Indirectly Heated Calcium Carbonate Looping - Reducing CO2 Emissions from Lime Plants. A Techno-economic and Environmental Assessment

Angela Rolfe ^{1*}, S. Rezvani ², F. Franco ², C. Brandoni ¹, O. De Priall¹, N. Hewitt ¹ and Y. Huang ¹ 1 CST, Ulster University, United Kingdom 2 ESTRA – Energy Technology Strategies Ltd, United Kingdom *Corresponding Author: <u>a.rolfe@ulster.ac.uk</u> (Angela Rolfe)

ABSTRACT

Lime plants produce non-avoidable CO_2 . Calcium carbonate looping carbon capture is used to reduce CO_2 emissions from lime plants. Indirectly heated calcium carbonate looping eliminates the air separation unit from the capture process. There are two integration methods considered, tail-end and fully integrated. A techno-economic and environmental assessment has been performed. The tail end case has a larger thermal input but produces more lime and electricity compared to the integrated case, which lowers its break-even selling price. The integrated case has lower project costs and direct CO_2 emissions. The capture rate is 90% for the tail-end and 91% for the integrated case.

Keywords: calcium carbonate looping, carbon capture, indirectly heated, techno-economic assessment, life cycle assessment

NONMENCLATURE

Abbrevi	ations
ASU	Air separation unit
BESP	Break-even selling price
CaCO₃	Limestone
CaO	Lime
CC	Carbon capture
CCL	Calcium carbonate looping
CO ₂	Carbon dioxide
DSK	Double-shaft kiln
FRS	Fossil resource scarcity
GW	Global warming
IHCal	Indirectly heated calcium carbonate
incui	looping
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment

RK	Rotary Kiln
SPECCA	Specific primary energy consumption for CO ₂ avoided
SRF	Solid recovered fuel

1. INTRODUCTION

Lime is used in agriculture and many manufacturing industries. In lime production plants, 60-65% of direct CO_2 emissions are a result of producing lime (CaO) from limestone (CaCO₃), generating CO_2 as a byproduct [1]. Due to this, the only method to significantly reduce CO_2 emissions is by employing carbon capture (CC) technology. Pre-combustion CC is unsuitable for lime plants as a large proportion of CO_2 would not be captured.

There are numerous CC technologies under investigation including membrane and solvent based. However, they have high energy penalties leading to unfavorable economics [2]. Calcium carbonate looping (CCL) is a second-generation post-combustion capture technology that is suitable for integration into many industrial and power plants, e.g., cement, steel and lime plants, and pulverized coal power plants.

In traditional CCL capture, flue gas emissions are fed to a carbonator, where the CO_2 is absorbed by CaO in a reversable exothermic reaction, to form CaCO₃ in accordance with Equation 1.

 $CaO_{(s)} + CO_{2(q)} \leftrightarrow CaCO_{3(s)} \pm 178.2 \, kJmol^{-1}$ (1)

The CaCO₃ is passed to the calciner, where the temperature is increased to 900°C by coal oxyfuel combustion. The CO₂ is released from the CaCO₃ to produce a high purity stream of CO₂ and CaO according to the reversable endothermic reaction in Equation 1. The CO₂ leaves the calciner for further purification and storage, and the CaO is sent back to the carbonator for the next cycle. The CO₂ lean fuel gas is emitted from the

carbonator. The oxygen for the oxyfuel combustion is supplied by an air separation unit (ASU) [3]. The heat rejected from the carbonator and emission streams is used to generate electricity for process utility use and export [4].

The ASU has a high electricity consumption that increases the overall energy penalty of the CCL process. The indirectly heated calcium carbonate looping (IHCal) configuration removes the ASU from the process thus deceasing the energy penalty [5]. The calcination heat is provided indirectly by heat pipes or other heat transfer method, connected to an external combustor. The flue gas from the combustor is directed to the carbonator with the base plant flue gas for CO_2 separation. In [6], other benefits of IHCal compared to traditional CCL are described.

