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Techno-Economic and Off-Design Analysis of Two CO₂ Purification Units for Low-Carbon Cement Plants with Oxy-fuel Calcination

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Abstract

This paper presents the techno-economic assessment of the CO₂ Purification Unit (CPU) included in cement plants based on CO₂ capture via oxy-fuel calcination. Two configurations, targeting two different outlet CO₂ specifications ('moderate' and 'high' purity), have been optimized in order to minimize the increase in the clinker production cost due to this process unit. Air infiltration rate is the parameter with the highest impact on the operational conditions (e.g. separation pressure) and on the incremental cost of clinker due to the CPU (up to 20% for the worst case considered). The work has been carried out within the framework of the CLEANKER project.

Keywords: CCS; CO₂ Purification Unit; CO₂ capture; Calcium Looping; Oxyfuel; Cement; CLEANKER.

Nomenclature

$\Delta c_{\text{clk,CPU}}$	Incremental cost of clinker due to the CPU
CaL	Calcium Looping
CCUS	CO ₂ Capture, Utilization and Storage
CPU	CO ₂ Purification Unit
CRR	CO ₂ Recovery Ratio

1. Introduction

The closure of the CO₂ Capture, Utilization and Storage (CCUS) value chain requires that the CO₂ captured stream complies with specific quality limits (i.e. composition, pressure, temperature, etc.), in order to meet the safety and techno-economic standards of the transportation (by either pipeline or ship), final utilization or storage systems [1].

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For this reason, the CO₂-rich mixture produced by oxy-fuel processes, although already highly concentrated in CO₂ after the capture step, is further processed in a purpose-designed CO₂ Purification Unit (CPU, also named Cryogenic Purification Unit) which produces purified and compressed CO₂ ahead of the final transportation, sequestration or utilization steps. This is the case of the CO₂ mixture released by the oxy-fuel calciner, the reactor of the Calcium Looping (CaL) process where CaCO₃ (i.e. the CO₂-loaded sorbent) is regenerated to CaO, while releasing CO₂ in the vapor phase as a result of the endothermic calcination reaction. In the integrated CaL process, currently under demonstration at TRL 7 in the framework of the H2020 CLEANER project [2], the calciner can be designed to use a variety of fuels (e.g. coal, high-density oil, refuse-derived-fuel, biomass or natural gas) and nearly pure oxygen (95% mol purity) as oxidizing agent. In CLEANER, the technical, environmental and economic performance of different CPU configurations for the full-scale integrated Calcium Looping cement plant are evaluated with process simulations and techno-economic assessments.

According to the most recent techno-economic assessment of oxy-fuel technologies applied to cement plants [3], the CPU is the largest power consumer of the CCS facility and has a non-negligible economic impact on the cost of CO₂ avoided. Therefore, achieving an efficient and cost-optimized CPU design is crucial to the technical and economic success of the overall CCUS chain. This work presents the methodology and results from the assessment of two CPU configurations, both tailored to process the CO₂-rich mixture produced by CaL, and designed for two different quality specifications of the CO₂ final product:

- Moderate purity, based on the less stringent limits set by the ISO standard 27913:2016 [1], and assuming CO₂ pipeline transportation with final storage in a saline formation (CO₂ concentration higher than 95% mol, N₂ content below 2% mol, maximum non-condensables species content below 4% mol, H₂O content lower than 1 ppmv).
- High purity, based on the quality specs typically suggested for CO₂ ship transportation [4], with a CO₂ concentration higher than 99.7% mol and O₂ content lower than 10 ppm_v as a safety measure for EOR applications (H₂O content still lower than 1 ppmv).

2. CPU process description and methodology for techno-economic optimization

The evaluated CPUs are considered to be applied to the CO₂-rich flue gases produced by the full-scale cement kiln equipped with the integrated Calcium Looping process described by De Lena et al. [5]. The full-scale plant has a clinker production capacity of 2,817 t/d, a representative size for a European cement kiln. Fig. 1 shows the general block flow diagram of the CPU for both configurations, which are based on low-temperature phase separation with auto-refrigeration (i.e. the cooling duty to achieve temperatures close to -50 °C is supplied by throttling and re-evaporating the liquid CO₂ stream), but envisaging two different arrangements of the cold box, depending on the level of purification required.

