



## Full Length Article

# Integration of chemical looping oxygen production and chemical looping combustion in integrated gasification combined cycles

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## ABSTRACT

Energy penalty is the primary economic challenge facing CO<sub>2</sub> capture technology. This work aims to address this challenge through a novel power plant configuration, capable of achieving 45.4% electric efficiency from coal with a 95% CO<sub>2</sub> capture efficiency. The COMPOSITE concept integrates chemical looping oxygen production (CLOP) and packed bed chemical looping combustion (PBCLC) reactors into an integrated gasification combined cycle (IGCC) power plant. Hot gas clean-up technology is implemented to boost plant efficiency. When commercially available cold gas clean-up technology is used, the plant efficiency reduces by 2%-points, but remains 2.3%-points higher than a comparative PBCLC-IGCC power plant and 8.1%-points higher than an IGCC power plant with pre-combustion CO<sub>2</sub> capture. It was also shown that the COMPOSITE power plant performance was not sensitive to changes in the performance of the CLOP reactors, implying that uncertainties related to this novel process component do not reduce the potential of the COMPOSITE concept. The outstanding efficiency obtained for this concept is made possible by a complex and highly integrated plant configuration, whose operability and techno-economic feasibility must be demonstrated.

## 1. Introduction

Energy penalty is the primary economic challenge facing CO<sub>2</sub> capture processes. The energy requirements of CO<sub>2</sub> capture not only increase fuel consumption, but also increase plant capital costs (a larger plant is required to produce a given amount of power) as well as the amount of CO<sub>2</sub> that needs to be captured, transported and stored. According to a recent review of the costs of CCS [1], a typical pulverized coal (PC) plant with post-combustion CO<sub>2</sub> capture will require about 32% more energy per unit electricity production than an equivalent plant without CO<sub>2</sub> capture. This is a major contributing factor to the ~62% increase in the levelized cost of electricity.

For this reason, energy efficiency has been the highest CO<sub>2</sub> capture research priority. Several second-generation CO<sub>2</sub> capture processes have been proposed with the primary aim of reducing energy penalty. Chemical looping technologies offer the most fundamental potential for achieving this goal because inherent separation between CO<sub>2</sub> and N<sub>2</sub> is achieved with almost no associated energy cost.

Chemical looping combustion (CLC) [2] is the most studied chemical looping configuration. It operates by transporting oxygen from air to fuel using an oxide oxygen carrier material (OCM). Air and fuel are

fed to two separate reactors where the OCM is oxidized by air, transported to the fuel reactor, reduced by the fuel, and then transported back to the air reactor. This way, CLC achieves oxyfuel CO<sub>2</sub> capture without the large energy penalty associated with air separation.

When applied to solid fuels, CLC can be implemented in two distinctly different configurations. Firstly, integrated gasification CLC (iG-CLC) feeds the solid fuel directly into the fuel reactor where it gasifies and reduces the oxygen carrier. A recent study estimated that iG-CLC can capture CO<sub>2</sub> for only €20/ton relative to a coal plant using a circulating fluidized bed (CFB) boiler [3]. The second alternative is integration of conventional gas-fuelled CLC into an IGCC power plant. This CO<sub>2</sub> capture pathway produces a similar cost increase (€23/ton) relative to an unabated IGCC plant [4].

Both these technology pathways have advantages and drawbacks. The iG-CLC pathway can capitalize on know-how from commercial deployment of CFB boilers. Even though CFB boilers are designed primarily for low-rank coal and have only recently been demonstrated at scale in efficient supercritical configurations, this similarity should be beneficial during the iG-CLC scale-up process. The capital costs of a CFB boiler is generally higher than that of a conventional pulverized coal (PC) boiler, but this capital cost drawback can be recovered by not

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**List of symbols***Regular symbols*

