

PROCESS INTEGRATION OF INDIRECTLY HEATED CARBONATE LOOPING IN LIME PLANT FOR ENHANCED CO₂ CAPTURE

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Abstract

Lime (CaO) production emits significant amounts of CO₂ through both the calcination reactions and the fuel combustion process. In order for the lime industries to reduce their mainly non-avoidable CO₂ emissions, the deployment of carbon capture and storage (CCS) technologies appears as a necessity. One very promising carbon capture method is the indirectly heated carbonate looping (IHCaL). This paper aims to investigate two novel IHCaL concepts, the tail-end and the fully integrated process in order to provide a better understanding of their integration in the lime sector. The concepts are developed and simulated in Aspen PlusTM, heat and mass balance equations are established and a detailed sensitivity analysis is performed. The tail-end process is ideal for retrofitting of existing lime plant, whereas a newly-build lime plant could use the fully integrated IHCaL concept in order to achieve higher CO₂-capture and lower energy consumption. The numerical results for the aforementioned scenarios, revealed that high carbon capture efficiency, 92% and 94% respectively, can be achieved at both concepts. Moreover, the power generation from the heat recovery steam cycle is calculated. Key parameters for effective integration of the concepts are the preheating of the combustion air, the efficiency of the sorbent solid-solid heat exchanger and the utilization of the sorbent purge as lime product.

Keywords: Calcium looping, CO₂ capture, lime production, indirect heating, process model

1. Introduction

Lime (CaO) is a key product for various sectors and plays an essential role for downstream industries. As an important element for soil treatment, it is traditionally used in agriculture, but it has also multiple applications in the manufacturing industry [1].

However, the production of lime remains a carbon-intensive process, since carbon dioxide (CO₂) is released from both the calcination of the raw material, limestone (CaCO₃), and the combustion of the required fuel. It is important to underline that the CO₂ emissions from the calcination process are non-avoidable and cannot be tackled using renewable energy sources. Many carbon capture technologies are being developed with the view for the lime industry to become more CO₂-efficient and maintain its competitiveness [2].

A very promising carbon capture technology is the Indirectly Heated Carbonate Looping (IHCaL), which is considered a very competitive option in comparison to MEA scrubbing and oxy-fuel technology [3].

In calcium looping process, a continuous recirculation of calcium-based sorbents takes place between two main reactors, the carbonator and the calciner. In the fluidized

bed carbonator, the carbon dioxide (CO₂) is captured, as it reacts exothermically, typically at 600°C with lime particles (CaO) and forms limestone (CaCO₃). The sorbents regeneration and release of CO₂ takes place typically at 900°C in the second fluidized bed, the calciner. Since the calcination reaction is strongly endothermic in high temperatures, an amount of thermal energy is needed. In conventional Carbonate Looping (CaL) process, the heat is provided from simultaneous fuel combustion inside the calciner. Moreover, an Air Separation Unit (ASU) is used to provide pure oxygen and a nitrogen-free environment, resulting to a high-purity CO₂ stream at the exit of the calciner. In the case of the IHCaL, the heat for the calcination is transferred indirectly from an external combustor to the calciner with heat pipes, a fact that reduces the efficiency penalty considerably compared to the direct firing inside the reactor.

The main objective of this study is to investigate the integration of Indirectly Heated Carbonate Looping (IHCaL) in a lime plant, by simulating two novel IHCaL concepts, the tail-end and the integrated process.

2. Process Concepts

The reference unit studied in the present paper is a lime plant from CaO Hellas located in Thessaloniki, Greece. The plant comprises a double-shaft kiln, which burns limestone into lime with a production capacity of 150 tonnes per day in normal operating conditions. The raw material of the process is rich in limestone (98%) and the fuel used to provide the necessary heat is petcoke.

The steady-state process models are developed in ASPEN Plus™ to calculate the balances of the existing plant and of the integration scenarios. Two IHCaL scenarios are studied based on the specific lime plant, the first one is the retrofitting of the CO₂ capture unit in the existing lime plant (tail-end process) and the second one is the fully integrated process.

2.1 Tail-end process

In the tail-end solution, the IHCaL facility is located downstream the lime process line as shown schematically in Fig.1. This concept entails a low amount of integration, which makes it suitable for retrofitting. The reference lime plant facility from CaO Hellas is depicted on the left within the dotted area. The raw material stream consisting mainly of limestone and the required fuel for the combustion, petcoke, enter the double shaft kiln. The calcination of limestone takes place and the produced lime exits the kiln and is cooled down. In addition, the flue gases leave from the upper part of the kiln and an amount of their heat is recovered and used in the calcination process for the raw material preheating.

