

# The mOxy-CaL Process: Integration of Membrane Separation, Partial Oxy-combustion and Calcium Looping for CO<sub>2</sub> Capture

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CO<sub>2</sub> capture and storage (CCS) is considered as a key strategy in the short to medium term to mitigate global warming. The Calcium-Looping process, based on the reversible carbonation/calcination of CaO particles, is a promising technology for post-combustion CO<sub>2</sub> capture because of the low cost and non-toxicity of natural CaO precursors and the minor energy penalty on the power plant in comparison with amines capture based technologies (4-9 % compared to 8-12 %). Another interesting process to reduce CO<sub>2</sub> emissions in power plants is oxy-combustion, which is based on replacing the air used for combustion by a highly concentrated (~95 % v/v) O<sub>2</sub> stream. This work proposes a novel process (mOxy-CaL) for post-combustion CO<sub>2</sub> capture based on the integration of membrane separation, partial oxy-combustion and the Calcium-Looping process. An oxygen-enriched air stream, which is obtained from air separation by using highly permeable polymeric membranes, is used to carry out partial oxy-combustion. The flue gas exiting partial oxy-combustion shows a CO<sub>2</sub> concentration of ~30 % v/v (higher than 15 % v/v typical in coal power plants). After that, the flue gas is passed to the CaL process where the CO<sub>2</sub> reacts with CaO solids according to the carbonation reaction. Thermogravimetric analysis show that the multicycle CaO conversion is enhanced as the CO<sub>2</sub> concentration in the flue gas stream is increased. Process simulations show that the mOxy-CaL process has a high CO<sub>2</sub> capture efficiency (~95%) with lower energy consumption per kg of CO<sub>2</sub> avoided than previously proposed post-combustion CO<sub>2</sub> capture technologies. Moreover, the overall system size is significantly lower than state-of-the-art CaL systems, which allows for an important reduction in the capital cost of the technology.

## 1. Introduction

Despite the necessary increase in energy production share from renewable sources, power plants based on natural gas or coal currently generate most of the energy consumed in the world (IEA, 2016). Thus, CO<sub>2</sub> capture and storage (CCS) is a key technology to be developed in the short to medium term to mitigate global warming. Several processes have been proposed in the last years to be integrated in coal power plants (CFPP) for post-combustion CO<sub>2</sub> capture, such as the already commercial amine-based capture systems and the Calcium-Looping (CaL) process, which has been demonstrated at the MWth pilot scale level (Arias et al., 2013). The CaL process, based on the reversible carbonation/calcination reaction of CaO particles, show several advantages in comparison with amines-based technologies such as a minor energy penalty over the power plant (4-9 % compared to 8 – 12 %) as well as nontoxicity (Perejón et al., 2016). The main shortcoming of the CaL system is a marked sorbent deactivation with the number of cycles (Abanades, 2002), which requires an extra energy consumption. Moreover, larger equipment size is needed because of the massive presence of poorly reacting solids in the system causing high investment costs. Another interesting process to reduce CO<sub>2</sub> emissions in power plants is the oxy-combustion technology, which is based on replacing the air used for combustion by a highly concentrated (~95 % v/v) O<sub>2</sub> stream, leading to a highly-concentrated CO<sub>2</sub> flue gas

stream that can be stored or reused after a purification process. The main drawback of this technology is the high energy consumption required to obtain a high purity O<sub>2</sub> stream, which must be carried out by means of an air separation unit (ASU) via cryogenic distillation (Kather and Scheffknecht, 2009).