The heat pipe IHCal concept has been tested in a 300 kW_{th} pilot plant, which maintained stable CO₂ capture for over 400 hours [7]. A technical and economical assessment of the concept coupled to a coal fired power plant is given in [6]. The energy penalty was reduced by 1.5%-points compared to the traditional CCL process. In [8], the heat pipe IHCal process was assessed with two concepts for integration into a host rotary kiln (RK) lime plant. In the first concept, the tail-end solution, direct CO₂ emissions were reduced by 70.5% compared to the host case with a 154% increase in fuel consumption. For the second concept, the fully integrated solution, the direct CO₂ emissions are reduced by 87.4% and an increase in direct fuel consumption increase of 63% compared to the host plant. The same two integration concepts were studied in relations to a double shaft kiln (DSK) lime plant in [9]. The carbon capture efficiency was found to be 92% for the tail-end solution, and 94% for the fully integrated solution. Electricity generation was calculated as 19.7 and 9.8 MWe for the tail-end and fully integrated solutions, respectively. An overview of the ANICA project, which incorporates [8, 9], is given in [10], along with the conditions for upcoming test campaigns at the 300 kW_{th}scale, under lime and cement plant conditions.

The work presented here builds on the previous assessments and provides a techno-economic and environment assessment of the RK lime plant with two integration concepts of the IHCal process described in [8, 9, 10].

2. PROCESS SOLUTIONS

2.1 RK Lime Plant – Base Case

In the base case lime plant, limestone is fed into the RK via a preheater, which is heated by the kiln flue gas. The limestone is calcined to burnt lime in the RK, with the

required heat provided by lignite/air combustion. The burnt lime exits the kiln and is cooled and further processed. The heat recovered from the burnt lime is used to preheat the combustion air. After heat exchange, the flue gas is quenched and filtered before being emitted.

2.2 RK Lime Plant – Tail-end Case

In the IHCal tail case solution, the flue gas is fed from the RK plant to the carbonator to undergo the CO_2 separation process. The tail-end case is suited to an existing RK plant, as retrofitting requires very little modification to the base plant. A drawback of the tailend case is that the process CO_2 is released twice, and the solids mass flows in the IHCaL reactor exceed that of the cement/lime plant, leading to very large plant sizes [10].

2.3 RK Lime Plant – Fully Integrated Case

The fully integrated case, shown in *Figure 1*, is not suitable for retrofit as the base plant is completely replaced with the IHCal unit to incorporate CC within the process [11]. The lime plant's raw material $(CaCO_3)$ is used as sorbent in the IHCaL process, and the purge from the IHCaL process consists of lime (CaO), which is the main product of the lime plant [10].

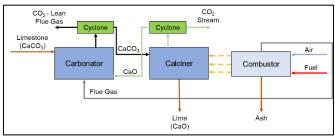


Figure 1: RK Plant – fully integrated IHCaL

3. METHODS

The inhouse ECLIPSE modelling and simulation software [12] is used to provide the mass and energy balance, and the techno-economic assessment. The ECLIPSE models were validated against other ANICA models. The SimaPro[©] software package was used to carry out the Life Cycle Assessment (LCA) [13].

4. RESULTS

4.1 Techno-economic Results

Table 1 details the technical performance for the RK lime plants included is the Base, Tail-end, and Integrated case. The thermal input for the Tail-end case is much larger than the integrated case, however, lime production is greater and indirect CO₂ emissions are reduced due to electricity generation and export. The

 CO_2 capture rate is 90% and 91% for the tail and integrated case respectively.

Table 2 shows the total project costs (TPC) for the RK lime plants. For the tail-end, the total capital cost is 304M, rising to 382M once contingency and interest payments are accounted for. For the integrated case, the total capital cost is 128M rising to 161M.

	Base case (RK)	Tail-end case (RK)	Integrated case (RK)
Thermal input, MWh	38.27	218.44	73.92
Raw meal input, t/h	44.82	94.63*	44.82
Lime production, t/h	25.69	56.65	25.24
CO ₂ emissions (direct), t/h	32.84	12.51	3.90
CO ₂ emissions (indirect), t/h	0.57	-7.08	-1.83
CO ₂ captured, t/h		106.20	41.44
CO ₂ capture rate, %		89.5	91.4
Specific CO ₂ emissions, tCO ₂ /t Lime	1.30	0.097	0.082
Energy consumption, GJ/t Lime	5.36	10.27	8.43
SPECCA (GJ/t CO2 avoided)**		3.54	1.99

Table 1: Technical performance for RK lime plants

*The lime derived from the sorbent make-up stream is included ($F_{CaCO3}/F_{CO2} = 0.2$). ** Indirect CO₂ emissions are included.