In the IHCaL unit, the flue gases from the lime plant of CaO Hellas, together with the flue gases produced from the combustion of fuel for IHCaL, insert the carbonator

with the aid of a blower. In the carbonator, the CO₂ reacts with the CaO that comes from the calciner forming CaCO₃. The resulting multiphase solid-gas stream exits through the top of the carbonator and is separated in Cyclone 1. The gas which consists of the CO₂-lean flue gas, exits the cyclone through the top and the solids leave through the bottom.

The solids from the carbonator flow into the second reactor and undergo full calcination at high temperature, releasing CO₂. A high purity CO₂-stream is separated then from the solid particles of CaO in Cyclone 2. Part of the CaO stream returns back to the carbonator and the remaining part is extracted as a purge stream through a sorbent extraction point. The purged CaO has high lime concentration (>98%) and therefore, it can be considered as lime product output. Thus, the implementation of the tail-end solution expands the lime production capacity of the plant, while allowing the carbon capture from its flue gases.

Regarding the energy optimization of the process, a solid-solid heat exchanger is installed to transfer heat between the circulating sorbent streams that exit the cyclones, reducing the energy requirement in the calciner to heat up the entering solids. Another parameter that reduces the IHCaL fuel consumption in the combustor is the preheating of the air from the combustion flue gases. Finally, the excess heat from the CO₂-lean flue gases, the CO₂-rich flow cooling and the regulation of the carbonator temperature is exploited to produce electricity power through a heat recovery steam generator [4].

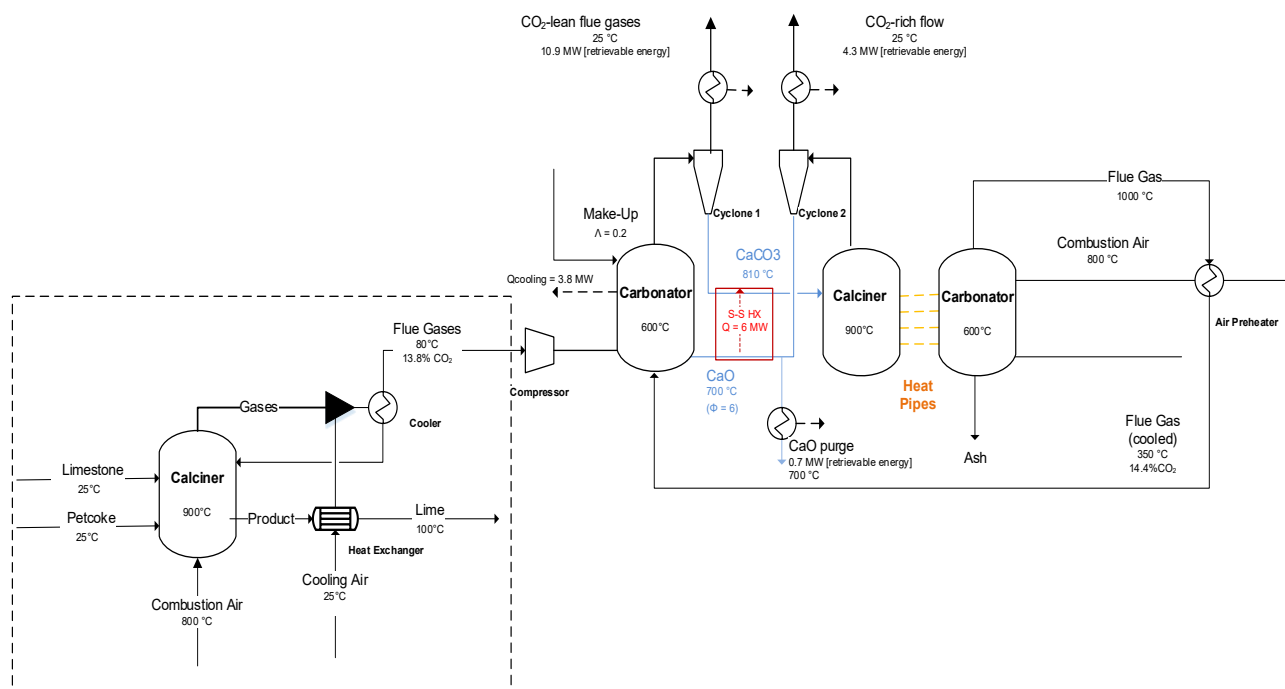


Figure 1: Flowsheet diagram of the tail-end integration of the IHCaL downstream to the lime plant of CaO Hellas

2.2 Fully integrated process

The second concept is the full integration of the process. The lime production kiln is holistically replaced by the IHCaL process, which serves both as a calcination unit and as a carbon capture facility, as it is illustrated in Fig.2. The same operation parameters as in the tail-end process are used.