The present work proposes a novel technology for post-combustion CO<sub>2</sub> capture based on the integration of membrane separation, partial oxy-combustion and the Calcium-Looping process. In this integrated system, partial oxy-combustion of the fuel is carried out for electric power production using an oxygen-enriched gas (OEA) which has been previously produced through a highly permeable polymeric membrane system. Since an O<sub>2</sub> purity of ~20 – 40 % v/v is sufficient for partial oxy-combustion, air separation can be carried out by means of a polymeric membrane, which highly reduces the energy consumption associated with oxy-combustion. The flue gas exiting the combustor, which presents a CO<sub>2</sub> concentration ~30 % v/v, is sent to the CaL process where CO<sub>2</sub> reacts with the CaO solids according to the carbonation reaction. The higher concentration of CO<sub>2</sub> entering the CaL system compared to a typical air-combustion plant (~15 % v/v CO<sub>2</sub> in flue gas) enhances carbonation and therefore the CO<sub>2</sub> capture process (Ortiz et al. 2017). Simulation results show that the mOxy-CaL system has an energy consumption per kilogram of CO<sub>2</sub> avoided (SPECCA) of around 3 MJ/kgCO<sub>2</sub>, lower than ~4 MJ/kgCO<sub>2</sub> for the case of amine-based systems (Politecnico di Milano – Alstom UK (CAESAR project), 2011), whereas the CO<sub>2</sub> capture efficiency is increased above 95 %.

## 2. The mOxy-CaL process

This section aims at describing the mOxy-CaL process from the three technologies integrated within it: membrane-based air separation, partial oxy-combustion and Calcium-Looping.

### 2.1 Membranes technology to produce oxygen-enriched air

Membranes are used in a huge amount of industrial applications among which we find air separation (Chong et al. 2016). Polymeric membranes are today the most developed membranes for gas separation although other possibilities are gaining attention in the last years such as ion transport membranes. Polymeric membranes are generally non-porous and separate gases from the solution-diffusion transport mechanism from applying a transmembrane partial pressure difference. In a typical membrane-based gas separation process the feed gas passes is put into contact with the membrane surface under a certain pressure, which is the driving force of the process. The most permeable gas crosses the membrane and leaves the system as permeate stream (at lower pressure than the feed gas), whereas the remaining gas exits at high pressure as the retentate stream without crossing the membrane. Membranes for gas separation are usually analyzed in terms of permeability and selectivity, which highly influence the efficiency and cost of a membrane-based gas separation process. As pointed out by (Robeson, 1991), in polymeric membranes there is usually a trade-off between permeability and selectivity. The O<sub>2</sub>/N<sub>2</sub> separation factor for the best commercially available polymer membranes is 6–8 (Murali et al., 2013). In the same work, data can be found for permeability and selectivity of a large number of polymeric membranes.

The pressure ratio (PR) between the upstream and downstream membrane sides is a key operating parameter in the separation process. In order to reach a O<sub>2</sub> purity ≈ 40 % it would be enough a pressure ratio across the membrane of 0.1 by using a single-stage process for a membrane selectivity ≥ 3 (Matson et al., 1986). Nevertheless, it is remarkable that the higher pressure ratio the higher permeability of polymeric membranes (Robeson, 1991). Thus, in order to increase the permeability and therefore reduce the membrane area needed, a higher PR must be imposed. The energy consumption associated to a separation process can be calculated from the adiabatic compression work of a perfect gas, which gives a realistic estimation of energy consumption (Humphrey and Keller, 1997). The pressure ratio can be imposed either by increasing pressure on the upstream side (i.e. compressing the gas entering the membrane system) or by creating vacuum on the downstream side, which is the selected technique in the present work. Since a vacuum strategy is considered, the pressure must be increased again in the permeate (OEA) before entering the boiler. Thus, energy consumption ( $E_{sep}$ ) can be calculated as (Bounaceur et al., 2006):

$$E_{sep} = m_p \frac{\gamma RT}{\gamma - 1} \left[ \left( \left( \frac{P_{in}}{P_p} \right)^{\frac{\gamma-1}{\gamma}} - 1 \right) + \left( \left( \frac{P_b}{P_p} \right)^{\frac{\gamma-1}{\gamma}} - 1 \right) \right] \quad (1)$$

where  $m_p$  is the mass of gas permeating through the membrane,  $\gamma$  is the adiabatic expansion factor of the gas,  $T$  is the temperature of the system and  $P_{in}$ ,  $P_p$  and  $P_b$  are the pressures at the upstream side, downstream side and boiler inlet, respectively.

