



Research Paper

Towards decarbonisation of sugar refineries by calcium looping: Process integration, energy optimisation and techno-economic assessment

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ABSTRACT

The sugar refinery process as a food industry is at the pinnacle of the energy hierarchy. Depending on the source of carbon in this industry that could be either from biomass or fossil fuel hydrocarbons, this sector has the potential to become a carbon-neutral or negative industry depending on the type of fuel for power generation. This work will deliver a new proof of concept model for simultaneous sugar refining and carbon capture, thereby transforming sugar into the carbon negative commodity which relies on the utilisation of CO₂ and fresh lime in the sugar refineries carbonisation tanks and in return byproduction of calcium carbonate that can be converted in the calciner of a calcium looping process. This study begins with the design and development of the calcium looping (CaL) process for the integration to the sugar refineries using various coupled and decoupled scenarios for the boiler units based on the type of the fuel, continues with an energy optimisation and techno-economic assessment of the proposed process, and finally culminates with the parametric study on the thermodynamic and economic performance of the sugar refinery retrofitted with the calcium looping. The process simulations revealed that the integrated CaL-sugar refinery can support the electricity exporter by installation of an onsite steam cycle which is able to generate electricity from surplus carbonation heat. The cost of CO₂ avoided for integration of CaL to the reference sugar refinery for natural gas boilers and calciner will be 62 £/tCO₂ which drops to 25 £/tCO₂ if the carbon tax is considered in the analysis and negative carbon emissions are credited. This is equivalent to 61% costs reductions associated refining sugar combined together with carbon capture and storage (CCS).

1. Introduction

The origin of the art of refining sugar stems from Khorasan in Persia, Iran. The modern technology for refining sugar is an energy intensive process which needs power and steam to convert the sugar cane to sucrose [1–3]. Most sugar refineries nowadays around the world utilise fossil fuels, mostly natural gas and some bagasse to generate steam and electricity thereby feeding different sugar refining processes including affination, crystallisation and carbonation. This can emit significant quantities of CO₂ and other pollutants to the atmosphere. Most of the carbon from refining sugar is energy related. This can come from direct combustion in combined heat and power (CHP) units to produce the required steam and electricity. The flue gas composition of these Rankine-based thermal power plants is similar to common coal-fired power plants, in terms of CO₂ content and global warming potentials [4,5]. The decarbonisation scenarios proposed for sugar refineries utility

units can match those ones found in the same large-scale coal-fired plants including conventional post combustion technologies such as chemical absorption with amines [6]. Despite the numerous technological challenges associated with these post-combustion capture technologies including high energy penalty, limited operational flexibility and significant water utilisation, these technologies are relatively mature possessing high technology readiness levels (TRL) 8–9 [7,8]. For instance, the amine based post-combustion capture technologies were once widely used in large-scales power plants using coal, while they offer significant drawbacks including solvent toxicity and amine degradation [9]. These technologies might pose greater challenges in sugar refineries as sugar is a food industry and scrupulous hygiene and cleanliness are requisites during purification and detoxification. The second-generation post combustion carbon capture technologies aim to overcome most of the bottlenecks in conventional carbon capturing systems associated with the energy penalty and installed capacity. The calcium looping process for carbon capturing of a power plant can result

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Nomenclature	
<i>Symbols and Abbreviations</i>	
a_1, a_2	sorbent maximum average conversion-model fitting parameter [-]
A_j	heat exchanger area of equipment j [m^2]
AC	cost of CO ₂ avoided [$£/t_{CO_2 - avoided}$]
ADt	Air dried tonne
ASU	air separation unit
b	sorbent maximum average conversion model fitting parameter [-]
CAPEX	Capital expenditure
C_j	capital cost of equipment j [$£$]
CaL	calcium looping
CCS	carbon capture and storage
CEPCI	Chemical Engineering Plant Cost Index
CHP	Combined heat and power
CF_t	discounted cash flows through the project lifetime [$£$]
CCU	CO ₂ compression unit
DEA	deaerator
ETS	Emission Trading System
e_{CO_2}	direct CO ₂ emissions from the sugar refinery [kg_{CO_2}/ADt]
$e_{CO_2,e}$	specific CO ₂ emissions associated with power generation [$kg_{CO_2}/MW_{el}h$]
$e_{CO_2,eq}$	equivalent CO ₂ emissions [kg_{CO_2}/ADt]
f_1, f_2	sorbent maximum average conversion model fitting parameter [-]
f_i	reaction extent [-]
FCI	Fixed capital investment
FEED	Front-End Engineering Design
F_0	make-up rate (fresh limestone and lime mud) [$kmol/s$]
F_R	sorbent looping rate [$kmol/s$]
G	generator
GWh	Giga watt hour
HX	heat exchanger
$i_{p\&c}$	piping and integration costs indicator [%]
HP	high pressure
IP	intermediate pressure
kWh	kilo watt hour
LCOE	levelised cost of electricity [$£/kWe$]
LCOS	levelised cost of sugar [$£/ADt$]
LP	low pressure
\dot{m}_F	fuel flowrate [kg/s]
m_{O_2}	O ₂ production rate [kg/s]
m	News newsprint production per year [ADt/a]
m	Sugar production per year [ADt/a]
M£	Million pound
MEA	monoethanolamine
MTPD	metric tonne per day
MW_{el}	Mega watt electrical
MW_{th}	Mega watt-thermal
n	cost exponent for the correction of capacity [-]
NDt	Nitrogen-dried tonnes
NPV	net present value [$£$]
OCAPEX	other capital cost [$£$]
OPEX	Operating costs
P_e	specific energy [$MW_{el}h/ADt$]
P_{net}	net power output [MW_{el}]
PC	project contingency [$£$]
Q	direct fuel consumption [MJ_{LHV}/ADt]
q_{eq}	equivalent fuel consumption [MJ_{LHV}/ADt]
Q_j	heat flux of equipment j [kW_{th}]
R	discount rate [%]
r_0	fraction of never calcined limestone in the system [-]
S	target capacity [ADt/d]
SPECCA	specific primary energy consumption for CO ₂ avoided [$MJ_{LHV}/kg_{CO_2,avoided}$]
SPB1	Steam power boiler 1
SPB2	Steam power boiler 2
T	project lifetime [y]
TCI	total capital investment [$£$]
TCR	total capital requirement [$£$]
TIC	total installed cost [$£$]
TPC	total plant cost [$£$]
TPD	Tonne per day
TWh	Tetra watt hour
TRL	Technology readiness level
W_j	brake power requirement/output of equipment j [kW_{el}]
WHRB	Waste heat recovery boiler
X_{ave}	average sorbent conversion [-]
$€/t_{CO_2}$	Euro per tonnes of carbon dioxide
$£/t_{CO_2}$	Pound per tonnes of carbon dioxide
$£/MWh$	Mega watt hour
<i>Greek letters</i>	
η_e	electric efficiency [-]
<i>Subscript</i>	
0	reference value
BRKP	brake power
Calc	calciner
Cap	sugar refinery with CO ₂ capture
Carb	carbonator
COND	condensate
E	electric
Eq	equivalent
ECON	economiser
FP	fuel preparation
HPW	high-pressure water
HRSG	heat recovery steam generator
LS	live steam
Ref	reference sugar refinery without CO ₂ capture
SC	steam cycle
ST	steam turbine

in two times higher net power output comparing to other solvent based techniques such as amine absorption [10], making this process a viable option for energy management teams and power planners in almost every industry.

The invention and recognition of calcium looping dates back to late 20th century when two Japanese scientists, Shimizu [11] and Hirama [12] applied this technology to separate the carbon dioxide from the combustion flue gases. Originally proposed for pulverized coal power plants [13], this technology has found its application as a capture route

in many industries. The approved application of CaL technology in co-firing or standalone biomass power plants [14] has shown that it might be interesting for industries such as sugar which relies on both biomass or hydrocarbon energy to invest on this technology for carbon capturing and removal capture [15]. Owing to the renewable nature of biomass as a fuel, a sugar refinery which relies on its sugar cane bagasse for utility units might be carbon negative as the bagasse will be burned at the same rate it was produced in a sugar refinery. This makes the sugar refinery a carbon negative industry and the sugar as a carbon

negative commodity even if the carbon capture rate in the carbonator is quite low [16].

The application of CaL is verified for conventional hydrocarbon and biomass fired power plants, with or without co-firing. However, its application in energy intensive industries is quite rare. CaL has a strong synergy for integration to many industries including cement, pulp and paper, and the steel making industry as most of these processes require fresh lime in their operating units. De Lena et al. [17] noted that CaL process is a capital-intensive process with cost of CO₂ avoided between 52 and 58.6 €/tCO₂. They have also pointed out that the extra heat that can be recovered from CaL is high enough to compensate for the increased fuel flow rate of the integrated cement kiln. Santos et al. [18] have evaluated this technology in pulp and paper industry and demonstrated that this technology is superior to amine scrubbing CCS technology in terms of cost of CO₂ avoided. This technology could change this industry from an electric importer to an electric exporter with cost of CO₂ avoided 39.0 €/tCO₂ in an integrated lime kiln-calciner design. The application of this technology in other energy intensive industries turns out to also be promising. The economic and technological adaptation of this second-generation CCS into the sugar industry is not well known. In a related study, Neto et al. [16] have demonstrated that the bagasse based boiler can also be integrated with CaL for carbon removal and capturing.

The sugar production is seasonal beginning from early September to the end of January every year. The annual CO_{eq} of refining sugar varies by location and capacity of refinery. For instance, it is around 272 ktonnes for Suiker Unie in Nederland. Around 10 % of the CO₂ emissions are produced in the lime kiln where natural gas is used to for conversion of limestone into lime at high temperatures. The sugar refinery lime kiln can be retrofitted to the CaL calciner to simultaneously regenerate the calcium carbonate for removing the impurities from the refining and carbonation process. In addition, it can also exploit the carbon dioxide that the calciner captured during the purification phase in the carbonation process as the carbon captured during this technology is quite pure (98 % \gg) and possesses the food qualities. The integration into this industry is simpler than any other carbon capturing technology such as amine absorption as it does not need any significant change to the layout and the operation of the sugar refinery. In addition, the sugar refinery shares some commonalities with CaL in terms of materials used which can be processed in other technology.

The UK food industry is responsible for consuming 24 TWh every year, accounting for 9 Mtonnes of carbon dioxide that is around 1 % of all UK CO₂ emissions. In this work, the decarbonisation of a sugar refinery using CaL technology is envisaged using different scenarios that every sugar refinery might rely upon in terms of type of the fuel they use, location and malleability in technology adaptations for use of biomass-based fuels. Moreover, the integration of the CaL to the sugar refinery is investigated to produce fresh lime for carbonation process during the separation of sucrose from fructose, glucose, gums, and impurities. Finally, the techno-economic assessment of the integrated technology is analysed and benchmarked against other technologies. A sensitivity analysis is also performed to explore the uncertainties associated with the simplifying assumption of the economy models.

2. Process integration of sugar refining to calcium looping

A sugar refinery with the capacity of refining 40,000 kg/hr sugar cane (=240 air-dried tonnes) was chosen as the reference plant. The sugar cane from this refinery consists of 59.232 NDt (nitrogen-dried tonnes, 99.61 % dry content) of bagasse per hour, 36.72 ADt/h (air-dried tonnes per hour) of refined sugar to be sold in the market. The process of refining that much sugar is an intensive industry which needs 71 GWh electricity. This equals to 378.02 tonne per day of live steam at 400 °C and 45 bars to run the equipment. The relative quantities of data for running the basic sugar plant was extracted from the technical papers considering different sugar refining scenarios [19]. The sugar refinery is

assumed to be operating 250 days a year and 20 h each working day.

2.1. Sugar refining process

The raw sugar contains other species such as sucrose, glucose and fructose, inorganic ash (mainly calcium and potassium salts) and other organic matters including gums, amino acids, colors from the cane. The impurities must be removed from the sucrose during refining. The refined sugar is composed of 97.5 % sucrose which will be obtained from raw sugar in sugar refineries [20]. The refined sugar is referred to the sugar crystals from the sugar juice or sugar beet which is obtained from raw sugar within four steps (1) affination, 2) carbonation, 3) char filtration and 4) crystallization. A key process which shares some commonality with calcium looping is carbonation as both need CaO and generate CaCO₃ [21]. Fig. 1 gives a schematic of the sugar refining process which is integrated to the CaL for carbon capturing.