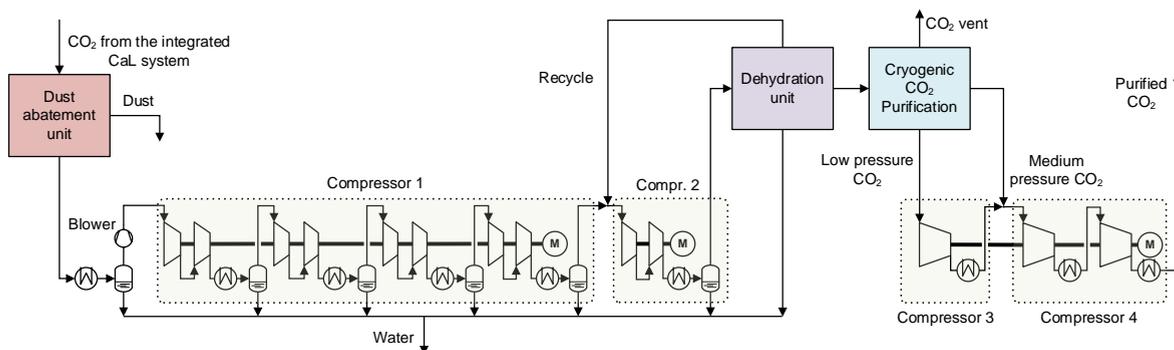


Fig. 1. Block-flow diagram of the CPU proposed for the Cleanker project (adapted from [6]).

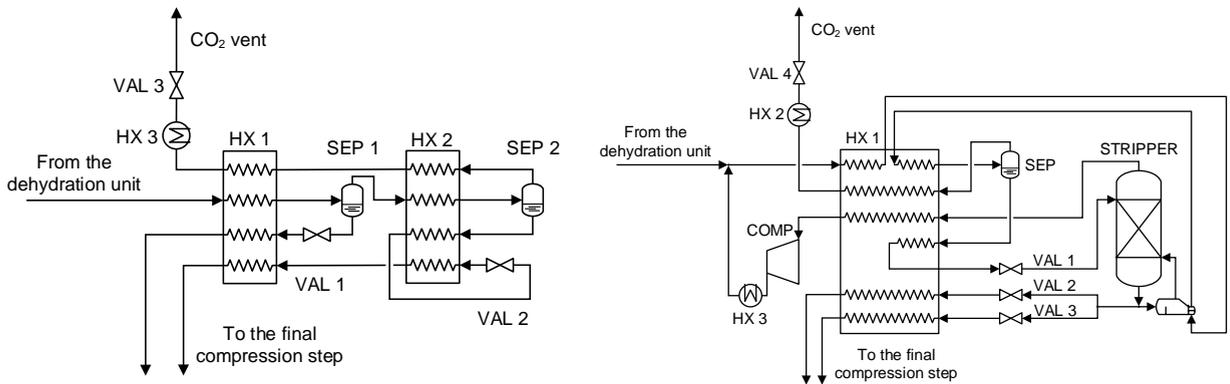


Fig. 2. Detailed schemes of the cold box for the (a) Moderate purity, and (b) High purity cases.

In the moderate purity case, the cold box, reported in Fig. 2-a, includes two multi-stream low-temperature heat exchangers with two sequential phase separators, aimed at maximizing the CO₂ Recovery Ratio (CRR), i.e. the amount of specification-compliant CO₂ made available by the CPU compared to the total amount of CO₂ entering it. On the other hand, for high purity levels, CO₂ distillation is required and therefore the cold box (Fig. 2-b) comprises a low temperature multi-stage separation column, with reboiler, integrated with the phase separator and the multi-stream heat exchanger. These process schemes have been defined from a literature analysis [6], while taking into account also design considerations and information publicly reported on the existing CPU pilots for oxy-fuel purification (e.g. Callide [7], Schwarze Pumpe [8], Ciuden [9]).