$\alpha$	Volume fraction
$\varepsilon$	Void fraction
$\phi$	Thiele modulus
$\eta$	Effectiveness factor
$\rho$	Density (kg/m <sup>3</sup> )
$\vec{v}$	Velocity vector (m/s)
$\tau$	Tortuosity
$\bar{\tau}$	Stress tensor (kg/s <sup>2</sup> m)
$\xi$	Normalized radius
$C$	Molar concentration (mol/m <sup>3</sup> )
$D$	Diffusivity (m <sup>2</sup> /s)
$d$	Diameter (m)
$\vec{g}$	Gravity vector (m/s <sup>2</sup> )
$h$	Enthalpy (J/kg)
$\dot{H}$	LHV flow rate (MW)
$\vec{J}$	Diffusive mass flux (kg/m <sup>2</sup> s)
$K$	Equilibrium constant
$K_{sg}$	Interphase exchange coefficient (kg/m <sup>3</sup> s)
$k$	Reaction rate constant ((m/s) (mol/m <sup>3</sup> ) <sup>1-n</sup> )
$M$	Molecular weight (kg/mol)
$\dot{m}$	Mass transfer rate (kg/m <sup>3</sup> s)
$\dot{M}$	Molar flow rate (kmol/s)
$N$	Moles (mol)
$n$	Reaction order
$P$	Pressure (bar)
$p$	Pressure (Pa)
$Q$	Interphase heat exchange (J/m <sup>3</sup> s)
$\vec{q}$	Diffusive energy flux (J/m <sup>2</sup> s)
$R$	Universal gas constant (8.314 J/mol.K)
$R^H$	Heterogeneous reaction rate (mol/m <sup>3</sup> s)
$S$	Mass source term (kg/m <sup>3</sup> s)
$S^{\vec{v}}$	Momentum source term (kg/m <sup>2</sup> s <sup>2</sup> )
$S^h$	Energy source term (J/m <sup>3</sup> s)
$s$	Active surface area (fraction)
$T$	Temperature (K)
$V$	Volume (m <sup>3</sup> )
$w$	Degree of solids conversion (fraction)

$x$	Mole fraction
$Y$	Mass fraction

*Subscripts*

$c$	Active core
$eff$	Effective
$eq$	Equilibrium
$g$	Gas
$gr$	Grain
$i$	Species index
$p$	Particle
$ox$	Oxidation
$pq$	Interphase exchange
$q$	Phase index
$red$	Reduction
$s$	Solids

*Acronyms*

ASU	Air separation unit
CGE	Cold gas efficiency
CLC	Chemical Looping Combustion
CLOP	Chemical Looping Oxygen Production
CLOU	Chemical Looping with Oxygen Uncoupling
CGCU	Cold gas clean-up
HGCU	Hot gas clean-up
HHV	Higher heating value
HP	High pressure
HRSG	Heat recovery steam generator
HT	High temperature
HTW	High temperature Winkler
HV	Heating value
IP	Intermediate pressure
IGCC	Integrated Gasification Combined Cycle
LHV	lower heating value
MP	Medium pressure
OCM	Oxygen carrier material
PBCLC	Packed Bed Chemical Looping Combustion
TOT	Turbine outlet temperature

having to include downstream flue gas scrubbers [5]. However, increasingly strict emissions standards may require flue gas treatment even from CFB plants [5].

IGCC plants are more capital-intensive than PC plants and there are only a few operating plants globally. However, the IGCC configuration is inherently capable of higher efficiencies and lower emissions than PC boilers. It therefore remains a relevant prospect for solid fuel combustion in an increasingly carbon-constrained world with strict emissions standards. IGCC also has significant headroom for future cost reductions through hot gas cleanup and advanced gas turbine technology. By the year 2030, the latest version of the International Energy Agency's electricity cost projections [6] gives similar costs for IGCC (60–88 \$/MWh) and advanced ultra-supercritical PC (58–82 \$/MWh) plants. The IGCC-based process proposed in this paper can become a commercial reality by the year 2030 and beyond when IGCC should be more competitive.

Regarding the CLC units in the two configurations, the primary technical challenges are in-situ gasification in the iG-CLC configuration and pressurized operation in the IGCC configuration. The iG-CLC technology poses challenges related to fuel slip from syngas produced near the top of the fuel reactor, the need for a carbon stripper unit to

prevent char from leaking to the air reactor, and the demand for a very cheap oxygen carrier that can have a short active lifetime due to ash exposure or losses with ash removal [7,8]. For IGCC, pressurized operation greatly increases the required solids circulation rate per unit reactor volume and requires special measures to carry the pressure load on all pressurized components. Technical challenges are also presented by the need for high-temperature filtration of fines that can damage the downstream gas turbine.

In this work, the IGCC pathway is studied. The starting point is based on an earlier work with integrated packed bed CLC (PBCLC) for highly efficient CO<sub>2</sub> separation [9,10]. The PBCLC configuration keeps the oxygen carrier in a single reactor where it is alternatively exposed to air and fuel gases. This simple standalone reactor configuration should be simpler to scale up than the conventional dual fluidized bed CLC configuration, especially under pressurized operation. We propose an extension of this PBCLC-IGCC power plant configuration to further boost the already attractive efficiency by replacing the air separation unit (ASU) with a chemical looping oxygen production (CLOP) unit. A more detailed description of this novel process is given in the next section.

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