2.2. Carbonation

After removing the surface impurities from raw sugar by dissolving it into the syrup and implementation of centrifugal force, the liquor is heated and added to a carbonation tank where a milk of lime (calcium hydroxide, Ca(OH)₂) is added and CO₂ is bubbled through the mixture formed in the tank ahead of any further decolourisation process [22]. The CO₂ in the flue gas of the onsite boilers could act as a source of carbon for the reactions in sugar refinery. The CO₂ is around 7–8 %, 10–12 %, and 12–14 % of natural gas, bagasse and coal combustion exhaust gas [23]. In carbonation tank, the carbonation reaction



occurs under a specific temperature–pressure range and calcium carbonate forms and precipitates with the sugar impurities including gums, polysaccharides, amino acids, and colour within one hour. This process usually takes place in two stages to remove all the impurities and solution from the syrup. In first stage, the large particles and impurities are conglomerated by calcium carbonate particles [24]. In the second stage, the pH of the solution is controlled in the syrup to ensure the complete precipitation of the lime. The calcium carbonate (CaCO₃) precipitate, including the impurities, is removed from the solution in a pressure filtration stage using a polypropylene filter cloth as supporting media and exploiting the calcium carbonate as a filter aid. The carbonation process is a quite successful purification step during the sugar refining which can lead to between 40–50 % colour reduction together with a reduction in ash content of about 20–25 %.

This process of sugar refinery is a key for integration to CaL as it can feed the calciner of calcium looping unit with CaCO₃ and in return, can receive fresh lime (CaO) from the calcination process, thereby establishing a second loop among the feed and product streams of the carbonation tanks with calciner. In addition, the onsite food quality carbon dioxide (98 % \gg) is available and no flue gas aftertreatment is needed to remove the solid particles, residue and SO_x emissions. Owing to the higher percentage of the carbon in the CaL captured stream, lower superficial gas velocities and as a result gas mass flow rates are needed which brings the capital investment for this stage of the sugar refinery down. Due to the nature of the carbon capturing process which is to remove a species from flue gas stream in an exothermic process, heat should be released that could be potentially used to generate steam for sugar refinery utility units. The grade of heat in CaL process is high enough to generate the superheat steam required in the affination and crystallisation process to refine the sugar. Therefore, CaL can save some fuel consumption in boiler utility by compensating for a part of heat from carbonation and exhaust streams of carbonator and calciner. Fig. 2 demonstrates the process of carbonation which is integrated in calcium looping for simultaneous carbon capturing and feeding sugar refinery.

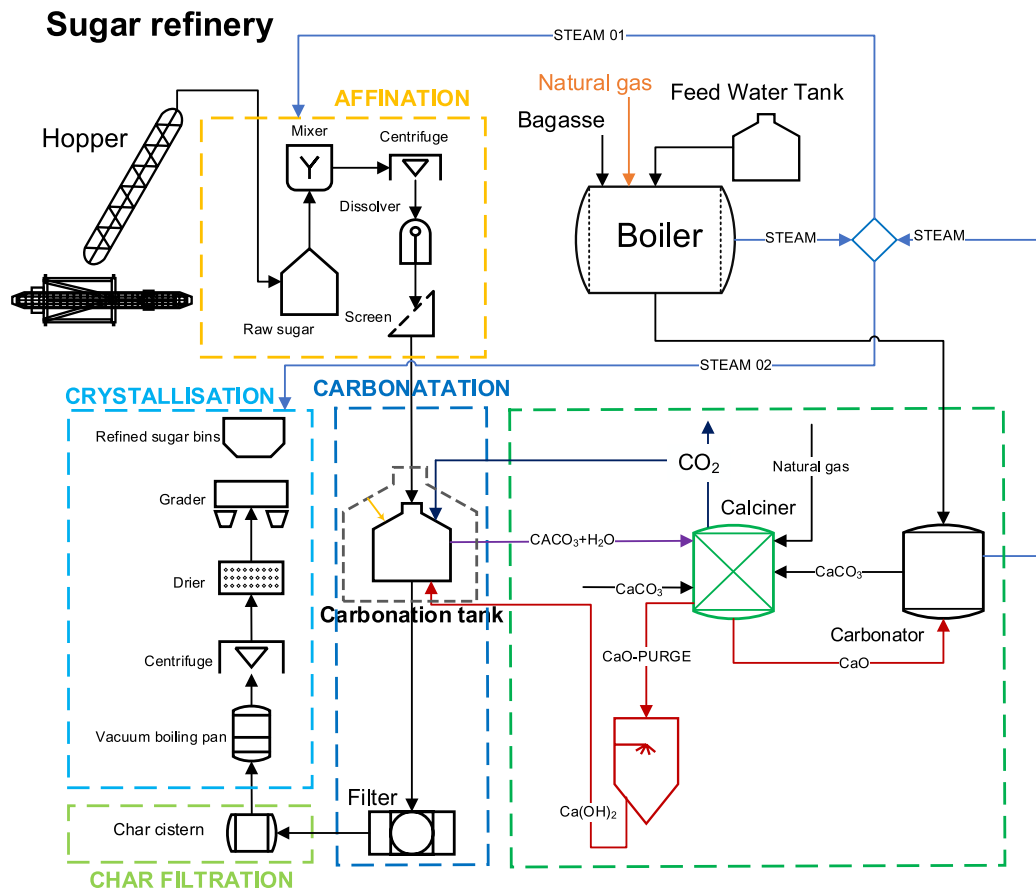


Fig. 1. Process of sugar refinery integrated with calcium looping for simultaneous carbon capture and regeneration of calcium carbonate lime milk for carbonation.

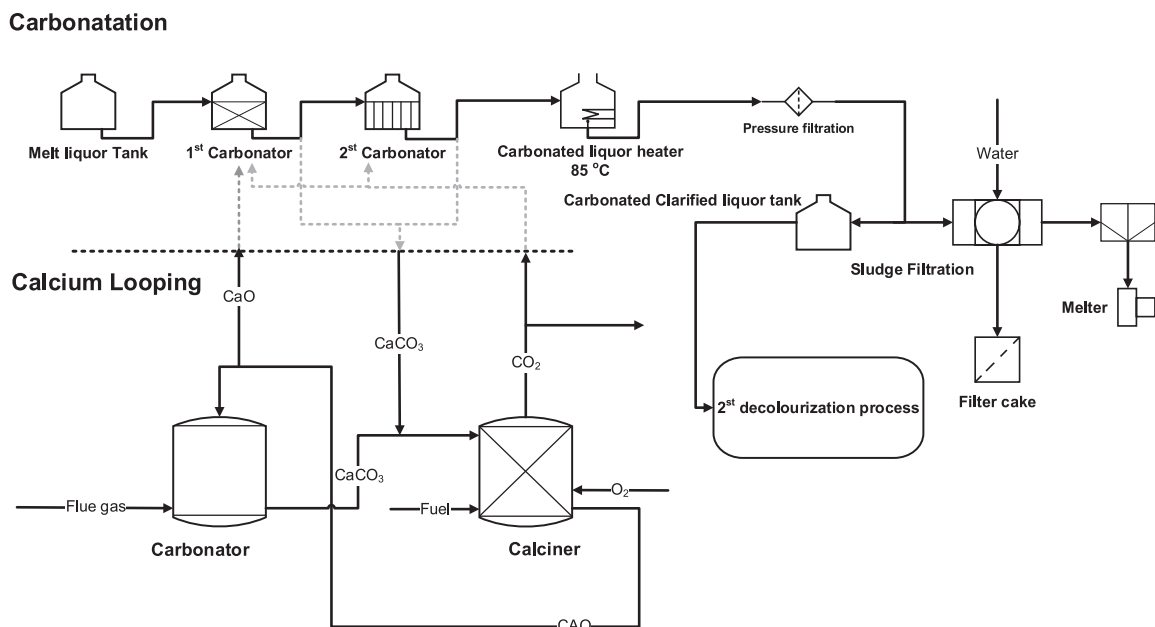


Fig. 2. Detailed schematic for integration of first and second carbonatation tanks with the calciner of the calcium looping process, whereby establishing the second loop for receipt of calcium oxide and carbon dioxide and disposal of CaCO_3 .

2.3. Sugar refinery case studies

For this study, different versions of CaL are envisaged for decarbonisation of sugar refineries. The operation of sugar refineries

integrated with CaL is assessed for different scenarios via Aspen Plus V12.2®. Table 1 enlists the sugar refinery case studies for integration to CaL and deployment of sugar as an either carbon neutral or negative product to the market. The scenarios include both coupled and

Table 1

Case studies for assessment of CaL technology for integration to sugar refineries and deployment of sugar as a carbon neutral or negative commodity.

Case study	Type	Boiler fuel	Reactors	Calciner Fuel	Sugar carbon
Case 1	Coupled	Natural gas	Carbonator/ Calciner	Natural gas	Neutral
Case 2	Coupled	Natural gas	Carbonator/ Calciner	Bagasse (indirect)	Neutral
Case 3	Coupled	Natural gas	Carbonator/ Calciner	Bagasse (direct)	Negative
Case 4	Coupled	Bagasse	Carbonator/ Calciner	Natural gas	Negative
Case 5	Coupled	Bagasse	Carbonator/ Calciner	Bagasse (indirect)	Negative
Case 6	Coupled	Bagasse	Carbonator/ Calciner	Bagasse (direct)	Negative
Case 7	Decoupled	Natural gas	Carbonator	NA	Neutral
Case 8	Decoupled	Bagasse	Carbonator	NA	Negative

decoupled CaL process. Coupled versions is the conventional CaL in which flue gas CO₂ is absorbed in the carbonator and released in the calciner during oxyfuel combustion whereas decoupled version is a new technology license in which the carbonator is installed in the sugar refinery premise and calciner is somewhere faraway in which facilities for carbon compression and storage is more accessible. The decoupled technology of CaL is of more interest where there is limited land for installing both carbonator/calciner or the possibility of burning biomass-based fuel is not available on the sugar refinery for limestone calcination.

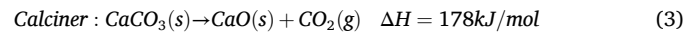
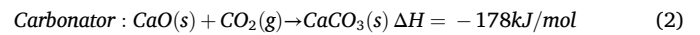
The schematic of the carbonator/calciner and boiler units are presented in Fig. 3 for different case studies. A reference sugar refinery which can process 40,000 kg/h of sugar cane is considered as the base line of decarbonisation. Table 2 gives the constituents and properties of the sugar cane, the feedstock for the sugar refinery and the bagasse- a sugar cane residue which can be used as a biomass fuel in utility boilers and calciners.

Two types of sugar refineries are considered based on their boiler fuel type-1) bagasse and 2) natural gas. Boilers generate the required steam for refinery and onsite power equipments. Around 31 % of the sugar cane input to the refinery is considered to be bagasse [27,28]-a sugar cane biomass residue which can feed the boiler to generate the steam for the refinery. This quantity of fuel can generate the required steam both as a feedstock and onsite steam cycle for electricity generation in the refining process. Two grades of steam are required during the refining process- 1) high pressure steam at 45 bars and 400 °C which is fed to the turbines for electrical purpose- 2) low pressure steam at 5 bars and 280 °C which is fed to the process units for washing, maintenance, and refining. The second grade is an exhaust from the steam turbine which processes the high-pressure steam. Therefore, the energy integration network between the CaL and utility boilers is designed to generate only the first grade of steam-high pressure steam to be deployed in their refinery steam turbine.

For this study, the boiler units including their steam generator are modelled in Aspen Plus. The sugar refinery is equipped with three boilers to generate electricity and steam for the refining process- 1) waste heat recovery boiler (WHRB) which is to burn the fuel with the exhaust gas of the onsite utility gas turbines, 2) steam power boiler 1 (SPB1)-this boiler is to generate superheat steam for refining the sugar, 3) steam power boiler 2 (SPB2)). The exhaust section of these three boilers is mixed and conducted to the design CaL unit for carbon capturing with appropriate material and energy integration. Table 3 gives the flue gas pressure, temperature, mass flow rate and composition of the utility boilers SPB1, SPB2 and WHRB.