Membrane separation is simulated within the whole mOxy-CaL process from the energy consumption as given by Eq (1). Thus, by considering a gas pressure at the permeate side of 0.235 bar (similarly to previous works

such as (Bhide and Stern, 1991)), the energy requirement for separation is 90 kWh/t O<sub>2</sub>. A stage cut, defined as the ratio of permeate flowrate to feed flowrate of 0.1 is assumed (Belaïssaoui et al., 2014).

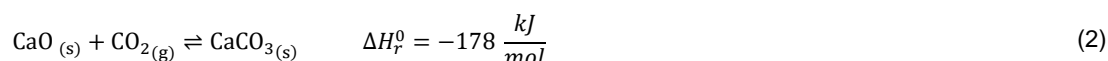
## 2.2 Partial Oxy-combustion

After passing across the membrane, the OEA stream is sent to the main boiler for partial oxy-combustion of coal. Partial oxy-combustion has been previously analysed to improve the boiler efficiency (Ariono and Wardani, 2017) or to enhance the performance of coal gasification (Belyaev et al., 2003). In this work, the concentration of O<sub>2</sub> entering the boiler has been chosen in order to achieve a CO<sub>2</sub> concentration of 30 %v/v in the flue gas. For practical proposes, it is assumed that the flue gas exiting the boiler has a concentration of 2.5 % to ensure a complete combustion. As for total oxy-combustion, a flue gas recirculation is needed to control the flame temperature inside the boiler.

A pulverized coal power plant with partial oxy-combustion is employed where coal is burned with the OEA stream in a boiler by a model based on chemical and phase equilibrium through Gibbs' free energy minimization methods (Ortiz et al. 2016). A compressor is considered to overcome the pressure drop in the flue gas recirculation stream. Power is produced using a reheat supercritical steam cycle ( $P_{vv} = 290$  bar,  $T_{vv} = 600/620$  °C). Flue gas exiting the steam generator is cooled and slightly compressed to reach the pressure needed at the carbonator inlet.

## 2.3 CaL process

The CaL process is performed using two interconnected circulating fluidized-bed (CFB) reactors, both operated under atmospheric pressure. CaO particles react in the carbonator reactor with the flue gas coming from the oxy-combustion boiler according to the carbonation reaction (Eq. (2)) at 650 °C. CO<sub>2</sub> capture in this reactor is modeled according to the model described elsewhere (Ortiz et al. 2015).



At CaL conditions involving short residence times in the carbonation reactor there is not a complete carbonation and an important amount of CaO remains unreacted (Perejón et al. 2016). CaO conversion (X) can be defined as the mass ratio between the reacted CaO to the total CaO entering the carbonator. As shown in previous works (Ortiz et al. 2017), CaO conversion is notably enhanced by increasing the CO<sub>2</sub> concentration in the flue gas entering the carbonator. The partially carbonated particles are then circulated into the calciner reactor in which fast decomposition of CaCO<sub>3</sub> occurs at high temperatures of ~950 °C to regenerate CaO and a rich CO<sub>2</sub> stream ready to be compressed and transported for storage or other uses. The regenerated CaO particles are recovered at the calciner exit by a cyclone and sent back to the carbonator for a new cycle. Given the large flow of solids recirculated between reactors in the CaL cycle, a heat exchanger (simplified by a heat transfer between solids with a temperature approach of 20 °C) is inserted to take advantage of sensible heat transfer between the solids leaving the calciner (with a temperature of about 950 °C) and the solids entering into it, which must be heated up to the calcination temperature (Martínez et al., 2012). The heat produced in the exothermic carbonation reaction as well as the sensible heat recovered by the streams exiting the calciner is used in a secondary steam cycle for electricity generation.