The raw material, limestone, is fed directly to the carbonator, where it is mixed with the circulating lime solids. Similarly, the flue gases from the IHCaL combustor enter the carbonator, where the CO₂ they contain reacts with the lime particles forming CaCO₃. The solids and the gases exiting the carbonator are separated by Cyclone 1. The solid CaCO₃ stream is calcined producing a high purity CO₂-stream and a solid CaO stream which are separated in Cyclone 2. One part of the solid CaO reenters the carbonator, while the other part exits as purge product of the process.

Similar to the tail-end process, a steam cycle that recovers the excess heat for power generation is considered.

The particularity of this solution is that the CO₂ from the limestone calcination is produced entirely in the calciner, avoiding the need for an extensive downstream separation for mainly particle matter removal. Because of this, the only CO₂ emissions to be captured in the carbonator are the ones related to the burning of the fuel in the combustor to generate the heat for the calciner. This poses a huge advantage with respect to the tail-end scheme as much lower heat penalties and recirculation rates for the same conditions are happening.

Additionally, since in the integrated solution the calcination is carried out with indirectly added heat, there is no contamination of the lime with fuel-related particles like ash or sulfur, thus, a purer product than in the conventional way is to be expected.

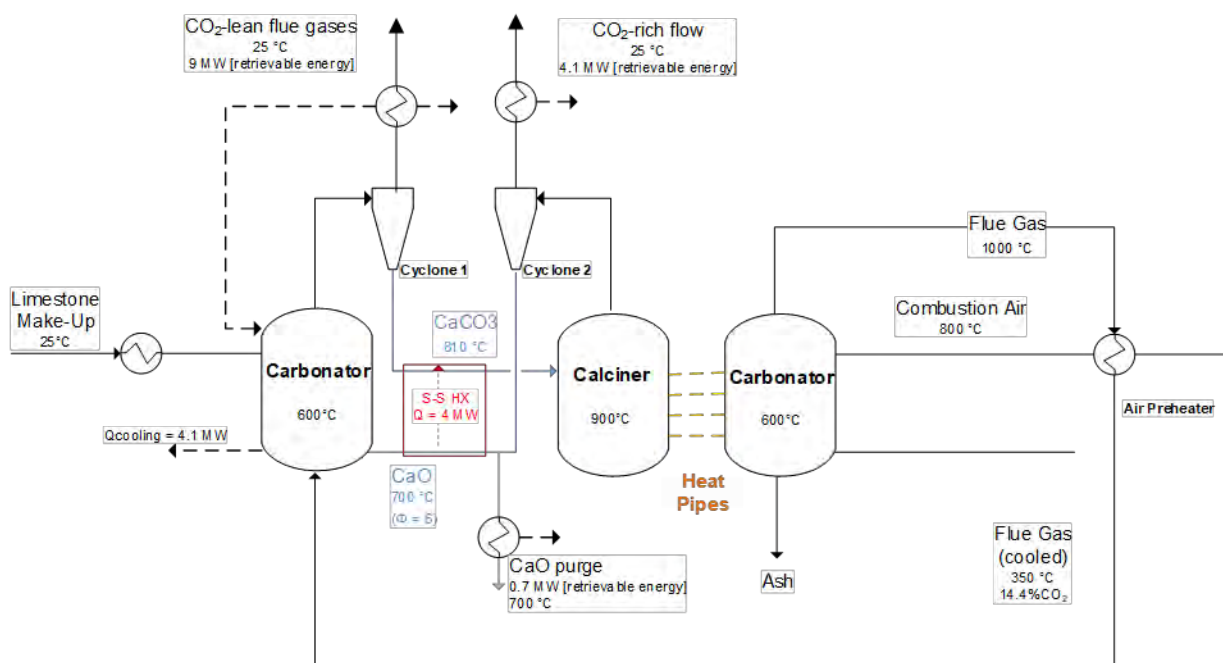


Figure 2. Flowsheet diagram of the fully integrated process

3. Methods

3.1 Process modeling

In this paper, the software ASPEN Plus™, version V11, was used to perform the mass and energy balances calculations. ASPEN Plus™ is a process simulation software, which has a powerful database of properties for various substances [5]. For the process, all simulations were developed in steady-state condition and the calculation of the properties of the substances were based on the Redlich-Kwong-Soave model [6]. The solid-gas separation in the cyclones was assumed to be ideal. The system pressure was set to 1.013 bar, while pressure drops were neglected.