3. Process model simulations

Boiler units are modelled to include combustion and high-pressure superheat steam generation for power and sugar refining. To model the combustion in the boilers, Gibbs equilibrium reactors are chosen to show the equilibrium species concentration of combustion products. The water jacketed boiler is envisaged in sugar refineries, and no heat lost to the environment is considered in the modelling of heat transfer to the water. In the process designed for decarbonisation, it is envisaged that the flue gas from WHRB and SPBs boilers are merged into one stream (composition included in Table 3) and via a fan blower directed into the carbonator, a fluidised bed fed with CaO pellets, where the reaction (1) takes place. In the carbonator, the CO₂ will be captured by lime from the flue gas which was regenerated in boiler units as CaO turns into CaCO₃. The carbonated lime (CaCO₃) will be transferred to the calciner by means of an electric conveyor for regeneration through reaction (2).



This process requires heat which is provided directly or indirectly from natural gas and bagasse combustion. Cases 1 and 4 are designed so that their calciners directly burn the natural gas with pure oxygen in oxyfuel technology to regenerate the lime (CaO). Calcination in case 2 will employ an indirect heat transfer from bagasse combustion in a conventional air combustion. Calciners in cases 3 and 6 directly burn bagasse via oxyfuel technology. Case 5 uses both direct oxyfuel and indirect conventional combustion technologies in the calcination process. Case 6 calciner also burns a specific quantities of natural gas to compensate for the shortage of bagasse obtained from factorised sugar. In cases 1–6, the sorbent (CaO) circulates within the carbonator-calciner in a frequent series of cycles. Cases 7 and 8 do not include the calcination process and fresh sorbent should be provided to capture the carbon from the merged flue gas. The process developed to integrate the carbon capture unit to the reference sugar refinery involves a CaL process, a steam cycle and a CO₂ compression unit (CCU). Fig. 4 demonstrates the process flow diagram of the designed system for carbon capturing, power generation and CO₂ compression, CASE1 (other case studies flow chart diagram are given in the Supplementary file). The calcium looping model was developed based on a previous model [15] and validated with experimental data for a 1.7 MW_{th} pilot plant at INCAR-CSIC [29]. A stoichiometric reactor is employed to model the carbonation process and carbon capturing from the boiler's flue gas streams. A Gibbs reactor is utilised to model the calcination process using both indirect and direct heat. In the integrated model, a stream for lime-mud is embedded in the calciner to receive the byproduct of the carbonation tanks, process them and make a fresh calcium oxide for the carbonator. As the conversion drop of sorbent occurs during frequent carbonation-calcination cycles [30,31], a fresh limestone stream (F₀) is also considered in the calciner to keep the average sorbent conversion in the carbonator constant. Here, limestone is chosen as the sorbent as it best represents the lime mud behavior. The limestone is used to balance the lime conversion drop [32]. The maximum average conversion (X_{ave}) depends on the carbonation (f_{carb}) and calcination extent (f_{calc}), the make-up rate (F₀), the solid looping rate (F_R) and the sorbent characteristics (a₁, a₂, f₁, f₂ and b). X_{ave} is estimated using the model proposed by Rodríguez et al. [33] as the following:

$$X_{ave} = (F_0 + F_R r_0) f_{calc} \left[\frac{a_1 f_1^2}{F_0 + F_R f_{carb} f_{calc} (1 - f_1)} + \frac{a_2 f_2^2}{F_0 + F_R f_{carb} f_{calc} (1 - f_2)} + \frac{b}{F_0} \right] \quad (4)$$

The sorbent parameters are used from the 1.7 MW_{th} INCAR-CSIC pilot

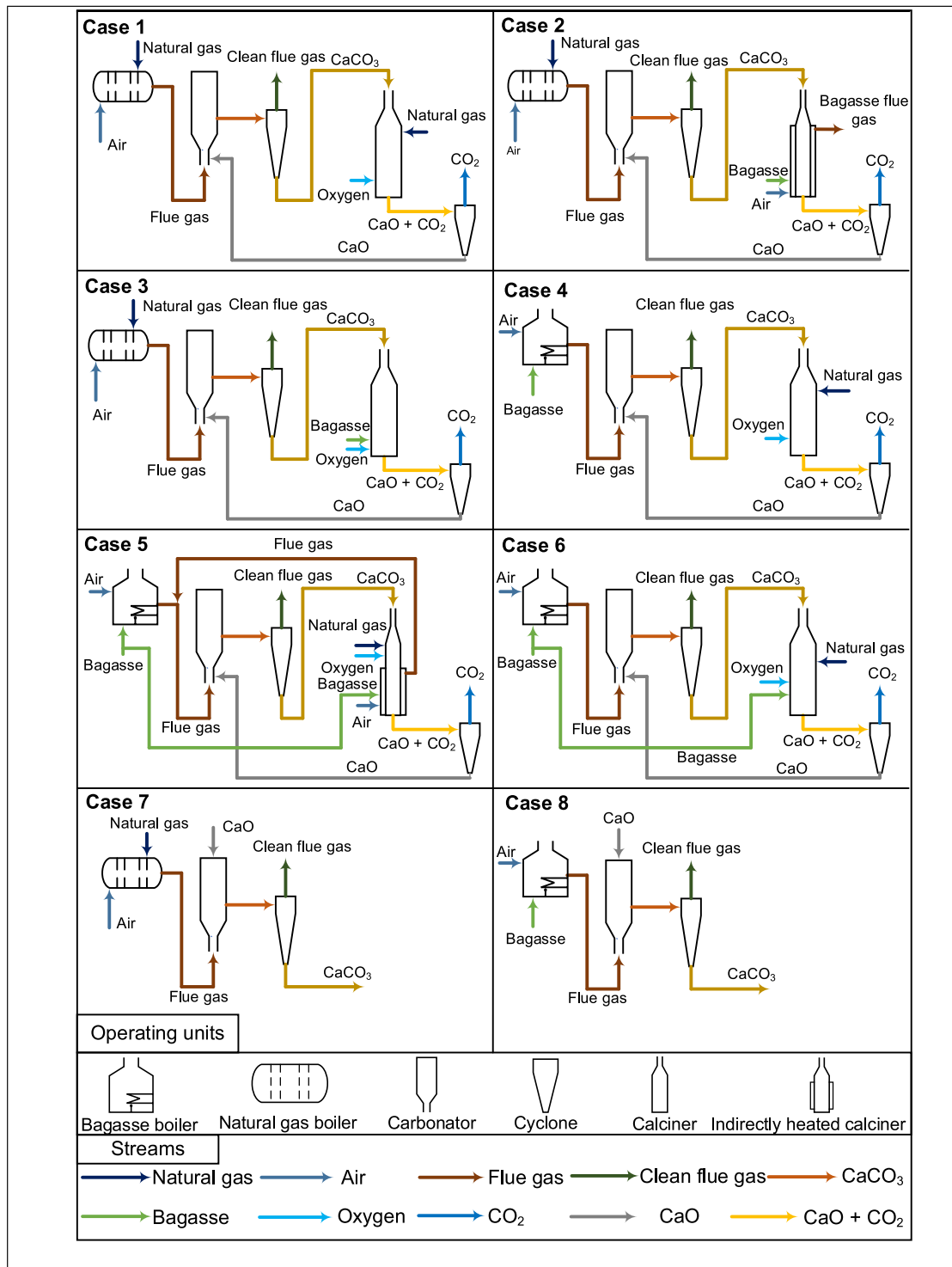


Fig. 3. Process flow diagram of carbonator integration to the reference case sugar refinery boiler unit and calciner integration to the carbonation and fuel delivery system.

plant [29]. The gas stream from the calciner is transferred to the steam cycle for heat recovery, then dehydrated and sent to compression and storage unit. Depending on the studied case, the gas stream from the calciner could include different composition of CO₂ and vapor which needs to be dehydrated before CO₂ compression and storage. Water dehydration is not needed in case 2 as calciner works from indirect heat of combustion. In that calciner, outlet gas stream only includes the gaseous product of reaction 3 which is carbon dioxide. In other cases, 1, 3–6, the water is in both the carbonator byproducts as well as that from

oxyfuel combustion of either natural gas or bagasse or both. After heat recovery and dehydration, the purified CO₂ ($\cong 99\%$) is compressed to a pressure well above the critical pressure of carbon dioxide ($\cong 80$ bar) in a multi-stage compressor. For storage, the CO₂ is cooled down to 25 °C and compressed at 110 bar to before it transmitted towards the storage pipelines [34].

A heat exchanger network is designed for heat recovery from the CaL unit, thereby utilising it for steam generation. High grade heat can be generated in the carbonator and can be recovered from both the

Table 2
Properties and composition of sugar cane for the baseline study.

	Property	Value	Unit	Ref
Sugarcane properties	Pressure	1.01325	bar	
	Temperature	10	°C	
	Mass flow rate	40,000	kg/h	
Sugarcane mass composition	Water	68.53	%wt	[25]
	Sucrose	14.17	%wt	
	Non-Sucrose	2.24	%wt	
	Fiber	15.06	%wt	
Bagasse properties	Pressure	1.01325	bar	[26]
	Temperature	10	°C	
	Bagasse to cane ratio	30.85	%	
	Mass flow rate	3085	kg/h	
Bagasse Composition	Water	50.95	%wt	[25]
	Sucrose	1.49	%wt	
	Non-Sucrose	2.16	%wt	
	Fiber	47.39	%wt	

carbonator and calciner exhaust gas. This extra heat can be used to generate additional steam during the carbon capture process, thereby either compensating for boiler duty in generation of steam for refining the sugar or it can be used in an additional steam cycle to generate more electricity transforming the sugar industry to an electricity exporter industry. In this work, two scenarios have been considered 1) the first one is that the boiler is working with 100 % duty during operation of CaL and extra heat is implemented in an additional steam cycle to generate more electricity, 2) the second one is that the heat of CaL (carbonation heat + that recovered from carbonator and calciner exhaust gases) is implemented to compensate for a specific part of boilers duty, therefore balancing the reductions in generated steam for the boilers. In the second case, an optimisation point for case studies 1–6 is found by reducing the duty of the SPW1 boiler and seeing at which specific boiler fuel consumption CaL extra heat can balance the reduction in the boiler duty. Depending on the specified scenarios, either a steam cycle or steam generators are considered for installation in the integrated sugar refinery plant.

The designed steam cycle is modelled based on a superheated Rankine cycle without reheating. It includes a heat recovery steam generator (HRSG), one gas–gas heat exchanger (superheat steam heater (SSH), and three gas–liquid heat exchangers (condensate heater (CONDH), high pressure water heater (HPWH), and ECONOMISER). SSH and HPWH receive the heat from the exhaust gas of the calciner ($\text{CO}_2 + \text{H}_2\text{O}$). ECONOMISER and CONDH recover the heat of the flue gas coming out of the carbonator. CONDH is used to preheat the circulating water at an intermediate pressure of 11 bar. HPWH is doing the same as CONDH on the water at the desired steam pressure 45 bar. ECONOMISER produces saturated steam at 45 bars and HRSG generates saturated vapor at 45 bars and 257.5 °C. SSH generates live steam at 45 bar and 400 °C to feed the steam turbines. The live steam generates electricity with four steam turbines at 25, 11, 5.21 and 0.69 bar.

The main streams for modelling of the calcium looping include

Table 3

Flue gas properties and composition of boilers SPB1, SPB2 and WHRB feeding with natural gas and bagasse (Data obtained from Aspen plus simulation for Gibbs reactor in operating conditions of WHRB and SP boilers).