## 3. mOxy-CaL process simulation and results

The mOxy-CaL process has been modeled by means of ASPEN PLUS™. In addition to those previously mentioned, main model assumptions are: (i) the system operates at steady conditions; (ii) minimum temperature difference is 20 °C for every heat exchangers; (iii) 89 % isentropic efficiencies are considered as constant for all turbomachinery; (iv) ideal behavior of cyclones, (v) calcination is fully achieved in the calciner; (vi) Solids pneumatic conveying consumes 20 MJ/t; (vii) Energy consumption for total oxy-combustion in the calciner is 200 kWh/t O<sub>2</sub>, although this value could be as lower as 120-140 kWh/t O<sub>2</sub> (Escudero et al., 2016).

Results from the simulation are shown in Figure 1, where main stream data are detailed. Combustion of 42.2 kg/s of coal (Pittsburgh No. 8) with 281 kg/s of OEA (38 %v/v O<sub>2</sub>) releases 322.9 kg/s of flue gas with a CO<sub>2</sub> concentration of 30 % v/v. Flue gas recirculation with a recycle ratio of 0.63 is used to keep the flame temperature at 1,400 °C. Heat released in partial oxy-combustion and transferred to the steam cycle produces a net power of 513 MWe, which gives a 36.2 % of thermal to electric efficiency. Note that the energy consumption by using membranes is 90 kWh/t O<sub>2</sub> whereas in the case of cryogenic distillation is ~ 200 kWh/t O<sub>2</sub>. The flue gas reacts in the carbonator with a solids stream entering it at a mass flow rate of 2,535 kg/s (~94 % CaO while the rest is CaSO<sub>4</sub> and ash formed in previous cycles). This yields a ratio CaO/CO<sub>2</sub> entering the carbonator of 15. Multicycle CaO conversion has been extracted from published data (Ortiz et al., 2017). By considering a solids inventory in the carbonator of 400 t, the CO<sub>2</sub> capture efficiency is 95 %. Thus, an amount of 270 kg/s of

CaCO<sub>3</sub> is produced while 2,236 kg/s of CaO remains as unreacted. After carbonation, the flue gas containing ~2.5 %v/v O<sub>2</sub> is vented to the atmosphere after recovering its sensible heat by means of several gas-gas heat exchangers (Figure 1). In the calciner, oxy-combustion of 22.3 kg/s of coal with 59.5 kg/s of high purity O<sub>2</sub> is needed to carry out the calcination reaction. At the calciner exit, a mass flow rate of 203 kg/s of high concentrated CO<sub>2</sub> (90 %v/v) is purified and compressed up to 100 bar to be stored or reused. Taking advantage of the heat released during carbonation, 288 MWe are produced by means of a secondary steam cycle. An important amount of this power produced (~60 %) is used for several process and utilities along the CaL cycle, mainly for O<sub>2</sub> production (through cryogenic distillation) to be burned in the calciner, for compression of the captured CO<sub>2</sub> and for solids transport.

