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Techno-economic and exergetic assessment of an oxy-fuel power plant fueled by syngas produced by chemical looping $CO₂$ and $H₂O$ dissociation

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ABSTRACT

Natural Gas Combined Cycle (NGCC) is presently the most efficient fossil fuel power plant but with no carbon capture. The efficiency penalty resulting from the integration of carbon capture and storage (CCS) is, however, a major challenge. The present study proposes an oxyfuel NGCC integrated with Chemical looping (CL) syngas production (OXY-CC-CL), for power generation with CCS. The chemical looping CO_2/H_2O dissociation would produce syngas (CO and $H₂$ with methane reduction step in redox cycle) from recycled exhaust gas for additional power generation within the power plant. This integration of CL unit with the existing conventional oxy fuel power plant would be expected to decrease the efficiency penalty. Therefore, the thermodynamic (both energetic and exergetic), economic and environmental performance of the integrated chemical looping unit oxyfuel NGCC power plant with carbon capture were assessed. A 500 MW scale plant was modelled and compared with a conventional NGCC and oxyfuel NGCC plant with carbon capture (OXY-CC). The net efficiency penalty of the proposed OXY-CC-CL unit was 4.2% compared to an efficiency penalty of 11.8% of the OXY-CC unit with a 100% carbon capture. The energetic efficiency obtained hence was 50.7%, together with an exergetic efficiency of 47.1%. Heat integration via pinch analysis revealed the possibility to increase the system energetic efficiency up to 61%. Sensitivity analyses were performed to identify relative impacts of system operational parameters. The specific capital cost of the proposed OXY-CC-CL was obtained as 2455 \$/kW, with a corresponding LCOE of 128 \$/MWh without carbon credits.

1. Introduction

Burning fossil fuels, resulting in anthropogenic carbon dioxide $(CO₂)$ emissions are presently recognised as the primary contributor to climate change, with 36.2 Gt being emitted in 2016 [\[1](#page--1-0)–3]. Notwithstanding substantial investment and decline in prices of renewable energy, fossil fuels continue to play an indispensable role in the World's energy landscape [[4](#page--1-1)]. Indeed, even though the trend is on a decline, such technologies continue to play a major role as the primary energy source, especially in developing countries [[5](#page--1-2)]. Hence, it is expected that the relevance of fossil fuels in the primary energy mix will continue to a significant extent in the considerable future.

Carbon capture and storage (CCS) technologies have been shown to have considerable potential to reduce such anthropogenic $CO₂$ emissions as part of the global transition towards a low carbon energy

system [[2](#page--1-3)[,6,](#page--1-4)[7\]](#page--1-5). These technologies are typically categorized into three categories: pre-combustion, post-combustion, and oxy-combustion [[8](#page--1-6),[9](#page--1-7)], the common idea being $CO₂$ capture and subsequent storage. The $CO₂$ is derived from the complete or partial combustion of fossil fuels generated either in power plants or in respective industrial sectors. The captured CO₂, nevertheless, needs to be subsequently compressed to approximately 110 bar prior to transportation via pipeline to a storage site $[2,10,11]$ $[2,10,11]$ $[2,10,11]$ $[2,10,11]$ $[2,10,11]$. However, in recent studies, recycle and re-use of the captured CO₂ via innovative methods such as chemical formation for subsequent use [\[12](#page--1-10)–14], has also received much attention as an alternative to storage.

The oxy-fuel combustion is currently one of the most promising alternatives among the portfolio of all the low-emission technologies (LETs) [\[15](#page--1-11),[16\]](#page--1-12). In this technology, the fuel (coal or natural gas or biomethane) is burnt in an oxygen (O_2) rich environment (near

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stoichiometric O_2 flows), instead of air, thereby improving combustion efficiency [\[17](#page--1-13)] and eliminating NO_x emissions and generating only $CO₂$ and $H₂O$ as the product of the combustion unit. The oxygen is supplied via an air separation unit (ASU). Burning fuels under these conditions generate combustion gases, which, after condensation yields a very high purity of $CO₂$ exhaust. Oxy-combustion can also be applied to natural gas combined cycle (NGCC), however, subject to the redesign of gas turbines. This is due to the alternation in the physical properties of the metal occurs from an increased $CO₂$ concentration in the flue gas [[11](#page--1-9)[,17](#page--1-13)]. Nevertheless, ease and ability to retrofit existing systems at low cost are the primary attractions [\[16](#page--1-12)], together with the high efficiency of 96–99% carbon capture [\[18](#page--1-14)] of such systems.

Similar to LETs, technical challenges exist for the oxy-fuel combustion process. The most critical limitations lie in the higher energy penalties associated with air separation unit (ASU) for O_2 production and $CO₂$ processing unit (CPU) for $CO₂$ purification and compression [[2](#page--1-3),[19,](#page--1-15)[20\]](#page--1-16) after the combustor unit. The existing commercialized technology for air separation for utility-scale application is the cryogenic air separation process (CASU). It works on the principle of the cryogenic distillation via compression of air to its liquefaction stage, followed by the fractional distillation of its constituent components, such as N_2 , O_2 , Ar and other rare gases. The primary advantage is that this process can produce liquid or gaseous streams of N_2 and O_2 as per the specification of the end user and for large-scale requirements also. Indeed, $O₂$ production, via such a process of cryogenic distillation of air, demanding 160 to 250 kW h per tonne of O_2 produced [[21](#page--1-17),[22\]](#page--1-18) is acknowledged as the bottleneck [[2](#page--1-3),[16\]](#page--1-12). State of the art of ASU can consume between 10 and 40% of the gross power output after retrofitting a conventional coal-fired power, resulting in a net energy penalty as high as 8–13 percentage points [[7](#page--1-5)[,23](#page--1-19)]. The efficiency penalty from integrating the ASU to a conventional NGCC unit without carbon capture for oxy-fuel combustion could be as much as 13% [\[24](#page--1-20)]. The penalties incurred by the use of the ASU would therefore offset any advantages gained by oxyfuel combustion. This has prompted many researchers to investigate the use of alternative air separation systems. However, to date, none of the alternative technologies for air separation have been able to produce high purity oxygen at large utility scale, either due to high costs, such as for adsorption processes, or the technology is still under development or in demonstration stage, as for membrane technologies such as oxygen transport membranes [[25,](#page--1-21)[26\]](#page--1-22). True, with a lower purity of $O₂$ of about 95%, if acceptable for such oxy-fuel applications, the energy requirement for oxygen production with ASU can be further reduced, together with the energy penalty [[16\]](#page--1-12).