Parameter	Natural gas boiler			Bagasse boiler		
	SPB1-2	WHRB	Overall (exhaust)	SPB1-2	WHRB	Overall (exhaust)
Pressure [bar]	1.2	2	1.5	1.2	2	1.5
Temperature [°C]	135	110	120	138	115	120
Mass flow [KTPD]	402.22	149.21	551.43	361.30	134.03	495.33
N ₂ [%vol]	73.62	70.42	72.37	72.21	70.12	70.47
O ₂ [%vol]	2.48	3.06	2.63	3.40	6.00	4.46
H ₂ O [%vol]	15.61	18.14	16.25	9.91	11.51	10.17
CO ₂ [%vol]	8.30	8.36	8.28	13.87	12.35	13.24
SO _x [ppm]	0	0	0	39	76	48
NO _x [ppm]	27	51	33	70	93	75

natural gas and bagasse, air and pure oxygen for combustion, water for stream generation, limestone for balance the conversion, by product calcium carbonate from carbonator, natural gas and bagasse composition and fresh lime (CaO) in case 7 and 8. Table 4 enlists the stream properties and compositions which are used for the modelling.

The model for CaL also includes some design consideration and thermodynamic assumption for the installed equipment. The model includes boiler units, CaL unit, steam cycle and CO₂ compression unit which comprise of reactors, heat exchangers, fan blowers, pumps, turbines, compressor, flash drums, mixer, and splitters. The model assumptions for installation are given in Table 5.

4. Energy optimisation

The energy network of process for case studies 1–8 is optimised for the best operation of the integrated sugar refinery + calcium looping carbon capturing unit. The extra heat that the carbonation and exhaust section of the carbonator and calciner produce, is considered to balance a part of boiler duty. To this end, an objective function (Q_{overall}) forms which gives the total energy for the process of refining. The following function is defined, including the duty of the energy systems:

$$Q_{\text{overall}} = q_{\text{boiler}} + q_{\text{carbonation}} + q_{\text{carbonator-exhaust}} + q_{\text{calciner-exhaust}} \quad (5)$$

The q_{boiler} , $q_{\text{carbonation}}$, $q_{\text{carbonator-exhaust}}$ and $q_{\text{calciner-exhaust}}$ represent the duty of the boiler, duty of carbonator, maximum extractable energy from the carbonator and calciner flue gas.

q_{boiler} is the duty of boilers overall which is 71 GWh for this sugar refinery without CaL optimization. This subfunction depends on the mass flow rate of bagasse or natural gas burned in boilers.

$q_{\text{carbonation}}$ is the carbonation heat coming out of the adsorption and endothermic reactions of CaO with CO₂. This heat is captured from the carbonator towers using water jacket towers so that the final exhaust temperature is 120 °C. This permits facile vented from stock.

$q_{\text{carbonator-exhaust}}$ is the heat recovered from the carbonator exhaust which is the boiler exhaust gas with 90 % of carbon dioxide captured at 650 °C.

q_{calciner} is the heat that can be removed from the calciner exhaust which is a mixture of CO₂, H₂O and O₂ at 900 °C.

The objective of energy optimisation is to find a mass flow rate for fuels burning in boiler units that all energy overall obtained from boiler-carbonator-calciner system reach 71 GWh which is required to power the sugar refinery. Energy optimisation route for all case studies 1–8 are comprehensively explained in the [supplementary material](#) of this manuscript. Technoeconomic assessment is done for energy optimized case studies and their results are given in the following.

5. Technoeconomic feasibility assessment

This section provides a plan for the technoeconomic assessment of the sugar refinery with and without CaL thereby, delivering a new proof

CASE 1

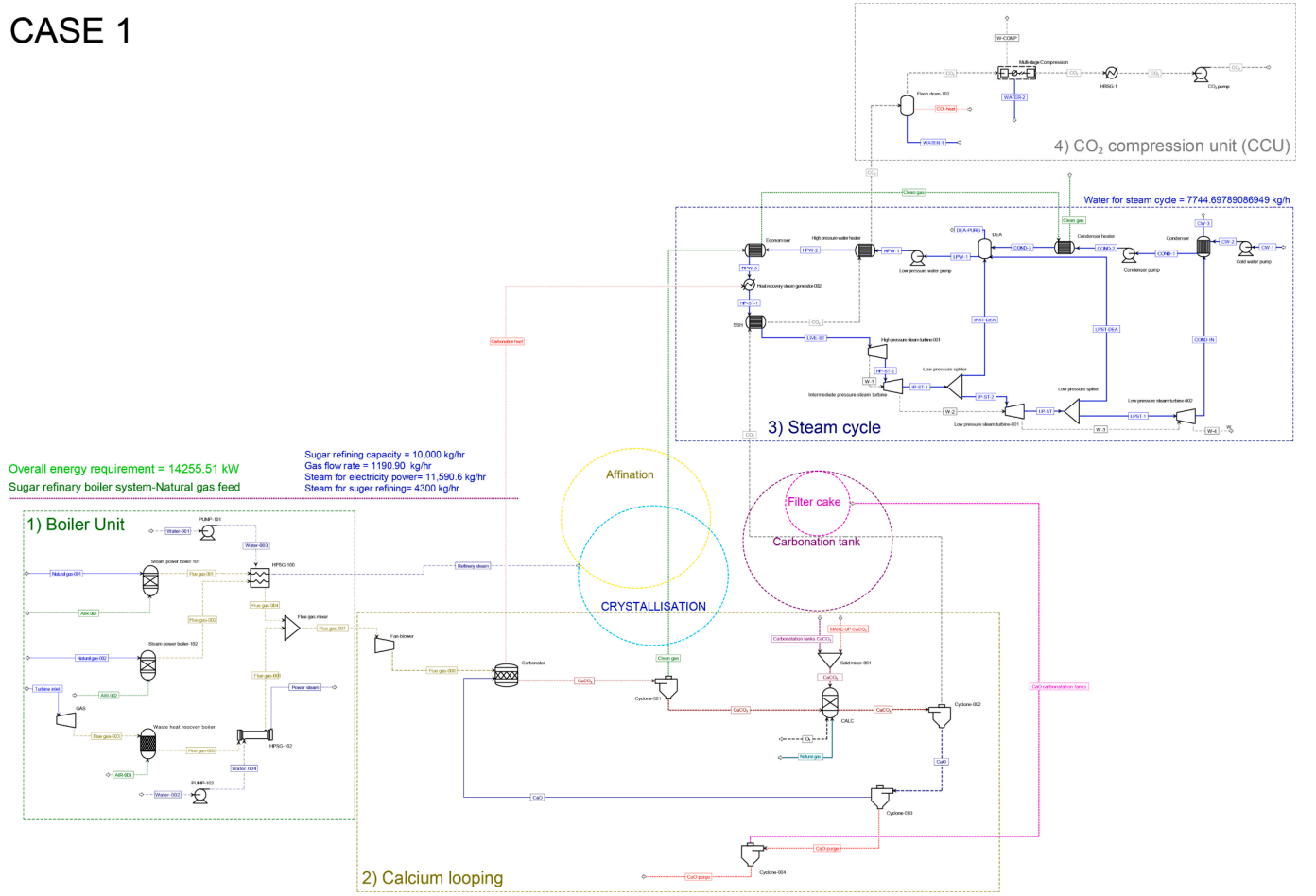


Fig. 4. Process flow diagram of the case 1 sugar refinery integrated into the CaL including 1) boiler unit, 2) Calcium looping, 3) steam cycle and 3) CO₂ compression unit.

of concepts for calcium looping carbon capturing technology in sugar refineries. The process models built for the sugar refinery with bagasse and natural gas boiler units presented in 8 case studies are used here to show the techno-economic assessment. To this end, the thermodynamic and economic performance indicators are defined as the following:

5.1. Thermodynamic performance indicators

Parameters including equivalent fuel consumption (q_{net}), Eq (6), equivalent CO₂ emissions ($e_{CO_2,eq}$), Eq (7), and specific primary energy consumption for CO₂ avoided (SPECCA), Eq (8) are employed to assess the synergy of CaL to the sugar refinery:

$$q_{eq} = q + \frac{3600 \cdot P_e}{\eta_e} \quad (6)$$

The equivalent fuel consumption is the sum of direct (q) and indirect fuel consumption ($3600 \cdot P_e / \eta_e$) which is related to the electricity imported (net power output, P_e) and electricity efficiency.

$$e_{CO_2,eq} = e_{CO_2} + P_e \cdot e_{CO_2,e} \quad (7)$$

The equivalent CO₂ emissions, $e_{CO_2,eq}$, is the summation of direct, $e_{CO_2,eq}$, and indirect emissions, $P_e \cdot e_{CO_2,e}$. The indirect CO₂ emissions are related to the electricity imported from the grid and emission of the power plant.

If the integrated sugar refinery is to become a net electricity producer, the indirect emissions, $P_e \cdot e_{CO_2,e}$ become negative, resulting in negative CO₂ emissions (same as in cement [17] and pulp and paper

industry [18]).

$$SPECCA = \frac{q_{eq,cap} - q_{eq,ref}}{e_{CO_2,eq,ref} - e_{CO_2,eq,cap}} \quad (8)$$

The subscripts ref and cap indicate the reference sugar refinery without CO₂ capture and with CO₂ capture, respectively. The specific primary energy consumption for CO₂ avoided (SPECCA) depends on the power generation, electric efficiency, and specific CO₂ emissions. The indirect CO₂ emission is done based on the average non-CHP energy mix in 27 EU Member States and the UK in 2022 ($e_{CO_2,e} = 233 \text{ kg}_{CO_2}/\text{MW}_{el}\text{h}$ and $\eta_e = 45.9\%$).

5.2. Economic performance indicators

The techno-economic assessment of the integrated sugar refinery + CaL is performed based on the project plan which is given in Table 6. The levelized cost of sugar (LCOS), levelized cost of electricity (LCOE) and the cost of CO₂ avoided (AC) are calculated using net present value (NPV):

$$NPV = \sum_{t=1}^n \frac{CF_t}{(1+r)^t} - TCR \quad (9)$$

where CF_t denotes the discounted cash flow during the project lifetime (t), TCR indicates total capital investment of the project with interest rate (r).

Table 4

Stream properties and composition of flue gases, generated steam, make-up limestone, exhaust calcium carbonate, fresh lime, natural gas and bagasse implemented to construct the CaL model.

Stream	Parameter	Value	Unit	REF	
Flue gas natural gas	Natural gas	3009	kt/y	[34]	
	Bagasse	4107	kt/y		
	Temperature	120	°C		
Limestone (make-up)	Pressure	1.01325	bar		
	CaCO ₃	95	%wt	[35]	
	MgCO ₃	3.5	%wt		
	SiO ₂	0.6	%wt		
	Fe ₂ O ₃	0.4	%wt		
Carbonator calcium carbonate	Al ₂ O ₃	0.5	%wt		
	CaCO ₃	14.49	%wt	[25]	
	SiO ₂	0.533	%wt		
	MgO	0.0915	%wt		
	Fe ₂ O ₃	0.061	%wt		
	Al ₂ O ₃	0.0762	%wt		
	H ₂ O	84.75	%wt		
Fresh lime CaO	CaO	92.15	%wt	[35]	
	CaCO ₃	1.66	%wt		
	MgO	0.16	%wt		
	SiO ₂	5	%wt		
	Fe ₂ O ₃	0.5	%wt		
	Al ₂ O ₃	0.52	%wt		
Air	N ₂	77.32	%wt	[36]	
	O ₂	20.75	%wt		
	H ₂ O	0.97	%wt		
	CO ₂	0.04	%wt		
	Ar	0.92	%wt		
	Temperature	10	°C		
	Pressure	1.01325	bar		
	Oxygen	N ₂	4	%wt	[37]
		O ₂	95	%wt	
		Ar	1	%wt	
Temperature		10	°C		
Natural gas	Pressure	1.01325	bar		
	N ₂	3.6412	%mol	[38]	
	CO ₂	0.7739	%mol		
	CH ₄	91.6956	%mol		
	C ₂ H ₆	2.9963	%mol		
	C ₃ H ₈	0.5853	%mol		
	i-C ₄ H ₁₀	0.0892	%mol		
	C ₄ H ₁₀	0.1389	%mol		
	C ₅ H ₁₀	0.0397	%mol		
	i-C ₅ H ₁₀	0.0397	%mol		
	Temperature	10	°C		
	Pressure	1.5	bar		
	Bagasse	H ₂ O	10.39	%wt	[16]
C		43.59	%wt		
H ₂		25.26	%wt		
O		38.37	%wt		
N		0.14	%wt		
Cl		0.02	%wt		
S		0.04	%wt		
ASH		2.19	%wt		
LHV		15.62	kJ/kg		
HHV		17.02	kJ/kg		

5.3. Sugar refinery cost estimation

The sugar refinery is assumed to operate 250 days a year with sugar refining capacity of 240 TPD. The chemical engineering plant cost index is employed to estimate the cost of sugar refining. The reference capital cost for sugar refinery is transferred to the capacity of studied sugar refinery using the following semi-empirical correlation:

$$\frac{C}{C_0} = \left(\frac{S}{S_0}\right)^n \quad (10)$$

where C indicates the actual capital cost and S denotes the target capacity. Subscript 0 refers to the reference value for sugar refinery costs in 1957 which was estimated M£ 1.08. The capital cost of the sugar refinery is adjusted for the year 2022 using chemical engineering plant cost index

Table 5

Technical data and assumption for sugar refinery boiler unit, carbonation and calcium looping integrated system and steam cycle equipment.