To model the fuel combustion in ASPEN Plus™, the fuel was inserted into a yield reactor that decomposes the stream into its elementary molecules and heat. The resulting stream is burned with air in a Gibbs reactor, which minimizes the Gibbs' free energy, assuming chemical equilibrium. The temperature of the IHCaL combustor is set to 1000°C to allow for 100°C temperature difference between the combustor and the calciner. The calcination and carbonation process in the IHCaL reactors are modelled also with a Gibbs reactor, assuming chemical equilibrium.

However, since in the carbonator only the active solids take part in the reactions, in order to account for the limitations of the kinetics the $X_{max,ave}$ is used. $X_{max,ave}$ is the turning point from the fast to the slow carbonation

regime, which shows the average maximum carbonation conversion of the solids and represents their CO₂ carrying capacity. In the simulation, a fraction ($X_{max,ave}$) of CaO enters the carbonator to react with CO₂ and the rest ($1 - X_{max,ave}$) by-passes the carbonator and is introduced back to the calciner unreacted.

$X_{max,ave}$ is influenced by the specific sorbent circulation as well by the make-up flow and is calculated based on the following equation of the model of Abanades [7]:

$$X_{max,ave} = \frac{f_m(1 - f_w) * F_o}{F_o + F_R(1 - f_m)} + f_w \quad (1)$$

The f_m and f_w are constants which depend on sorbent's physical and chemical characteristics. F_o and F_R represent the make-up flow and the circulation flow respectively.

Moreover, another important parameter is the molar conversion of CaO into CaCO₃ (X), which is defined as the ratio of moles of CaCO₃ to the moles of Ca (Eq. (2)).

$$X = \frac{n_{CaCO_3}}{n_{Ca}} \quad (2)$$

In particular, there are two characteristic X -values that are used in the analysis of CaL processes, the molar conversion of the sorbent exiting the carbonator (X_{carb}) and the molar conversion of the sorbent exiting the calciner (X_{calc}) [8]. The upper limit for the former molar ratio is the $X_{max,ave}$.

The fuel and raw material composition is defined according to the reference plant. Petcoke (NHV=8287 kcal/kg) is implemented as fuel for the combustor in the reference lime plant of CaO Hellas and in the IHCaL. Similarly, the limestone composition from the reference plant (98 wt% CaCO₃) was used for all the limestone inputs in the model.

3.2 Key Performance Indicators

The following key performance indicators were identified in order for a detailed evaluation of the concepts to be achieved. Many key components of the study are based on previous work developed by Charitos et al. [9].

The carbon capture efficiency, E , is defined as the ratio of the captured CO₂ to the generated CO₂, in terms of molar flow rate. There values considered in Eq.3 for the calculation of E , are the output mole flows of CO₂ from the calciner ($F_{CO_2}^{calc,out}$) and from the carbonator ($F_{CO_2}^{carb,out}$).

$$E = \frac{F_{CO_2}^{calc,out}}{F_{CO_2}^{calc,out} + F_{CO_2}^{carb,out}} \quad (3)$$

An important indicator for this study is the product ratio (PR), i.e., the ratio of the production capacity of the new process ($\dot{m}_{CaO,prod}^1$) to the original production ($\dot{m}_{CaO,prod}^0$) as defined in Eq. 4.

$$PR = \frac{\dot{m}_{CaO,prod}^1}{\dot{m}_{CaO,prod}^0} \quad (4)$$

The heat ratio, HR, Eq.(5), is used to present the heat requirement for CO₂ capture and lime production. It is calculated considering the lime produced and the heat requirement in the original process, (Q_{in}^0) and in the entire process including CO₂ capture solution (Q_{in}^1).

$$HR = \frac{Q^1 / \dot{m}_{CaO,prod}^1}{Q^0 / \dot{m}_{CaO,prod}^0} \quad (5)$$

3.3 Operational Parameters

One dimensionless parameter is the specific make-up ratio (Λ), which is defined in Eq. (4) as the ratio between the molar flow of make-up calcium species into the IHCaL (F_o) and the total CO₂ molar flow. In order to sustain the continuous carbonate looping process for the capture of CO₂, it is necessary to purge the solid inventory and replace it with fresh limestone (make-up flow). This refreshing of the sorbent is necessary to (i) avoid the build-up of inert species such as ash and calcium sulfate (CaSO₄), and to (ii) ensure the proper activity of the sorbent.

$$\Lambda = \frac{F_o}{F_{CO_2}} \quad (6)$$

Another important dimensionless parameter is the specific sorbent circulation rate (Φ), which considers the molar flow rate of calcium species that are fed back to the carbonator from the calciner, F_R , as defined in Eq.(5).

$$\Phi = \frac{F_R}{F_{CO_2}} \quad (7)$$

Moreover, a base case was defined and its key parameters are shown in Table 1. In the sensitivity analyses that follow, the base case values undergo a range of variation, where each base case parameter changed leaving the others constant in order to study their influence on the process.