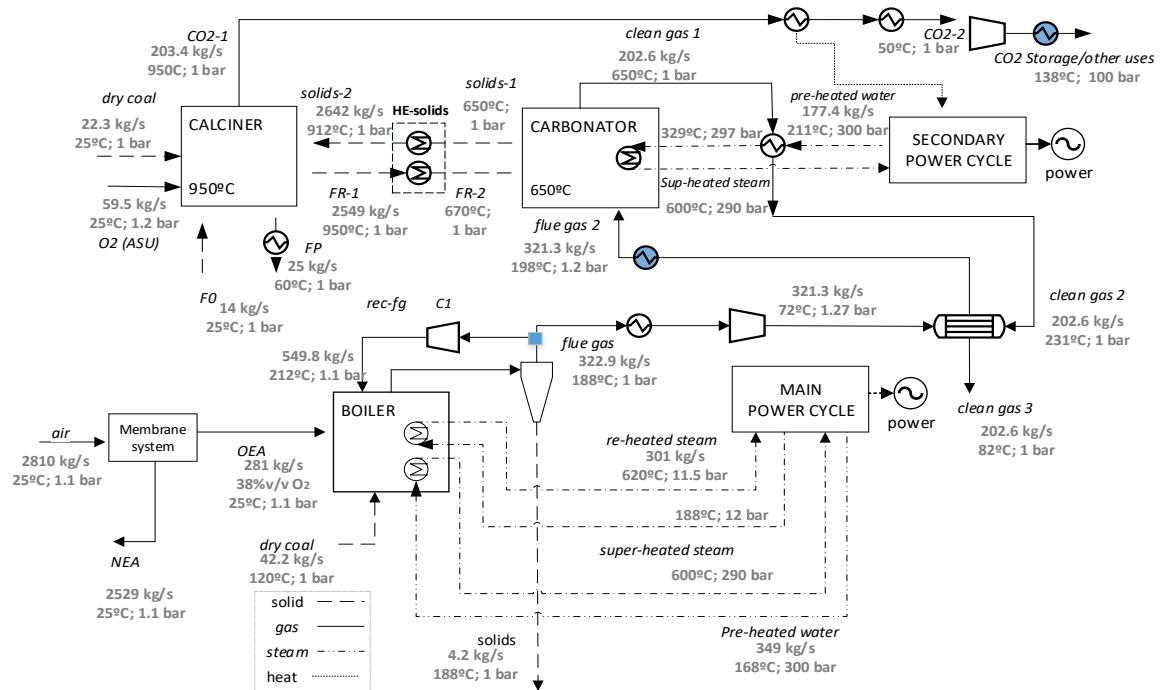


Figure 1: mOxy-CaL integration scheme and simulation results

CO<sub>2</sub> capture process in the CaL cycle leads to an energy penalty of 6.12 %, which, together with the penalty caused in the membrane/oxy-combustion process (1.63 %), yields an energy consumption (SPECCA) of 3.05 MJ/kg CO<sub>2</sub>. In order to analyze the mOxy-CaL performance, Table 1 shows a comparison between the mOxy-CaL process and previously proposed CO<sub>2</sub> capture systems (Ortiz et al., 2017). A reference coal fired power plant without CO<sub>2</sub> capture system is compared with an equivalent oxy-combustion plant and with a plant retrofitted with a CaL process for CO<sub>2</sub> capture. Moreover, the performance of an Oxy-CaL system (Ortiz et al., 2017) in which the OEA needed for the partial oxy-combustion is produced by using cryogenic distillation technology is also compared. As can be seen in Table 1, a notable reduction on energy consumption for CO<sub>2</sub> capture is achieved by means of the mOxy-CaL process. Compared with the Oxy-CaL system, the energy reduction achieved is directly attributable to the introduction of the membrane separation, which saves power for the production of O<sub>2</sub> enriched air.

Table 1: Main inputs and results for the mOxy-CaL30 process compared with other CO<sub>2</sub> capture systems over the same reference plant (Ortiz et al., 2017)