Unit operation	Parameter and value	Value	Unit
Boiler Unit			
Boiler	Pressure	1.01325	bar
	CO level (natural gas and bagasse)	0	%vol, dry
	O ₂ in natural gas boiler	2.65	%vol, dry
	O ₂ in bagasse boiler	4.87	%vol, dry
Chemical looping			
Carbonator	Pressure	1.01325b	bar
	Temperature	650	°C
	Carbonated sorbent fraction	0.7	[-]
	CO ₂ capture percentage	90	%
Calciner	Pressure	1	bar
	Temperature	900	°C
	Calciner sorbent fraction	0.95	[-]
	Excess oxygen	1	%vol, dry
	Relative make-up	0.04	[-]
Steam cycle			
Live steam	Temperature	400	°C
	Pressure	45	bar
High pressure turbine	Outlet pressure	25	bar
	Isentropic efficiency	92	[%]
	Mechanical efficiency	99.8	[%]
Intermediate pressure turbine	Outlet pressure	11	bar
	Isentropic efficiency	77.5	[%]
	Mechanical efficiency	96.5	[%]
Low pressure turbine	Outlet pressure	5.21	bar
	Isentropic efficiency	80	[%]
	Mechanical efficiency	96.5	[%]
Second low pressure turbine	Outlet pressure	0.69	bar
	Isentropic efficiency	60	[%]
	Mechanical efficiency	98	[%]
Condenser	Feed water temperature	10	°C
	CO₂ compression unit		
Multi-stage compressors	Polytropic efficiency	80	[%]
	Mechanical efficiency	99.6	[%]
	Intercooler temperature	40	°C
CO ₂ pump	Isentropic efficiency	80	[%]
	Mechanical efficiency	99.6	[%]
CO ₂ pump	Pressure	110	bar
	Temperature	25	°C
	Purity	≅ 99	[%]

Table 6

The project plant considered for integration of CaL to the sugar refinery [39,40].

Variable	Value	Unit
Sugar refinery + CaL lifetime	25	Year
Operational year	250	days
Capacity factor	80	%
Project interest rate	8.8	%
CO ₂ emission allowance price	28	£/tCO ₂
Average GBP/EUR exchange rate 2022	1.17	£/€

(CEPCI) as the following:

$$C_{2017} = C_{1957} \frac{CEPCI_{2022}}{CEPCI_{1957}} \quad (11)$$

After estimation of fixed capital costs from CEPCI index, the total capital investment (TCI) costs are estimated from the sum of the fixed capital investment and working capital. Afterwards, the total sugar cost is estimated using the cost variable of TCI. Finally, total revenue of the sugar refinery integrated with CaL is estimated using total product costs. The economic model parameter for estimation of CAPEX and OPEX are represented in Table 7.

5.4. Calcium looping cost estimation

The calcium looping unit involves a fan blower, carbonator, calciner, cyclones, air separation unit and CO₂ compression unit. Therefore, the total capital cost of the calcium looping is estimated from the following:

$$TCR_{cap} = C_{CaL} + C_{ASU} + C_{CCU} + C_{SC} \quad (12)$$

where C_{CaL}, C_{ASU}, C_{CCU}, and C_{SC} indicate the costs associated with CaL, air separation unit, CO₂ compression unit and steam cycle. The total capital investment costs for CaL unit and steam cycle are estimated based on their operational units:

$$C_{CaL} = (1 + i_{p\&c})(C_{FB} + C_{Carb} + C_{Calc} + C_{FP} + C_{Cyclone} + C_{HX}) \quad (13)$$

where C_{FB}, C_{Carb}, C_{Calc}, C_{FP}, C_{Cyclone} and C_{HX} denote the costs of flue gas, fan blower, carbonator, calciner, fuel preparation system, cyclone, and heat exchangers. The heat exchangers in CaL unit are only applied in decoupled versions where preheating of lime and flue gas streams is needed to reach the required carbonation temperature. The cost of piping and integrations for operation of CaL to the reference sugar refinery is considered 5 % of the total capital costs of CaL (C_{CaL}) [41].

Table 7
Economy model parameter for estimation of sugar refining price and integrated with CaL.

Cost parameter	Value
Total capital investment	
I) Direct costs	
A) Material and labor	
a) Purchased equipment costs	30 % FCI
b) Installation, insulation, and painting	35 % purchased equipment costs
c) Instrumentation and	10 % purchased equipment costs
d) Piping installed	40 % purchased equipment costs
e) Electrical installed	25 % purchased equipment costs
B) Building, Process, and auxiliary	30 % purchased equipment costs
C) Service facilities and yard improvements	50 % purchased equipment costs
D) Land	6 % purchased equipment costs
II) Indirect costs	
A) Engineering and supervision	10 % direct costs
B) Construction expenses and contractor's fee	15 % direct costs
C) Contingency	8 % FCI
III) Fixed capital investment (FCI)	Direct + Indirect costs
IV) Working capital (WC)	15 % TCI
V) Total capital investment (TCI)	FCI + WC
Total product cost	
I) Manufacturing costs	
A) Fixed charges	
a) Depreciation	10% FCI+2.5% building, process, and auxiliary
b) Local taxes	2% FCI
c) Insurances	0.6% FCI
d) Rent	10% building, process, and auxiliary
B) Direct production cost	
a) Raw materials	25% Total product costs
b) Operating labour	12% Total product costs
c) Direct supervisory and clerical labour	12% Operating labour
d) Utilities	12% Total product costs
e) Maintenance and repair	5% FC
f) Operating supplies	15% maintenance and repair
g) Laboratory charges	14% Operating labour
h) Patent and royalties	2% total product costs
C) Plant overheat costs	10% total product costs
II) General expense	
A) Administrative costs	5% Total product costs
B) Administrative costs	15% Total product costs
C) Research and development costs	5% Total product costs
D) Financing (interest)	5% TCI
III) Total product costs	Manufacturing costs + general expenses

$$C_{SC} = C_{HPWH} + C_{ECON} + C_{SSH} + C_{CONDH} + C_{HRSG} + C_{ST} + C_{PUMP} \quad (14)$$

The cost of steam cycle (eq (12) includes the equipment costs:liquid–gas (C_{HPWH}, C_{CONDH}, C_{ECON}) and vapor–gas heat exchangers (C_{SSH}, C_{HRSG}), pumps (C_{PUMP}), and steam turbine (C_{ST}). To estimate equipment costs as a part of TCI, empirical correlations available in the literature are used. Table 8 gives correlations for equipment and operating costs of

Table 8
Equipment costs correlations and operating costs for integration of sugar refinery to the CaL.

Cost variable	Cost correlation [€]	Ref
Unit operations		
Fan [Brake power requirement, W _{Fan,BRK} (kW _{el})]	$C_{Fan} = 88199 \left(\frac{\dot{W}_{Fan,BRK}}{445} \right)^{0.67}$	[41]
Carbonator	$C_{Carb} = 14180 \left(\dot{Q}_{carb} \right)^{0.67}$	[42]
Cyclone	$C_{Cyc} = 3.29 \times 10^{-9} D_{cyc}^2 + 2.26 \times 10^{-6} D_{cyc} + 0.013$	[17]
Fuel preparation system	$C_{FP} = 14158479 \left(\dot{m}_{FP} \right)^{0.24}$	[43]
Air separation unit	$C_{ASU} = 2.501 \times 10^7 \left(\frac{\dot{m}_{O_2}}{28.9} \right)^{0.7}$	[44]
Calciner	$C_{Calc} = 11231 \left(\dot{Q}_{cal} \right)^{0.67}$	[42]
Steam turbine [Brake power output, W _{ST,BRK} (kW _{el})]	$C_{ST} = 3,200.25 \left(\dot{W}_{ST,BRK} \right)^{0.7} - 52.39 \left(\dot{W}_{ST,BRK} \right)^{0.95}$	[45]
Steam cycle		
Heat exchanger high-pressure water [Heat exchange area, A _{HPW} (m ²)]	$C_{HPW} = 111 \left(\frac{A_{HPW}}{0.093} \right)$	[46]
Economiser [Heat exchange area, A _{ECON} (m ²)]	$C_{ECON} = 111 \left(\frac{A_{ECON}}{0.093} \right)$	[46]
Heat exchanger live steam [Heat exchange area, A _{LS} (m ²)]	$C_{LS} = 1957 (A_{LS})^{0.6}$	[47]
Heat exchanger condensate [Heat exchange area, A _{COND} (m ²)]	$C_{COND} = 111 \left(\frac{A_{COND}}{0.093} \right)$	[46]
Heat recovery steam generator [Heat exchange area, A _{HRSG} (m ²)]	$C_{HRSG} = 111 \left(\frac{A_{HRSG}}{0.093} \right)$	[46]
CO ₂ compression unit [brake power requirement, W _{CCU} (kW _{el})]	$C_{CCU} = 1.05054 \times 10^7 \left(\frac{\dot{W}_{CCU,BRK}}{13000} \right)^{0.67}$	[48]
Operating costs		
Piping and integration cost indicator [%]	5 %TCI	[41]
Fixed operating costs	1 %TCR	[39,40]
Variable operating costs	2 %TCR	[39,40]
Limestone price [€/t]	6	[39,40,49]
Natural gas price [€/MWh]	103.01	[50]
Electricity exported to the grid [€/MWh]	270	[50]
CO ₂ transport and storage cost [€/t]	7	[51]

necessary equipment for integration of the CaL to the sugar refinery. The operating cost for CaL also consists of the fixed and variable operating costs and price for fuel (bagasse, natural gas), limestone, CO₂ transport and storage. The revenue from the integrated units comprises of sugar, exported electricity, and other savings from integration of refinery to the CaL (lime for carbonation tanks and CO₂).

The cost of CO₂ avoided (AC) is also defined as the following to calculate a comparative figure between the levelised cost of sugar (LCOS) when CO₂ is captured to when the CO₂ is released to the atmosphere. This factor depends on the annual sugar production rate, \dot{m}_{sugar} and levelised cost of sugar and carbon equivalent emissions ($e_{CO_2,eq,ref}$ and $e_{CO_2,eq,cap}$).

$$AC = \frac{[LCOS]_{cap} - [LCOS]_{ref}}{e_{CO_2,eq,ref} - e_{CO_2,eq,cap}} \quad (15)$$

6. Results and discussion

The energy optimisation of the unit is performed here to see how the CaL contributes to the energy generation of the sugar refinery power plant. Fig. 5 gives a bar chart demonstrating the energy savings in the operation of boiler by integration of CaL to the reference sugar refinery based on the case studies defined earlier in Table 1. The exhaust energy that can be used to generate steam and contribute to the operation of the boiler is the energy of carbonation, exhaust gas from carbonator and CO₂ streams from the calciner. The energy of carbonation ideally exists at 650 °C and comes from the carbonation reaction. The exhaust gas energy can be obtained by cooling the carbonator flue gas from 650 to 120 °C and calciner CO₂ from 900 to 70 °C when the thermodynamic constraint allows. Note that when the bagasse is used indirectly in the calciner for carbon neutral sugar, the energy can be obtained also from the exhaust gas of bagasse cooling from temperature of the calciner 900 °C to flue gas regular temperatures 120 °C.