Parameter	Description	Value
Λ	Specific make up rate	0.2
Φ	Specific sorbent circulation rate	6
$T_{Preheated\ Air}$	Preheated air temperature[°C]	800
$T_{CaCO_3\ in}$	Sorbent temperature at calciner inlet[°C]	810
$T_{Calciner}$	Calciner operating temperature[°C]	900
$T_{Carbonator}$	Carbonator operating temperature[°C]	650
$T_{Combustor}$	Combustor operating temperature[°C]	1000
$E_{Cyclone}$	Cyclone separation efficiency[%]	100

Table 1. Main IHCaL parameters with their base case values

4. Results and discussion

4.1 Results of the reference plant

In the present work, a steady-state process model was developed of the reference lime plant of CaO Hellas without the IHCaL process. The heat and mass balances of the facility were calculated with less than 2% deviation from the reference values provided and therefore, the model is assumed validated.

Parameter	CaO Hellas Data	Simulation Data	Relative Error
CO ₂ in flue gases [v/v% _{wet}]	13.8	13.7	+0.7%
Direct fuel consumption [MJ/t _{CaO}]	5620	5575	-0.8%
Production [t/h]	150	148	-1.4%

Table 2. Validation of the lime plant simulation of CaO Hellas

4.2 Results of the IHCaL models with the base case parameters

A summary of the results of the IHCaL simulations is presented in Table 3. For the calculations the base case parameters were assumed for both concepts.

In the tail-end concept, the product ratio is as high as 2.66, which means that more lime is being produced in the IHCaL facility than in the reference lime plant. Additionally, in the integrated concept the product ratio is 0.98. It was expected that for the same raw material input as in the reference plant the amount of produced lime in the integrated process would be similar. The purge lime extracted from IHCaL showed a high purity (98 wt% CaO). Consequently, it can be sold as product of the lime production, which makes both IHCaL concepts especially suitable and profitable for application to lime plants.

The direct fuel consumption of the reference plant is 5575 MJ/t_{CaO}. In the tail-end process there is a 265% increase of the fuel consumption with respect to the reference plant. For the fully integrated process, the increase in fuel consumption is 239%. The electricity generated through heat recovery amounts to 67.6% and 65.3% respectively of the total thermal energy input. This implies a strong reduction in the net CO₂ emissions, considering the avoidance of the CO₂ produced from the grid's power generation.

Parameters	Tail- end solution	Integrated solution
System Parameters		
Heat Ratio (HR)	2.66	1.57
Product Ratio (PR)	1.63	0.98
CO ₂ capture efficiency(E)	92%	94%
Specific sorbent circulation rate (Φ)	6	6
Specific Make-up ratio (Λ)	0.20	0.48
Carbonator		
Operating Temperature [°C]	650	650
Flue gas from lime plant molar flow [kmol/s]	0.31	-
Flue gas from IHCaL combustion [kmol/s]	0.32	0.21
Total Flue gas flow to the carbonator [kmol/s]	0.63	0.21
Total CO ₂ molar concentration in the flue gas [kmol/s]	0.14	0.14
Flue gas inlet temperature [°C]	114	336
Sorbent temperature at carbonator inlet [°C]	700	700
Molar conversion of the sorbent exiting the carbonator, X_{carb} [mol _{CaCO3} / mol _{Ca}]	0.18	0.14
Max. average carbonation conversion, X_{max} [mol _{CaCO3} /mol _{Ca}]	0.25	0.33
Calciner		
Operating Temperature[°C]	900	900
Sorbent mole flow at calciner inlet[kmol/s]	2020	1427
Sorbent temperature at calciner inlet[°C]	810	810
Purge CaO flow[t/day]	93.9	145
Heat Input[MW]	20.2	11.9
Combustor		
Operating Temperature [°C]	1.000	1.000
Total fuel Consumption IHCaL[kg/s]	0.64	0.41