The methodology adopted for the techno-economic optimization involves the following steps, for each CPU configuration:

- i) for given boundary conditions (composition and flow-rate of the CO₂ from the integrated CaL process) and design conditions (key pressures, temperatures and split fractions), the process is simulated with Aspen Plus v10 to solve the mass and energy balances and compute the energy performance;
- ii) Simulation results are elaborated in Matlab to complete the economic assessment, the sizing and costing of each equipment unit is carried out (e.g. calculate compressors capital cost, heat exchangers area and capital cost, etc.), the net electricity import and cost is evaluated and the net CO₂ avoidance and emissions are computed;
- iii) The objective function to be minimized in the optimization, i.e. the incremental cost of clinker (due to the presence and operation of the CPU) $\Delta c_{clk,CPU}$ [€/t_{clk}], is finally calculated. $\Delta c_{clk,CPU}$ includes: CPU capital cost, CPU variable operating costs (electricity, carbon tax – on both direct and indirect emissions, water) and CPU fixed operating costs.

After each iteration of the step by step procedure above, the objective function value is returned to the numerical optimizer which automatically changes the design conditions in order to attain a lower CPU cost, until max number of iterations or convergence is reached.

2.1. Process modelling and simulation

The key assumptions are listed in Table 1. Mass and energy balances of both process schemes have been simulated using the Peng-Robinson equation of state with Van der Waals mixing rules, with binary interaction parameters k_{ij} for CO₂-O₂ and CO₂-Ar mixtures retrieved from [10]. Preliminary sizing and cost assessment of the CPU equipment is based on up-to-date literature correlations and/or engineering guidelines defined together with the industrial partners of the CLEANKER project [2]. The general methodology is fully described in Milestone MS11 of CLEANKER [11].

The design variables changed during the optimization for the moderate purity case (Fig. 2-a) are: separation pressure, temperature of separators #1 and #2, minimum temperature difference in the cold box heat exchangers,

temperature increase of liquid CO₂ stream from separator #2 across heat exchanger #2. The design variables changed during the optimization for the high purity case (Fig. 2-b) are: separator pressure, separator inlet temperature, stripper pressure, stripper height, valve #2 outlet pressure, minimum temperature difference in the cold box heat exchanger, split ratio of liquid CO₂ between valves #2 and #3, temperature increase of liquid CO₂ stream from the separator across the main heat exchanger.

Table 1: Main modelling assumptions.

Parameter	Value
Purified CO ₂ delivery pressure	110 bar(a)
Purified CO ₂ delivery temperature	30 °C
Heat losses in multi-flow heat exchangers	1 % of heat exchanged
Pressure drop in multi-flow heat exchangers	2 % of inlet pressure
Minimum process temperature to avoid freezing	$T_{CO_2, triple\ point} + 3^\circ C$
Intercoolers outlet temperature	28 °C
Electricity consumption of cooling system, as a function of rejected heat	0.01 kW _{el} /kW _{th}
Dehydration unit recycle rate	10 %
Dehydration unit pressure drop	1 bar
Electricity consumption of dehydration unit, as a function of removed water	2.29 kWh _{el} /kg _{water}
Compressors polytropic efficiencies	0.82 ÷ 0.88
Electro-mechanic efficiency of compressors and pump drivers	0.95

2.2. List of case studies

Four case studies have been investigated:

- Moderate purity target – Base case;
- High purity target – Base case;
- Moderate purity target – Off-design;
- High purity target – Off-design.

In base cases, air infiltrations are considered to be zero. On the other hand, in off-design cases the air infiltration rate is considered to grow from 0 to 10% of the flue gas flow rate along the year: hence, the results are reported as ranges between the two extreme conditions. All the considered case studies are assumed to burn coal as fuel in both oxy-fuel calciner and air-blown rotary kiln, with a target O₂ content at calciner outlet of 3.5 %_{mol} (dry basis). A 50 €/t_{CO₂} carbon tax is assumed to be in place. Table 2 summarizes the conditions of flue gases entering the CPU for the different air infiltration scenarios.

Table 2: Flue gases entering the CPU.

	No air infiltration	With air infiltration
Gas volumetric flow rate, Nm ³ /h	73'903	81'293
Temperature, °C	60	60
Pressure, bar(a)	0.93	0.93
Gas composition, %mol		
CO ₂	83.2	75.7
H ₂ O	11.5	10.5
O ₂	3.1	4.7
Ar	1.2	1.2
N ₂	0.9	7.9

2.3. Off-design analysis

Besides investigating the most efficient design conditions, the techno-economic assessment considers also the off-design operation. In a full-scale plant, the CaL calciner and the related CO₂ recirculation line would operate slightly sub-atmospheric. Air infiltration is a drawback that the full-scale calciner reactor is expected to show, as it typically

happens in other components of cement facilities (e.g. pre-calciner, pre-heating tower, cyclones etc.) [12], since they are not completely air-tight and operate slightly sub-atmospheric; therefore, they tend to entrain considerable amounts of air between two planned maintenance periods (here assumed to happen once a year); as a result, they are expected to be exposed to air infiltrations and, therefore, it is interesting to evaluate the impact of air infiltrations on the performance, design and costs of the CPU.