Costs in k€	Base case (RK)	Tail-end case (RK)	Integrated case (RK)
The equipment cost	27,947	122,307	51,578
Installation and integration cost	41,474	181,504	76,542
The installed cost	69,422	303,812	128,212
Owner's cost	2,776	12,152	5,124
Total project cost (inc. contingency)	82,610	361,535	152,571
Total project cost (inc. construction interest)	87,290	382,012	161,212

Table 2: Total project costs for full-scale RK lime plants

Table 3 shows the economic assessment results for the RK lime plants. The lime break even selling price (BESP) for the CC cases is high compared to the base case. The tail-end BESP is lower than the integrated case. There are two reasons for this, the first is that the tailend plant produces more lime than the integrated case. It also generates more electricity for export, which provides additional revenue. Utilizing waste derived fuels, should be considered to reduce the BESP.

	Base	Tail-end	Integrated	
	case (RK)	case (RK)	case (RK)	
BESP, €/t Lime	91.86	141.79	134.79	
BEST, Of CENTE	51.00	111.75	10 1.75	
Emissions, t CO ₂ /t Lime	1.300	0.097	0.082	
CO2 reduction, t CO ₂ /t Lime		1.20	1.22	
Cost increase, €/t Lime		49.93	42.93	
Cost of CO ₂ avoided (€/t CO ₂)		41.48	35.23	
*The lignite price = 1.2 €/GJ				

Table 3: Economic results for RK lime plants

4.2 Environmental Results

The goal of the LCA is to quantify and compare the environmental impact of the RK plant and the two IHCaL integration solutions. The study is a cradle to gate assessment and the functional unit is 1kg of lime produced. The ReCiPe midpoint method was used in the analysis. *Table 4* shows the life cycle inventory (LCI) for the processes in relation to the functional unit.

	Process	Base Case RK	Tail-end RK	Integrated RK	Unit
	Limestone Extraction	1.73	1.88	1.75	kg
Inputs	Lignite Extraction	0.25	1.43	0.42	kg
dul	Transport	0.03	0.15	0.35	tkm
	Prep Elec.	0.01	0.01	0.01	MJ
	Process Elec.	0.43	0.00	0.03	MJ
	ARGON	0.05	0.18	0.05	kg
Wastes & Emissions	CO ₂	1.29	0.31	0.11	kg
Vast imiss	Particulates	0.000	0.005	0.000	kg
~ Ш	Ash	0.00	0.06	0.02	kg
ts	Lime	1.00	1.00	1.00	kg
Products	CO ₂ stream	0.00	3.48	1.53	kg
Pre	Electricity	0.00	2.69	0.00	MJ

Table 4: LCI per kg Lime Produced

Figure 2 and Figure 3 show the results for the global warming (GW) and fossil resource scarcity (FRS) impacts, respectively. For GW, the Tail-end case has the lowest impact, however, this low score is highly dependent on the environmental credit for electricity generation and export.

The IHCal configurations have a higher FRS score than the base case lime plant. This is primarily due to the

increase lignite consumption to fuel the CC plants. The Tail-end plant's FRS impact is greater than the integrated case however, this is offset somewhat by the electricity export.