The findings here demonstrate that the integration of CaL to the reference sugar refinery leads to around 30–40 % energy and fuel saving in the boiler units when the coupled versions of CaL are considered. In the case of decoupled versions, the energy savings should be 15–20 %. Cases 1, 2, 3, 4, 5, and 6 result in 34 %, 45 %, 32 %, 42 %, 98 % and 38 % saving in the operation of the boilers. Case 7 and 8 culminate in the energy and fuel savings 17 % and 21 %, respectively.

One important finding here for boilers using bagasse is that it is technologically feasible to indirectly use bagasse in the calciner. Results for case 5 state 98 % savings (the bar for case 5 is not completely shown in Fig. 5). Findings from case 5 demonstrate that the bagasse obtained from processing the sugar cane is not enough for indirect application in a hot air combustion technology used for calcination. This means that the amount of bagasse from the sugar cane is less than what is needed for simultaneous carbon-capturing and steam generation in utility boilers used to refine the sugar. This is only feasible if the calciner is cofired with oxyfuel combustion of natural gas (case 5). However, it needs significant improvement in the boiler, capacity reduction of boilers and large capacity of CaL compared to other case studies.

Findings for case 6 are interesting. The bagasse obtained from the processed sugar cane is well enough to fuel the energy integrated CaL-sugar refining boiler units. If the boiler is working with bagasse, the energy saving of boiler units is balanced by installation of the calcium looping process. This balance of the energy is coming from the carbonation energy in the carbonator and energy recovered from exhaust gas of the carbonator and calciner. Thus, the bagasse which is extracted from the sugar cane is enough to feed both the boiler and calciner to provide the required onsite energy for refining the sugar.

The thermodynamic and techno-economic feasibility results of case studies are given in Table 9. This results are for optimised case studies when the recovered energy of calcium looping is used to balance a porportion of the boiler energies. The integrated CaL increases the thermal capacity of the reference sugar refinery from 10 MW to 17, 20, 21, 19, 19, 13, and 15 MW for CASES 1–8, respectively. The extra thermal energy, reflected in the increase of equivalent fuel consumption, can be employed in the designed steam cycles to generate electricity and overcome the energy demand of the retrofitted sugar refinery. Therefore, the integration of the CaL to the sugar refinery increases the net power output of the sugar refinery making it a net electricity exporter. While the reference sugar plant relies on 3.8 MW_{el} electricity import, the retrofitted refinery has now become a net electricity export asset, exporting 0.86, 1.47, 1.54, 1.09, 10.9, 1.09, 0.25, and 0.33 MW_{el} of electricity to the grid for CASES 1 to 8, respectively. In optimised retrofitted units, the net power output is considered zero and all the thermal onsite power generations through CaL process can be employed to offset the reduction of boiler duty.

The specific primary energy consumption variation for natural gas

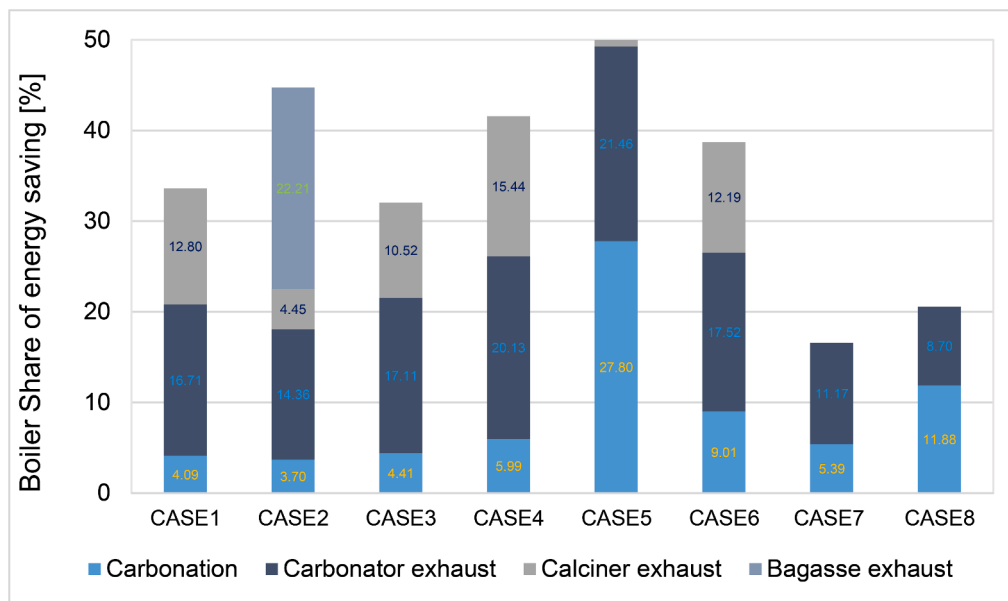


Fig. 5. Result of energy optimisation of the sugar refinery integrated to sugar refinery-the overall number gives the total saving in boiler operation energy and fuel consumption by contribution of energy from carbonator, carbonation, and calcination exhaust energy.

Table 9

Technoeconomic and thermodynamic assessment of the case studies with different integrated system of CaL to the sugar refinery.

Parameter	Reference plant	CASE 1	CASE 2	CASE 3	CASE 4	CASE 5	CASE 6	CASE 7	CASE 8
Thermodynamic assessment									
Gross power output [kW]	14,256	23,024	26,226	27,487	24,516	24,516	24,516	17,506	19,346
Onsite electrical power requirement [kW _{el}]	3857	4545	4356	4774	4725	4726	4725	3991	3971
Onsite thermal power requirement [kW _{th}]	10,398	17,623	20,399	21,169	18,702	18,702	18,702	13,263	15,049
Netpower output [kW]		-856	-1471	-1545	-1088	-1088	-1088	-251	-326
Equivalent fuel consumption [MJ _{LHV} /ADt]	35,572	42,285	47,110	47,685	40,032	40,032	40,032	37,542	34,051
SPECCA [MJ _{LHV} /kg CO ₂ avoided]	0	5	11	7	2	2	2	3	-2
Economic assessment									
Levelised cost of sugar [£/ADt]	7	22	11	12	25	25	25	20	26
Cost of CO ₂ avoided [£/tCO ₂]		62	25	15	54	54	54	103	107

boilers is quite large. When the calciner also uses natural gas (CASE 1), the SPECCA (4.95 MJ_{LHV}/kgCO₂) is comparable with pulp and paper industry (5.7 MJ_{LHV}/kgCO₂) [18]. In case studies 2–3, the SPECCA becomes 10.92 and 6.89 MJ_{LHV}/kgCO₂. The relativity significant up and down in SPECCA from case 1 to 2 and 3 is coming from the calciner carbon which is not captured in case 2 and captured in case 3 when bagasse is being used for calcination process. Case 4–6 results are exactly the same because when the retrofitted plant becomes energy exporter all the bagasse from sugar cane residue should be implemented in boilers and no extra is available for calcination. In these three cases, the boiler uses bagasse and calciner burns natural gas which is captured with SPECCA = 2.52–2.59 MJ_{LHV}/kgCO₂. This value is comparable with iron and steel (2.8 MJ_{LHV}/kgCO₂), [52] and cement industry (2.39–3.27 MJ_{LHV}/kgCO₂) [10,17]. For the decoupled integration of CaL to sugar refinery, the SPECCA is 2.86 MJ_{LHV}/kgCO₂ for natural gas boiler and -1.62 MJ_{LHV}/kgCO₂ for bagasse combustion system. This negative value of SPECCA is likely as CASE8 with bagasse boiler is also compared with a reference sugar plant which is using natural gas as the primary source of energy. The emissions associated with electricity: onsite generation or export, the efficiency and emissions of the reference power generation have an impact on the final figure. As capturing carbon from bagasse leads to net negative carbon emissions in the overall plant, the negative value for SPECCA is not far. Overall, the SPECCA of the test case studies exhibits the superiority of the CaL post combustion technology compared to amine scrubbing that requires 3–5.5 MJ_{LHV}/kgCO₂ for the solvent regeneration only [53–55].

The levelised cost of sugar in basic sugar refinery is around 7.01 £/ADt. The technoeconomic evaluation demonstrated that levelised cost of sugar increases to 22, 11, 12, 25, 25, 20, and 26 £/ADt for case studies 1 to 8, respectively. Regardless of around 270 % increase in the levelised cost of sugar, this industry remains highly profitable resulting in 100 % rate of return for almost all case studies. It is noteworthy that the recent significant rise in natural gas price from 2.56 £/GJ to 7.25 £/GJ influenced the price of most feedstocks including fresh lime, considered in the economic evaluation, giving rise to around 3–18 £/ADt increase in the levelised cost of sugar for the retrofitted plants. As for CASES 2 and 3 with their calciner running on bagasse fuel which is much cheaper than natural gas (the price considered zero as the byproduct of refining), the levelised cost of sugar demonstrates around £4 (for CASE2, CASE3=£5) increase per tonne of sugar produced from the reference plant. In the decoupled versions, the levelised cost of sugar increases from 7 to 20 and 26 £/ADt for CASE7 and CASE8 respectively. Here, the price of fuel indirectly contributes to the price of other goods and service including fresh lime which results in a levelised cost of sugar on par of coupled versions, regardless of lower investment required to install and commissioning the plant.

The estimated CO₂ avoided costs are 62, and 54 £/tCO₂ for CASE1, and CASE4–6, respectively. The cost of CO₂ avoided for CASE 2 and 3 is 25, and 15 £/tCO₂, which is due to the calcination with bagasse and absence of natural gas price in the operating cost of CaL. In CASE2, the carbon from the calcination is not captured—the main reasons for nearly double cost of CO₂ avoided = 25 £/tCO₂ compared to CASE3 = 15

£/tCO₂ with both combustion and calcination carbon capturing. For the decoupled versions, CASE7 and CASE8, the cost of CO₂ avoided increases to 103 and 107 £/tCO₂ despite the absence of the calcination process. The recent rise in fresh lime price results in the higher costs of CO₂ avoided of the decoupled versions compared to coupled versions. Indeed, the result of the case studies shows that the level of cost of CO₂ avoided is mostly dependent on the prices of good and service rather than the initial investment or capital cost for CaL post combustion technology in sugar refinery, this is confirmed in the sensitivity analysis of the economical evaluation. While, data for decarbonisation of sugar industry is scarce, the cost of CO₂ avoided for the coupled versions, CASE1, 4–6, is comparable with technoeconomic results with physical absorption [56] and amine scrubbing [57,58], while it is much lower in CASES 2 and 3. Even with the updated price for material and energy in 2022, the cost of CO₂ avoided for CaL looping is around 30–40 % lower than the amine scrubbing for CO₂ post combustion carbon capture which is around 44–112 £/tCO₂ even with an archaic outdated economic prices [55,59].

This work emphasises the feasibility of retrofitting the sugar refineries with CaL. The costs associated with optimisation of the utility units including boilers, steam turbines and CO₂ compression units are not considered in the economic assessment.

The sensitivity analysis for the effective parameters in the CaL is done here to demonstrate the potential of this technology in adaptation of economic uncertainties. To this end, the cost of CO₂ avoided is estimated within 25 % fluctuations in initial values of capital and variable operating costs, CO₂ transport and storage, good and service including natural gas and limestone. Natural gas price experiences most of the fluctuations. Depending on the case under study, the influence of these parameters is different on the economic uncertainty (see Fig. 6). The initial investment for CaL plant seems to be the most influential factor in the cost of CO₂ avoided as the sensitivity of this factor is more in most of the cases, except CASE7 and 8. The fresh lime price in CASE7 and 8 is more pronounced in the cost of CO₂ avoided with 25.46 % and 24.68 % sensitivity for 25 % variation. For CASES1–6, the fixed capital cost sensitivity for cost of CO₂ avoided is 19.94 %, 80.31 %, 73.78 %, 19.19 %, 19.19 %, and 19.19 %, respectively. The large sensitivity of CASE2 and CASE3 is due to the huge investment needed to design the bagasse calciner and low technology readiness level of this technology for calcium looping. The variable operating cost (≥ 15) and CO₂ capturing (≥ 7), and storage stand after the capital costs in the sensitivity hierarchy.