Off-design analysis is relevant since the CPU is expected to operate with flow-rates coming from the CaL system periodically changing on a yearly basis (i.e. between two maintenance periods) due to air infiltration rates growing from a minimum right after maintenance to a maximum right before maintenance. In this work, the air infiltration is assumed to change with a linear saw-tooth profile, from 0% (right after maintenance) to 10% (right before maintenance) of the design inlet flow to the CPU. The CPU is designed for the highest inlet flow-rate scenario (called “On-design”), while off-design is the situation right after maintenance, without air infiltration. In off-design, the CPU equipment geometries and sizes are given and a control strategy is defined for its regulation: centrifugal compressors regulated according to the “variable speed” control strategy, UA given for heat exchangers, throttling valves regulated to get separation temperature close to on-design, reboiler regulated to achieve 10 ppm O₂.

3. Results and discussion

Table 4 reports the results for base cases and off-design cases, i.e. key energy performance and cost for both moderate and high purity configurations. Results for the off-design cases are expressed as the range between the situations right after and right before maintenance, except for the incremental cost of clinker which is expressed as a yearly averaged value.

Key results for the base case are: the CO₂ recovery efficiency is 3 points higher for the moderate purity scheme, due to looser CO₂ specifications; the specific power consumption for the high purity scheme is 12% greater; the incremental cost of clinker is 16 €/t_{clk} for the optimized moderate purity scheme, while it grows to 19 €/t_{clk} for the high purity case, both due to larger electricity costs and increased carbon tax costs because of higher CO₂ emissions.

Table 3: Optimal performance and cost results of two different CPUs without (unit always operating on-design) and with air infiltration (unit operating off-design).

Parameter	Units	Base cases		Off-Design cases (range)	
		Moderate purity	High purity	Moderate purity	High purity
Gas mass flow rate at CPU inlet	kg/s	37.05	37.05	37.05 - 39.70	37.05 - 39.70
CO ₂ content in inlet gas	%mol _{dry}	94.1	94.1	84.6 - 94.1	84.6 - 94.1
CO ₂ Recovery Ratio (CRR)	%	99.31	96.15	94.28 - 98.47	92.24 - 96.68
CO ₂ purity	%mol	96.00	99.99	96.21 - 97.08	99.99
Energy-related Key Performance Indicators					
Electric consumption of the CPU	MW	13.51	14.66	14.47 - 16.10	15.95 - 17.18
Specific electric consumption	kWh/tCO ₂ purified	112.6	126.2	121.6 - 141.3	136.6 - 154.1
Economic Key Performance Indicators					
$\Delta c_{clk,CPU}$	€/t _{clk}	16.26	19.29	19.62	22.25

For all the cases investigated, the economic assessment highlights that electricity (which shall be imported from the grid in the integrated CaL plant with CPU) is the main cost driver affecting the $\Delta c_{clk,CPU}$, followed by annualized capital costs and CO₂ tax. From the point of view of capital costs, compressors are the main cost item of the CPU (~80% of CPU CAPEX share). The optimal separation temperature is always close to -50 °C, while the separation pressure changes depending on the scenario and typically ranges between 30 and 40 bar.

Off-design analysis underlines the significant impact of air infiltrations on the additional cost of clinker and CO₂ recovery rate, thereby inducing to design the CPU for the condition right before maintenance (i.e. the worst case scenario in which the flow rate is as high as possible and the CO₂ is more diluted), in order to avoid much lower CRR and excessively higher costs for CO₂ emissions. Off-design analysis also suggests larger minimum temperature differences in the cold box heat exchanger at design conditions, such that a flexibility margin is provided in the HEXs during off-design operation.

In case integrated CaL CO₂ capture is applied to a new cement kiln, the share of total clinker cost attributable to the CPU section alone is ~15%, thereby confirming the importance of properly designing and optimizing this unit in cement plants employing oxyfuel CCS technologies.

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