	Parameter	Reference CFFP (air combustion)	oxy- combustion	CaL	Oxy-CaL 30	mOxy- CaL30	
CFPP	$\dot{m}_{coal}$ (kg/s)	42.20	55.05	42.20	46.10	46.1	
	$\dot{m}_{air}$ (kg/s)	475	-	475	213	0	
	$\dot{m}_{O_2}(x_{O_2} = 0.95)$ (kg/s)	-	136.91	-	68.05	0	
	$\dot{m}_{OEA}(x_{O_2} = 0.38)$ (kg/s)	-	-	-	-	281.02	
	$Y_{fg}$	-	0.78	-	0.63	0.63	
	$F_{fg}$ (kmol/s)	17.12	3.85	17.12	10.25	10.25	
	$F_{CO_2}$ (kmol/s)	2.60	3.39	2.60	2.84	2.84	
	$v_{CO_2}$	0.15	0.89	0.15	0.30	0.3	
	$v_{O_2}$	0.023	0.025	0.023	0.025	0.025	
	$\eta_{boiler}$	0.90	0.90	0.90	0.89	0.89	
	$\eta_{CFFP}$	0.377	0.287	0.377	0.353	0.362	
	net work (MW)	490.47	488.80	490.47	500.4	513.59	
	Penalty	-	9.02 %	-	2.56 %	1.63 %	
	SPECCA (MJ/kg CO <sub>2</sub> )	-	4.06	-	0.89	0.55	
	CaL	$F_R/F_{CO_2}$	-	-	15	15	15
		$F_0/F_{CO_2}$	-	-	0.05	0.05	0.05
		$\tau$ (min)	-	-	3.05	2.79	2.79
$T_{calc}$ (°C)		-	-	950	950	950	
$T_{carb}$ (°C)		-	-	650	650	650	
$E_{CO_2}$		-	-	0.827	0.950	0.950	
$\dot{m}_{coal}$ (kg/s)		-	-	18.48	22.34	22.34	
$\dot{m}_{air}$ (kg/s)		-	-	0	0	0	
$\dot{m}_{O_2}$ (kg/s)		-	-	48.00	58.51	58.51	
$Y_{fg}$		-	-	0.7	0.7	0.7	
$\dot{m}_{gas,calc}$ (kg/s)		-	-	165.07	203.75	203.38	
$\dot{m}_{CO_2,calc}$ (kg/s)		-	-	150.45	185.78	185.43	
$x_{CO_2}$		-	-	0.030	0.019	0.019	
$x_{O_2}$		-	-	0.028	0.032	0.032	
$P_{HE,solids}$		-	-	640.92	696.86	698.06	
$\dot{W}_{sec}$ (MW)		-	-	75.80	119.93	118.14	
$\dot{W}_{solids}$ (MW)		-	-	45.70	49.70	49.73	
$\dot{W}_{ASU}$ (MW)		-	-	34.56	42.13	42.13	
$\dot{W}_{comp,CO_2}$ (MW)		-	-	57.55	71.06	70.94	
$\dot{W}_{comp,FG}$ (MW)		-	-	12.53	7.37	7.37	
$\eta_{int}$		-	-	0.303	0.295	0.301	
Penalty		-	-	7.43 %	5.82 %	6.12 %	
SPECCA (MJ/kg CO <sub>2</sub> )		-	-	3.28	2.48	2.49	
Total							
		Penalty <sub>total</sub>	-	9.02 %	7.43 %	8.38 %	7.75 %
		SPECCA <sub>total</sub> (MJ/kg CO <sub>2</sub> )	-	4.06	3.28	3.37	3.05

#### 4. Conclusions

A novel CO<sub>2</sub> capture process is presented in this work. The new process combines the advantages of each one of the subsystems that compose it to achieve a highly efficient CO<sub>2</sub> capture system. Thus, by integrating membrane separation, partial oxy-combustion, and the CaL process, energy consumption for CO<sub>2</sub> capture can be as low as ~3 MJ/kg CO<sub>2</sub>, whereas other capture systems consume up to 25 % more energy (as in the case of oxy-combustion). Moreover, results show a very high capture efficiency (>95 %), which involves a lower number of solids needed in the carbonator, and therefore a reduction of the reactor size and capital costs, which are main limitations of CO<sub>2</sub> capture technologies. Furthermore, a reduced equipment size would improve the flexibility of the plant. Thus, the proposed Oxy-CaL integrated system results in an improved cost-effective CO<sub>2</sub> capture system, which may help deploy this post-combustion CO<sub>2</sub> capture technology at the commercial scale.

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