Table 3. Results of the tail-end and the integrated solution with the parameters of the base case

	Tail-End	Integrated
CO₂ balance		
CO ₂ Molar flow rate from kiln [kmol/h]	156	-
Molar flow rate of CO ₂ from IHCaL combustor [kmol/h]	169	107
Molar flow rate of CO ₂ from calcined make-up in IHCaL [kmol/h]	65	106
Total CO ₂ output [kmol/h]	390	226
Direct CO ₂ emissions [kmol/h]	358	213
Captured CO ₂ flow rate [kgco ₂ /t _{CaO}]	1573	1562
Energy balance		
Total Heat Input [MW]	29.1	15
Direct fuel consumption [MJ/t _{CaO}]	14787	13352
Power generation (P_{el}) [MW _{el}]	19.7	9.8

Table 4. Model results: CO₂ and energy balances

4.3 Sensitivity Analysis

4.3.1. Tail-end process

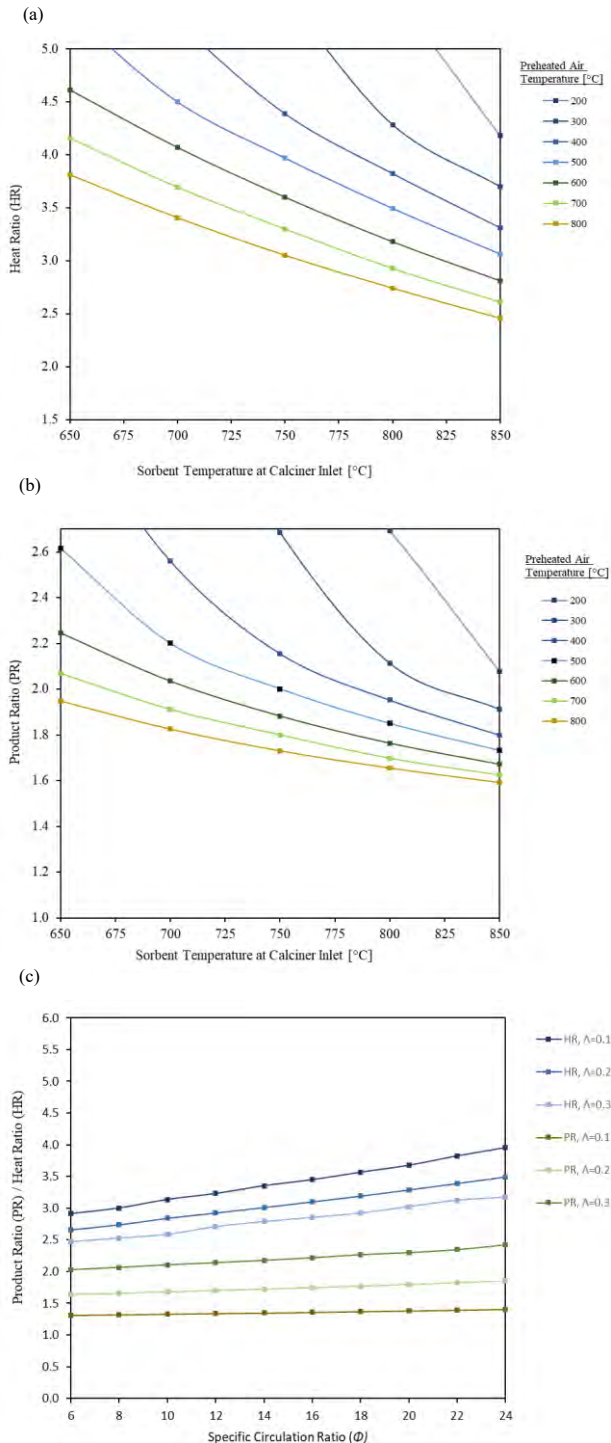


Figure 3: Sensitivity analysis for the tail-end integration concept. In (a),(b) the influence of $T_{PreheatedAir}$ and T_{CaCO_3in} on heat ratio (HR) and product ratio (PR) respectively is shown.

In (c) the variation of HR and PR for different specific circulation ratio (Φ) and make-up ratio (Λ) is depicted.

