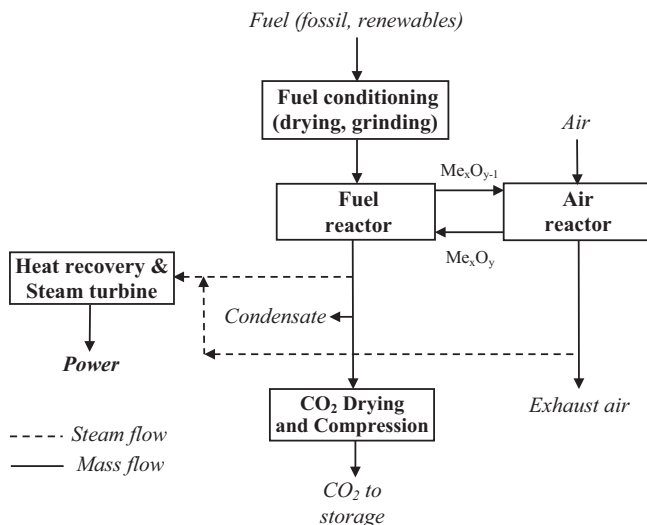


Fig. 1. Layout of CLOU system for power generation with carbon capture.

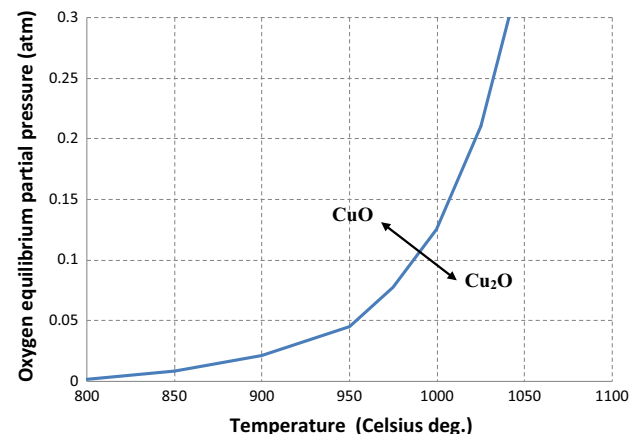


Fig. 2. Variation of oxygen equilibrium partial pressure vs. temperature in copper-based CLOU system.

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