Summarising the effect of integration of CaL into the sugar refineries, three economic scenarios are investigated:

Economic scenario 1: No CO₂ emissions taxes and no credits for negative emissions (test case scenarios 1–8).

Economic scenario 2: Fossil CO₂ emissions tax and no credits for negative emissions (current situation for economic evaluation). Only the natural gas fuel CO₂ emission is considered eligible for carbon tax and bagasse carbon is determined tax free.

Economic scenario 3: Fossil CO₂ emissions tax and credits for negative emissions. The negative CO₂ emission credits are determined in

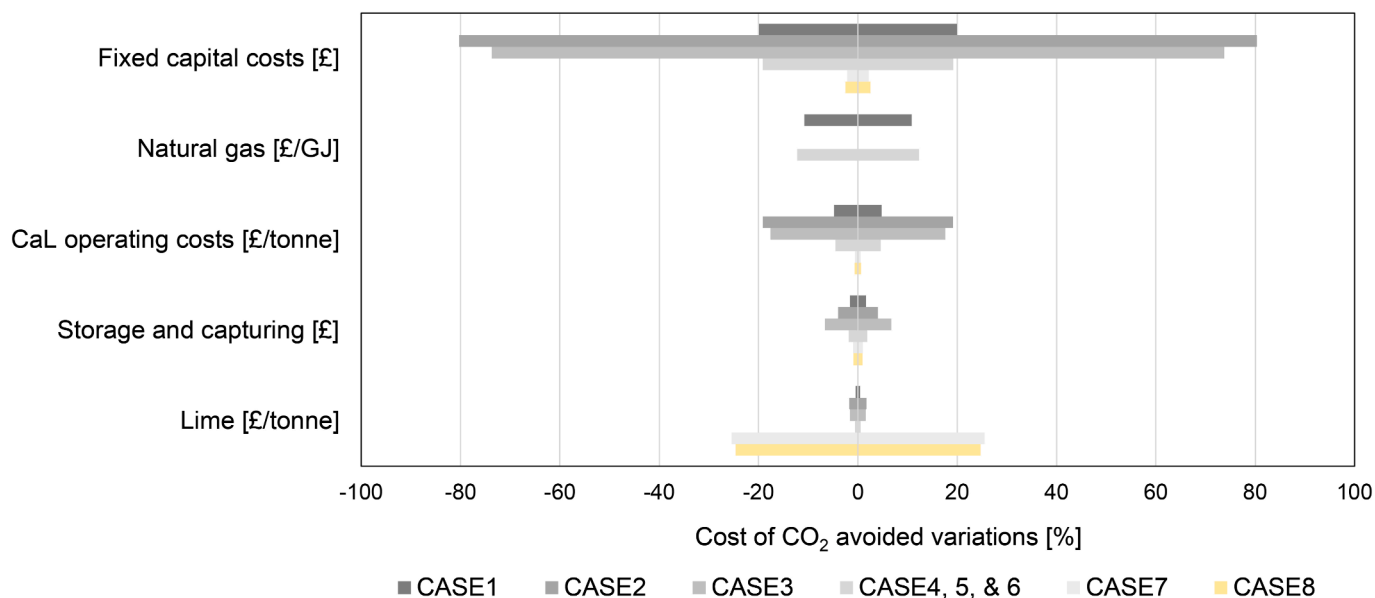


Fig. 6. Cost of CO₂ avoided sensitivity to 25% fluctuations in key investment factors of the integrated sugar refinery-CaL.

economic calculations when the bagasse carbon is captured-i.e, boiler, calciner or both.

The current 2024 average CO₂ emission price allowance under the EU Emission Trading System (ETS) 72.10 £/tCO₂ (=84.360 €/tCO₂) [60] is considered for negative CO₂ emission credit. The economic evaluation of the reference sugar plant is performed using economic indicators given in Table 7 for comparison of different scenarios.

Note that as of 2027, the current carbon tax will be replaced by a new carbon levy in the UK. In this new system, the products of goods and services imported into the UK from the countries with no or low carbon tax needs to pay extra money for the carbon. This may influence of the technoeconomics of integrated sugar refinery and CaL if any of the feedstock for the process of the refining sugar will come from overseas. If all feedstocks are provided from UK domestic companies, the current data and figures can be applied beyond 2027.

Fig. 7 exhibits the influence of different scenarios on the economic indicators of the reference sugar refining plant as well as retrofitted refinery with calcium looping. The fossil CO₂ emission is subjected to emission allowances prices and carbon taxes. Hence, the levelised cost of sugar from scenario 1 to 2 should increase in sugar reference plant and CASES 1, 2, 3 and 7 with natural gas fired boiler. The levelised cost of sugar increases from 7.01 to 17.37 £/ADt for the base reference plant, from 22.04 to 23.07 £/ADt for CASE 1, 11.08 to 12.12 £/ADt for CASE2, 11.56 to 12.6 £/ADt for CASE3, and 20.43 to 21.47 £/ADt for CASE8.

The retrofitted plants experience a much lower increase in the levelised cost of sugar compared to reference plant (10 % vs 150 %) as 90 % of carbon is captured in the carbonator and only 10 % of the carbon is subjected to the tax. No change in CASE 4, 5, 6 and 8 levelised cost of sugar from scenario 1 to 2 is due to the biogenic carbon which emanates from bagasse fired boilers.

From scenario 2 to 3, the results are opposite of those from scenario 1 to 2. Here, the bagasse fired boiler and calciner would be advantageous for the sugar industry as the negative carbon removal from biomass contributes to the revenue of the refining. That is the reason for the reduction in levelised cost of sugar CASE3 from 12.60 to 6.43 £/ADt, CASE 4,5 and 6 from 24.89 to 12.17 £/ADt, and CASE8 from 25.89 to 13.17 £/ADt, respectively. The results for CASE 1 and 7 remain intact as no renewable carbon capture will be envisaged. A similar situation is for CASE 2 as the bagasse indirectly contributes to the calcination with its carbon released to the atmosphere. The findings demonstrate that the levelised cost of sugar will be reduced by 48.91 %, 51.12 % and 49.14 % in CASE 3, CASE4-6 and CASE8 when scenario 2 changes to 3, respectively.

When carbon is taxed, the cost of CO₂ avoided decreases whatever the scenario is under the investigation. This increase coming from the rise of the levelised cost of sugar of the reference plant results in both positive and negative cost of CO₂ avoided depending on the case study. When scenario 1 shifts to scenario 2, the cost of CO₂ avoided decreases

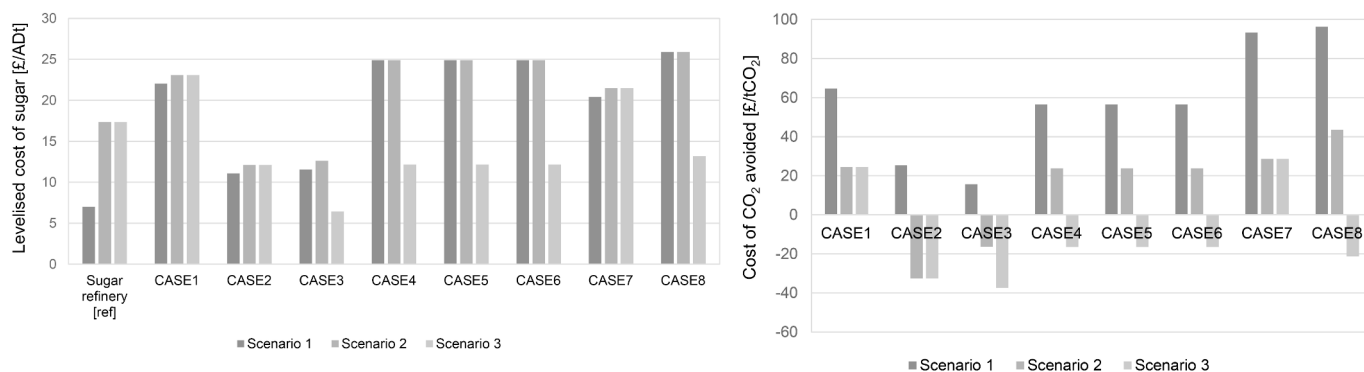


Fig. 7. Influence of different economic scenarios on (a) levelised cost of sugar and (b) cost of CO₂ avoided for different case studies (Ref corresponds to references sugar refinery plant).

from 64.65, 25.31, 15.59, 56.51, 93.41 and 96.32 to 25.54 ($\approx 62.04\%$ reduction), -32.58 ($\approx 228.74\%$ reduction), -16.34 (≈ 204.83 reduction), 23.78 (≈ 57.92 reduction), 28.52 (≈ 69.46 reduction) and 43.51 (≈ 54.84 reduction) £/tCO_2 for CASEs 1, 2, 3, 4–5, 7, and 8, respectively. The negative cost of CO_2 avoided in CASE 2 and 3 from scenario 1 to 2 even when the renewable carbon capture is not credited shows first the role of the fuel price in the technoeconomic value of CaL technology and second the low capital investment which is required for the integration of this technology to the sugar refinery which eventuate in lower levelised cost of sugar when the carbon is taxed. From scenario 2 to 3, the cost of CO_2 avoided exhibits a further decrease changing from -16.34 , 23.78 , and 43.51 to -37.45 , -16.43 , and -21.39 £/tCO_2 for CASE3, CASE4-5, and CASE 8, respectively. The is equivalent to 129.13% , 169.10% and 149.6% reduction in the cost of CO_2 avoided. The intact cost of CO_2 avoided for CASE 1,2 and 7 is due to indirect combustion of either natural gas or bagasse in the calcination process which offers no carbon negative credits compared to the scenario 2 with carbon taxations.

The CO_2 emission allowance price is another economic factor that may be subjected to monthly oscillation and is used here to further sensitivity analysis of the retrofitted sugar plant. When the economy scenario under scrutiny was changed from 1 to 2, the cost of CO_2 avoided should be negative as the saving from carbon tax in the economy plan increases the revenue of the sugar refining to an extent that the levelised cost of sugar of the retrofitted plants in CASE 2 and 3 culminated in a number lower than the reference plant for which the carbon is under the tax. This is also arising from lower annual investment, i.e., operating costs, which are required for calcination with bagasse if bagasse fuel price is considered zero as a byproduct of the sugar refinery. In 2022, the year coming to transition zone from phase 3 to 4 of EU plans for decarbonisation, the CO_2 emission allowance prices rocketed from 23.73 £/tCO_2 (≈ 20.29 £/tCO_2) to almost 80 £/tCO_2 (≈ 71.79 £/tCO_2) resulting into the significant revenues for sugar industry if carbon tax is implemented. Another sensitivity analysis provided here is for CASE2 and 3 under carbon allowance prices 0 – 70 £/tCO_2 for three different electricity export prices 180 , 270 and 350 £/MWh (Fig. 8). The findings show that for scenario 2 the cost of CO_2 avoided should be nearly zero for the carbon emission allowance price 30 – 35 £/tCO_2 . The price of electricity is also very influential in the cost of CO_2 avoided. When the

price of electricity export drops to 180 £/MWh , levelised cost of sugar approaches that of reference sugar refinery plant in phase 4 EU plan (carbon emission allowance price 70 £/tCO_2) leading to nearly almost zero cost of CO_2 avoided. For electricity grid price 350 £/MWh , the levelised cost of sugar for CASE2 and 3 are equal to the reference plant and the cost of CO_2 avoided is almost zero. Note that CaL technology for other case studies cannot result in negative cost of CO_2 avoided under scenario 2. However, when the negative carbon credit is coming into play, the results should be different.

If scenario 3 is under scrutiny with negative carbon credits (Fig. 9), CASE 4, 5, 6 and 8 along with CASE3 could have their levelised cost of sugar lower than the reference sugar plant as the revenue from negative carbon taxation adds into the refining profits. This is also for case studies with bagasse carbon captured either from the boiler or in the calcination process. Based on the current scenarios for carbon allowance prices, CaL technology leads into almost negative cost of CO_2 avoided and profitability of this technology in sugar refining process. The boiler fired natural gas with bagasse calciner (CASE3) has the negative cost of CO_2 avoided which is lower compared to other case studies for all the three electricity export prices. If the boiler is using bagasse with 100% load and calcination is ongoing using natural gas (CASEs 4–6), this technology leads to profitability for sugar refining as the CO_2 emission prices reaches 61 , 43 and 27 £/tCO_2 for electricity prices 180 , 270 and 350 £/MWh , respectively. For the decoupled version CASE8 with boiler fired bagasse system, the cost of CO_2 avoided becomes negative at 54 , 48 , and 44 £/tCO_2 for electricity prices 180 , 270 and 350 £/MWh , respectively.