In Fig. 3 (a) HR is shown as a function of the sorbent temperature at the calciner inlet (T_{CaCO_3in}) for different values of preheated air temperature ($T_{PreheatedAir}$) under the assumption of constant circulation rate (Φ) and make-up ratio (Λ). An increase in the temperature of the

combustion air and the temperature of the sorbent stream at the calciner inlet, leads to lower HR. This means that the heat requirement is influenced by the heat exchange at the preheater and at the solid-solid heat exchanger. The process becomes more efficient as the required amount of energy decreases and the value of HR drops accordingly for a fewer fuel consumption. The preheating of the air appears to be a very important factor of the IHCaL simulation as not only is less fuel needed in the combustor to raise the temperature at the desirable levels, but also the subsequent smaller CO₂-emissions from this unit decrease the heat needs of the carbonation/calcination loops. Additionally, in Fig. 3 (b) it is seen that in this case the PR decreases similarly. This fact is expected, because as the CO₂ emissions decrease due to smaller fuel consumption, less make-flow enters the calciner and the purge stream decreases proportionally.

Fig.3(c) presents the relation of heat ratio (HR) and product ratio (PR) to specific circulation ratio (Φ) for different values of make-up ratio (Λ). A rise in Φ results in an increase of HR, because higher circulation requires higher consumption of fuel, due to the sensible heat needed to balance the temperature difference of the two reactors, i.e. the carbonator (650 °C) and the calciner (900 °C). The product rate remains almost constant, because the circulation has an insignificant effect on purge production. Additionally, the influence of Φ on Λ is illustrated. The limestone make-up flow has a proportional relationship to purge lime flow, since inside the calciner it is decomposed to carbon dioxide and lime, therefore the amount of produced purge lime rises. Moreover, the HR value reduces for higher Λ . Hence, if HR decreases and PR increases for higher Λ , it is evident that it is more efficient to add extra limestone in the IHCaL process to be calcined than to calcine it in the traditional lime plant process. As every additional limestone particle added in the lime plant energy is needed to both calcine it and capture the respective CO₂ in the IHCaL plant. In contrast, the direct calcination of the limestone in the calciner of the IHCaL plant inherently creates a high purity CO₂ stream.

4.3.2. Fully Integrated process

For the fully integrated solution, the make-up stream is the main raw material input, limestone, for the calcination process. Consequently, the make-up ratio can only be controlled indirectly and is considered a dependent variable in the sensitivity analysis.

In comparison to the tail-end solution, there is no variation in the production flow due to changes in the temperatures. This is expected because the make-up flow rate is a constant value. Thus, there is no variation of the PR at different temperatures.