Several studies have alternately proposed to increase the efficiency for carbon capture. Improving the efficiency through a novel chemical looping air separation technology has been proposed by Moghtaderi [[22\]](#page--1-18). From a system perspective, chemical looping combustion has been shown to have considerable potential for a relatively high efficiency of power production together with carbon capture. For a pulverized coal power plant, around 39% efficiency was calculated while ensuring a $CO₂$ capture efficiency of almost a 100% [\[27](#page--1-23)]. Innovative system integration has shown the further possibility to decrease the energy penalty of carbon capture.

Natural gas combined cycle (NGCC) power plants are among the most efficient fossil fuel-fired power plants, able to reach net efficiencies of up to 57% based on the Lower Heating Value (LHV) of the fuel [\[28](#page--1-24)]. Correspondingly, the specific $CO₂$ emissions are low as well compared to other fossil fuel power generation units, at around 350 $gCO₂/kWh$, besides having much less SO_x and NO_x emissions due to the lower sulphur and nitrogen content of the fuel [\[28](#page--1-24)]. Addition of CCS units to considerably decrease the specific $CO₂$ emissions to much below 100 gCO₂/kWh have therefore been studied and presented in multiple literature via diverse technologies [28–[30\]](#page--1-24). Like solid fuel power units, the primary motivation of such studies included the decrease in the energy penalty of the capture process, thereby increasing the efficiency of the power plant alongside keeping the capture

Fig. 1. Conceptual scheme of the chemical looping syngas production through methane reduction and corresponding splitting of water and carbon dioxide, usually present in waste gas from industrial applications.

efficiency to its maximum potential.

Chemical looping for thermo-chemical dissociation of the captured $CO₂$ has received considerable interest [\[31](#page--1-25)–37] in the last couple of years after the initial success of the thermochemical water splitting [[38\]](#page--1-26). In fact, the simultaneous splitting of $CO₂$ and $H₂O$, resulting in the formation of a syngas has great potential to improve the system efficiency by providing additional fuel with high calorific value leading to $CO₂$ recycling. Chemical looping $CO₂/H₂O$ splitting, like chemical looping combustion (CLC) cycles, uses metallic oxygen carriers to convert the thermo-chemical energy into chemical energy [\[39](#page--1-27)]. The cycle can either be driven by concentrated solar energy or through the chemical energy of methane. The later, besides having multiple benefits, also lowers the temperature of thermal reduction as compared to direct thermal reduction [[40,](#page--1-28)[41\]](#page--1-29). A basic schematic of the methane driven Chemical Looping $CO₂/H₂O$ splitting is shown in [Fig. 1.](#page-1-0)

In the reduction step (Eq. 1), the metal oxide is reduced in the presence of methane, either up to stoichiometric or a non-stoichiometric extent, δ . The δ moles of oxygen released from the MeO_x forms CO and H_2 by partial oxidation of CH₄. In the subsequent reaction steps, usually exothermic, (eq. 2), MeO_{x-δ}reacts with CO_2 and/or H₂O to reincorporate the oxygen back into the metal oxide lattice, while reducing the CO_2 and/or H_2O into a stream of syngas (CO or H_2 respectively). Reactions (2a) and (2b) can be intrinsically assumed to result in complete oxidation at thermodynamically favourable temperatures depending on the metal oxide redox pairs. The oxygen production during the first step depends on the reduction extent and the highest valence state of the metal cation to be reduced. Highest possible dissociations are sought in principle; the higher the oxygen released during dissociation step, the higher the oxygen taken from $CO₂$ and $H₂O$ during oxidation.

Methane – reduction: $MeO_x + \delta CH_4 \rightarrow MeO_{x-\delta} + \delta (CO + 2H_2)$ (1)

 $Water - splitting (WS): MeO_{x-\delta} + \delta H₂O \rightarrow MeO_x + \delta H₂$ (2a)

$$
CO_2 - splitting (CDS): MeO_{x-\delta} + \delta CO_2 \rightarrow MeO_x + \delta CO \tag{2b}
$$

In this regard, multiple studies have been reported in the literature to identify the most suitable metal oxide redox pairs [41–[43\]](#page--1-29) and reactor scale experimental analyses for reactor modelling [[39](#page--1-27)[,41](#page--1-29)]. These include volatile metal oxide/metal system such as ZnO/Zn; SnO/Sn or non-volatile metal oxide/metal oxide pairs such as $Fe₃O₄/FeO$; $Mn₃O₄/$ MnO; $CeO₂/Ce₂O₃$, etc. Other metals oxides tested are ferrites with different valences, $Co₃O₄$, $Nb₂O₅$, $WO₃$, $SiO₂$, $In₂O₃$, CdO to name few

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