Note that the under the scenarios 1–3 investigated, the levelised cost of sugar is higher for the retrofitted plant compared to the reference plant when both the combustion and calcination is with natural gas-CASE1 which is a more realistic case study for the sugar plants in the UK (see Fig. 7). The levelised cost of sugar increases from 22.04 to 23.07 £/ADT for scenario 1 to 2 and remains unchanged from scenario 2 to 3 as there is no credit for capturing the anthropogenic carbon emission from natural gas. The cost of CO_2 avoided drops from 62.23 to 24.54 £/tCO_2 if the carbon is taxed under scenario 2.

Apart from government's carbon plans, outlooks etc, the operational characteristics of CaL also could change the operating points as well as economics of the retrofitted sugar refinery. Here, the ratio (F_0/F_R) representing the fresh make-up sorbent to the circulating lime between the

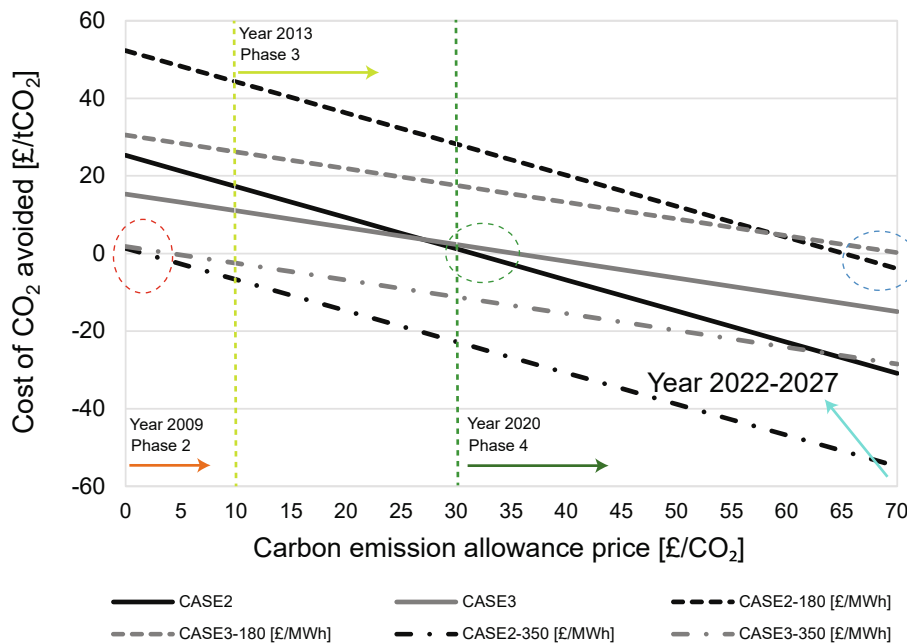


Fig. 8. Impact of CO_2 emission allowance price and electricity price on the cost of CO_2 avoided under scenario 2-CASE2 and CASE3 with price of electricity 180, 270 (solid line-2022 electricity price) and 350 £/MWh .

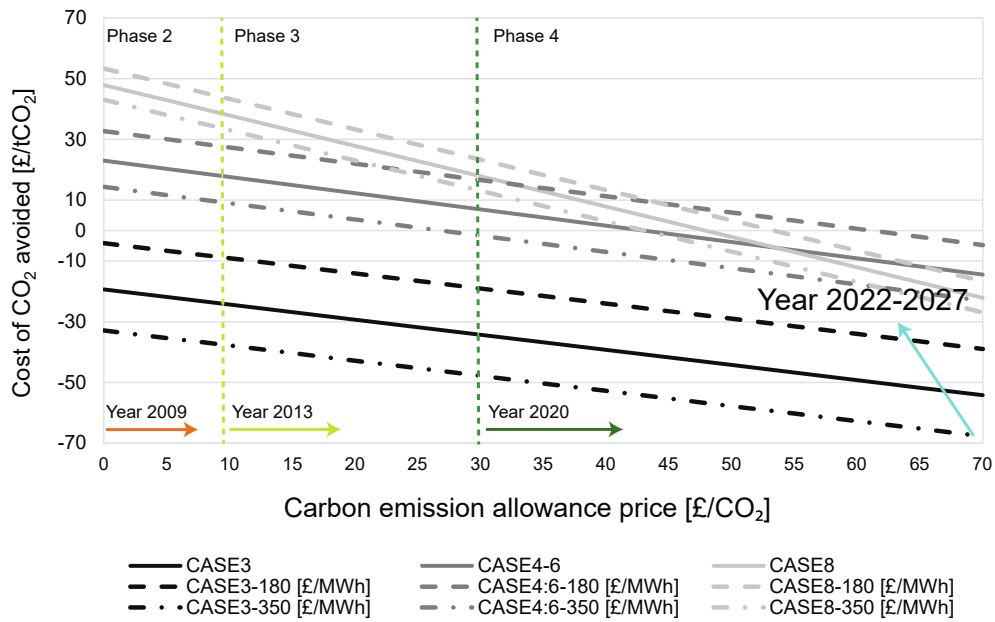


Fig. 9. Impact of CO₂ emission allowance price and electricity price on the cost of CO₂ avoided under scenario 3-CASE2 and CASE3 with price of electricity 180, 270 (2022 electricity price) and 350 £/MWh.

carbonator and calciner is used to exhibit the sensitivity of the retrofitted plant to operating conditions. This ratio can change the natural gas, oxidiser quantities and the rate of electricity export from the retrofitted plant. The variation of the economic indicators (levelised cost of sugar and cost of CO₂ avoided) in relation to relative sorbent ratio for CASE1 is presented in Fig. 10 (other case studies is available in supplementary material with almost a same trend). An increase in the sorbent make-up ratio increases natural gas for calcination, the rate of electricity export, carbon emissions from the retrofitted plant and as the result both the nominator and denominator in the SPECCA function. Once the retrofitted sugar refinery becomes an electricity exporter, the net power output increases. This has a negative impact on the SPECCA decreasing the equivalent fuel consumption and CO₂ emissions. Here, 0.02 increase from the original setpoints of sorbent make up fraction ($F_0/F_R = 0.04$) leads to 4.06% decrease in SPECCA and 4.44% increase in the specific exported electricity. The sorbent make up fraction can also increase the levelised cost of sugar and ironically the cost of CO₂ avoided. Indeed, an increment in F_0/F_R increases both the rate of carbon capture as well as levelised cost of sugar as more lime is available for calcination in the lime kiln. This leads to the relative increase of carbon capture rate comparing to the levelised cost of sugar, and resultant

decrease in the cost of CO₂ avoided. 50 % increase in the sorbent make up rate ($F_0/F_R = 0.04$) results in 4.89 % increase in the levelised cost of sugar and 2.78 % decrease in the cost of CO₂ avoided of the retrofitted plant. Note that the sorbent make-up fractions below 0.02 and above 0.06 are not practical as low and high rate of recirculation are accompanied respectively by low carbonation rate and higher investment for the retrofitted plants which makes this technology incompetent compared to the other available carbon capture processes.

7. Conclusions

This paper provides the conceptual design of a new carbonation process with inherent CO₂ capture for a sugar refinery. This design can be retrofitted to existing sugar refineries by integrating a CaL process in the existing lime cycle of the carbonation process in sugar refining. Eight case studies including different fuel types for both boiler and calciner, carbonation using fresh lime (decoupled CaL), and direct application of fuel in the calcinations are envisaged and investigated. Findings demonstrate that the energy that could be obtained from CaL can offset a part of boiler energy thereby leading to remarkable savings in the fuel consumption of the boiler (30–50 % for coupled versions and

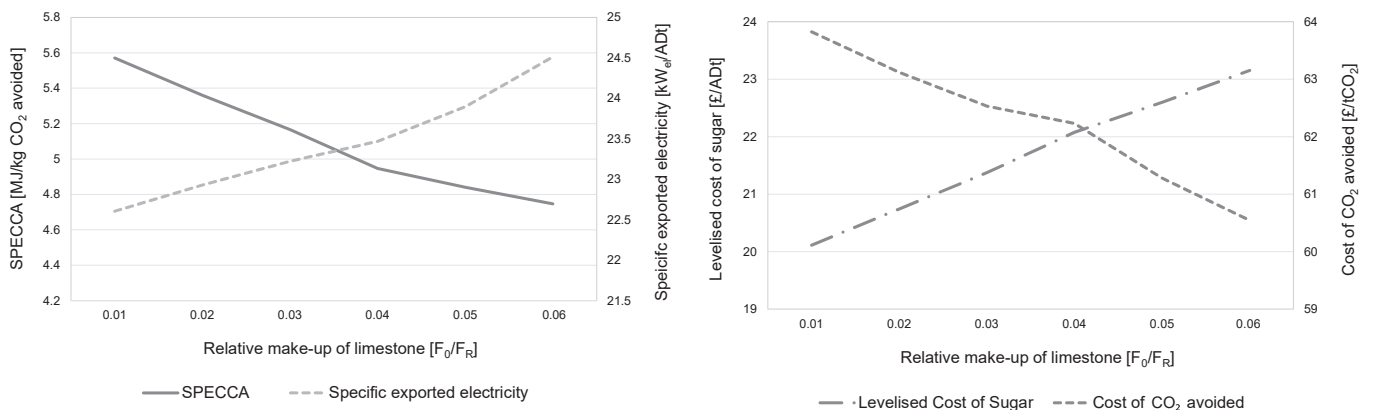


Fig. 10. Impact of fresh limestone make-up rate on SPECCA [MJ/kg CO₂ avoided], specific exported electricity [kW_{el}/ADt], levelised cost of sugar [£/ADt] and cost of CO₂ avoided for CASE1 [£/tCO₂].

10–20 % for the decoupled versions). In another case scenario, the retrofitted sugar plant can be an electricity exporter if the surplus of energy from carbonation is to be exploited in a designed steam cycle. In this case, the cost of CO₂ avoided becomes 62.23 £/tCO₂ if the carbon is not taxed and 24.54 £/tCO₂ if the phase 4 of carbon emission allowance price is considered for CASE1. The application of bagasse in calcination process is highly adventitious into the economy of the retrofitted plant although the TRL of the bagasse calciner needs further development to be applicable under the real case scenarios.

The technoeconomic investigation of the retrofitted plants is carried out for fresh limestone (CaCO₃) make-up rate, unit cost prices, carbon capture and storage prices, variable operating costs, lime (CaO) and fuel prices. The results here show that the cost of CO₂ avoided is more sensitive to the capital investment of CaL for most of decarbonisation plans. As the levelised cost of sugar and specific exported electricity is increasing in response to the sorbent make-up ratio, the trend of SPECCA and cost of CO₂ avoided is decreasing towards the application of the more sorbent make up fraction in the calcination.

This paper also demonstrates that the application of calcium looping process for carbon capture and storage in sugar refineries becomes economically highly feasible on the inclusion of biogenic carbon emissions in the EU ETS and attribution of credits for them. The findings confirm if the biogenic carbon is incentivised in the future policies in the cost associated with the operation of sugar refining units, the cost of CO₂ avoided will drop by 61% if CaL CCS is implemented in sugar refineries. The adoption of these policies will enable the sugar refining process to become carbon negative and deployment of sugar as a negative carbon commodity to the market.

CRedit authorship contribution statement

Bahamin Bazooyar: Software, Resources, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Ali Nabavi:** Formal analysis, Data curation, Conceptualization. **Peter Clough:** Supervision, Software, Project administration, Methodology, Investigation. **Edward J. Anthony:** Writing – review & editing, Visualization, Validation, Supervision. **Vasilije Manovic:** Writing – review & editing, Visualization, Supervision, Software, Resources, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

<https://doi.org/10.17633/rd.brunel.25864513.v1>.

Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.enconman.2024.118597>.

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