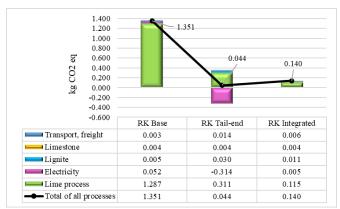


Figure 2: Global warming impact

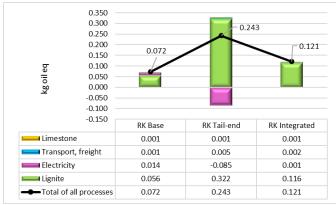


Figure 3: Fossil resource scarcity impact

5. CONCLUSIONS

Lime plants produces CO_2 as a byproduct of combustion and process. CC can be employed to reduce CO_2 emissions. CCL CC can be utilized in lime plants as a tail-end or fully integrated solution. Both solutions reduce the GW indicator, but the tail-end consumes more lignite, and has larger plant sizes and thus, increased TPCs. However, the BESP for the tail plant is reduced due to the increased lime production and the export of electricity generation.

Further works include an examination of the other environmental indicators, an economic sensitivity study, a risk assessment and a complete analysis replacing lignite with solid recovered fuel (SRF). The assessment will also be repeated for the DSK lime plant.

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6. **REFERENCES**

[1] AggNet, "Capturing CO2 from cement and lime production," AggNet, 25 April 2016. [Online]. Available: https://www.agg-net.com/news/capturing-co2-from-cement-and-lime-production. [Accessed 15 January 2022].

[2] M. Junk, M. Reitz, J. Ströhle and B. Epple, "Technical and Economical Assessment of the Indirectly Heated Carbonate Looping Process," ASME Journal of Energy Resources Technology, vol. 138, no. 4, p. 042210, 2016.

[3] J. Hilz, M. Helbig, M. Haaf, A. Daikeler, J. Ströhle and B. Epple, "Long-term pilot testing of the carbonate looping process in 1 MWth scale," Fuel, vol. 201, p. 892–899, 2017.

[4] A. Rolfe, Y. Huang, M. Haaf, S. Rezvani, D. McIlveen-Wright and N. Hewitt, "Integration of the calcium carbonate looping process into an existing pulverized coal-fired power plant for CO2 capture: Technoeconomic and environmental evaluation," Applied Energy, vol. 222, pp. 169-179, 2018.

[5] M. Junk, M. Reitz, J. Ströhle and B. Epple, "Thermodynamic Evaluation and Cold Flow Model Testing of an Indirectly Heated Carbonate Looping Process," Chemical Engineering & Technology, vol. 36, no. 9, pp. 1479-1487, 2013.

[6] M. Junk, M. Reitz, J. Ströhle and B. Epple, "Technical and Economical Assessment of the Indirectly Heated Carbonate Looping Process," Journal of Energy Resource Technology, vol. 138, no. 4, p. 042210, 2016.

[7] M. Reitz, M. Junk, J. Ströhle and B. Epple, "Design and operation of a 300kWth indirectly heated carbonate looping pilot plant," International Journal of Greenhouse Gas Control, vol. 54, no. 1, pp. 272-281, 2016.

[8] M. Greco-Coppi, C. Hofmann, J. Ströhle, D. Walter and B. Epple, "Efficient CO2 capture from lime production by an indirectly heated carbonate looping process," International Journal of Greenhouse Gas Control, vol. 112, p. 103430, 2021.

[9] K. Peloriadi, K. Atsonios, A. Nikolopoulos, K. Intzes, G. Dimitriadis and N. Nikolopoulos, "Process Integration of Indirectly Heated Carbonated Looping in Lime Plant for Enhanced CO2 Capture," in Conference on CO2 Capture, Transport and Storage, Trondheim, 2021. [10] J. Ströhle, C. Hofmann, M. Greco-Coppi and B. Epple, "CO2 Capture from Lime and Cement Plants Using an Indirectly Heated Carbonate Looping Process – The ANICA Project," in Conference on CO2 Capture, Transport and Storage, Trondheim, 2021.

[11] M. Greco-Coppia, C. Hofmann, J. Ströhle, D. Walter and B. Epple, "Efficient CO2 Capture from Lime Production by an Indirectly Heated Carbonate Looping Process," in 15th International Conference on Greenhouse Gas Control Technologies, GHGT-15, Abu Dhabi, 2021.

[12] Ulster University, "ECLIPSE process simulator," Energy Research Centre, University of Ulster, Coleraine, Copyright 1992.

[13]Pré, "SimaPro," 2022. [Online]. Available: https://simapro.com/. [Accessed 18 January 2022].