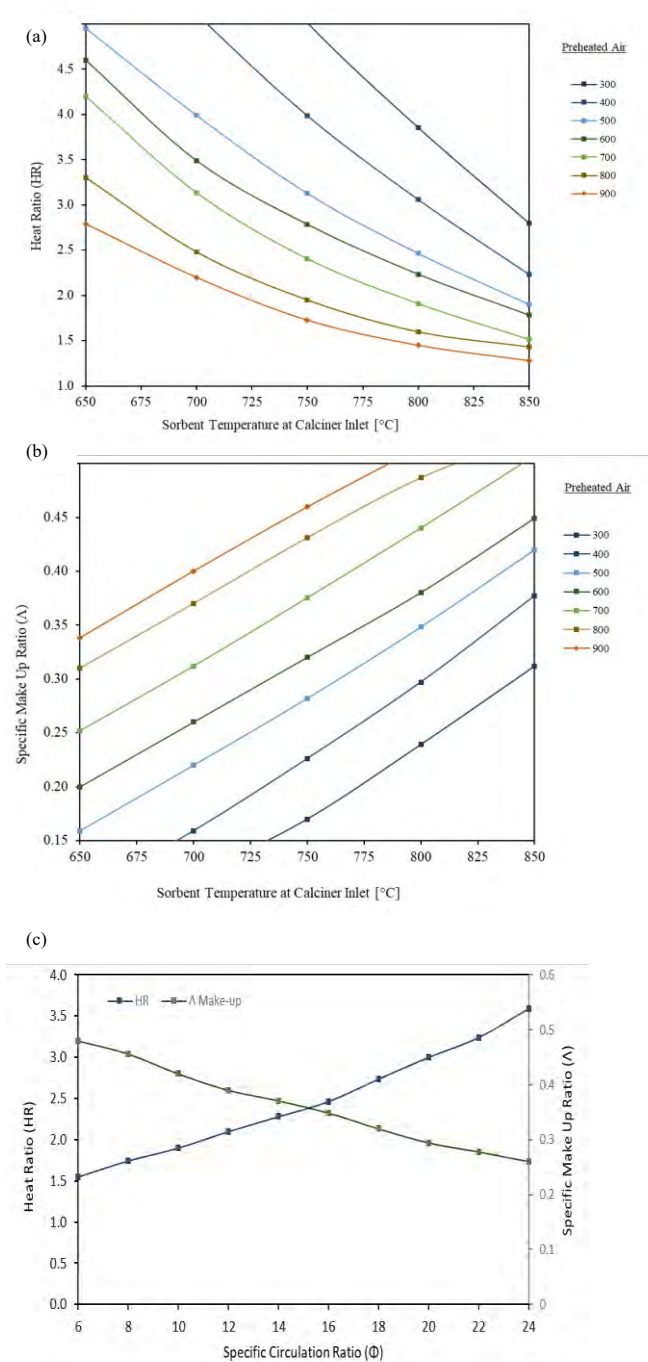


Figure 4: Sensitivity analysis for the full integration concept. In (a),(b) the influence of $T_{\text{PreheatedAir}}$ and $T_{\text{CaCO}_3\text{in}}$ on heat ratio (HR) and specific make up ratio (PR) respectively is shown. In (c) the variation of HR and Λ for different Φ is shown.

In Fig.4 (a) the value of HR decreases for higher $T_{\text{CaCO}_3\text{in}}$ and higher $T_{\text{PreheatedAir}}$ under the assumption of constant circulation rate (Φ). It is remarkable that, compared to the results from the tail-end solution, the values of HR are much lower for the same temperature values. Furthermore, towards the limit of the maximum theoretical integration, the HR tends towards 1.0, whereas in the tail-end solution, the minimum achievable HR is around 2.5.

The impact of higher $T_{\text{CaCO}_3\text{in}}$ and $T_{\text{PreheatedAir}}$ on Λ is shown in Fig.4 (b). The value of Λ will rise, because the process becomes more efficient and the CO₂ decreases.

Fig.4 (c) presents the relation of heat ratio (HR) and make-up ratio (Λ) to specific circulation ratio (Φ). A rise in Φ results in an increase of HR, because higher circulation requires more fuel consumption, due to the required sensible heat to balance the temperature difference of the carbonator and the calciner in the process. The make-up ratio has a negative correlation to the CO₂-flow, which increases for higher circulation and therefore Λ decreases accordingly.

5. Conclusions

This study presents two approaches for the integration of the IHCaL process into a lime plant using as process simulation tool the software of ASPEN PLUS™, version V11.

The tail-end solution is suitable for retrofitting to the existing lime plants since the CO₂ emissions were captured with an efficiency of 92% with an increase of 166% in the total direct fuel consumption. Additionally, the high product output mass flow from the tail-end process proves that a great increase in the production takes place, of 65%, and therefore, this concept appears to be a good option for existing lime production facilities that opt to minimize their carbon footprint by capturing CO₂ and expand their production capacity.

In the fully integrated solution, where the lime production is integrated into the IHCaL process, decreased CO₂ emissions can be achieved. It is worth noting that the CO₂ capture efficiency reaches 94% with an increase of 57% in the direct fuel consumption. Therefore, it can be proposed that a new lime plant could be constructed with a fully integrated process to achieve very high CO₂ capture at smaller cost.

From the sensitivity analyses, it is concluded that a low energy penalty in the IHCaL process can be opted with the maximization of the preheating temperature of the combustion air and with the best possible heat exchange in the solid-solid heat exchanger. Furthermore, the utilization of the produced lime in the IHCaL process is important for the viability of the tail-end solutions.

The developed models and results will be further used to establish boundary conditions for upcoming test campaigns at a pilot plant within the ANICA project framework, as well as to execute economic, environmental, and risk analyses on the integration of the IHCaL into the lime production process.

Acknowledgements

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Nomenclature

CaL	Calcium Looping
Calc	Calcliner
Carb	Carbonator
IHCaL	Indirectly Heated Carbonate Looping
CH	CaO Hellas
E	CO ₂ Capture Efficiency
F_0	Molar Flow of Make-up CaCO ₃
F_{CO_2}	Total Mole Flow of Produced CO ₂
F_R	Molar Flow of CaO fed to Carbonator
HMB	Heat and Mass Balance
HR	Heat Ratio
KPI	Key Performance Indicator
NHV	Net Heating Value of Fuel
PR	Product Ratio
\dot{Q}_{IHCaL}	Heat requirement IHCaL
$\dot{Q}_{Lime\ Process}$	Heat requirement for lime production
$T_{CaCO_3\ in}$	Sorbent temperature at calciner inlet
$T_{Preheated\ Air}$	Preheated air temperature
X	Molar Conversion of CaO to CaCO ₃
Λ	Specific Make-up Ratio
Φ	Specific Sorbent Circulation Ratio