



Techno-economic feasibility of fluid catalytic cracking unit integrated chemical looping combustion – A novel approach for CO₂ capture

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ABSTRACT

Oil refineries are collectively responsible for about 4–6% of the global CO₂ emissions, largely because of the regenerator part of the Fluid Catalytic Cracking (FCC) unit (25–35%). An advanced combustion technology, also called chemical looping combustion (CLC), has been recently presented as a novel CO₂ capture process for FCC units; however, no study provides the economic feasibility of a CLC-FCC unit. In this study, a techno-economic feasibility of the novel CLC-FCC unit was presented for the first time based on a case study with 50,000 barrels feed per day. A rigorous mass and energy balance estimation shows that 96 vol% of coke regeneration (combustion) was achieved in the FCC regenerator by using a stoichiometrically required amount of metal oxide (CuO modified catalysts) at 750 °C for 45 min. The preliminary energy penalty calculations of the proposed CLC-FCC unit (0.21 GJ/ton CO₂) is relatively lower compared to the post-combustion (3.1–4.2 GJ/t CO₂) via amine solvent and oxy-fuel combustion (1.8–2.5 GJ/t CO₂) units reported in the literature. The equipment purchase cost (EPC) is 1.1 times higher than a standalone FCC unit due to the increase in the number of processing equipment required. The cash flow analysis results reveal a yearly basis average CO₂ capture cost of 0.0106 US\$/kg of CO₂ (~10.6 US\$/ton CO₂) for the CLC-FCC unit, which is lower compared to the other conventional CCS technologies i.e. oxy-fuel combustion and post-combustion. Factors such as EPC, capital expenditure (CAPEX), and discount rate significantly influenced the capture cost. In contrast, the CO₂ capture cost is not influenced by a change in oxygen carrier and electricity cost.

1. Introduction

Over the past two decades, there has been a tremendous elevation in the average CO₂ emissions, and the concentration of atmospheric CO₂ has reached nearly 400 ppm [1]. Compared to the emissions level in the early 1850s, this value is more than 40% higher. The CO₂ increase is largely responsible for the challenges in the present world such as climate change and atmospheric pollution. Therefore, there has been a growing interest in CO₂ capture, utilization, and storage (CCUS) technologies to address this global challenge.

One of the most promising methods for natural CO₂ capture is through plantings and afforestation [2] In order to effectively reduce CO₂ levels, innovative designs incorporating artificial plant-based green

buildings have emerged as potential solutions, specifically by implementing virgin ivy plants on building surfaces such as walls and roofs [2]. This approach, known as direct air capture, has the potential to capture over 3.5 billion tons of CO₂ annually, which is equivalent to approximately 6.9% of global greenhouse gas emissions [3]. Furthermore, a variety of processes have been developed to address sequential CO₂ separation or CO₂ capture, including post-combustion, pre-combustion, and oxyfuel combustion [1]. The post-combustion capture of CO₂ from the exiting flue gas has been completed via processes such as adsorption, absorption, or cryogenic separation [4,5]. In contrast, pre-combustion technology implements CO₂ before combustion is completed through an integrated gasification and water gas shift reaction [6]. During oxy-fuel combustion, high-purity oxygen combined with recycled flue gas is used for combustion to produce ultra-pure CO₂

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Abbreviations

CAPEX	Capital expenditure
CCUS	CO ₂ capture, storage, and utilization
CCS	Carbon Capture and Storage
CEPCI	Chemical Engineering plant cost index
CLC	Chemical Looping Combustion
iG-CLC	In-situ Gasification-Chemical Looping Combustion
CLOU	Chemical Looping with Oxygen Uncoupling
EPC	Equipment purchase cost
FCC	Fluid Catalytic Cracking
FCI	Fixed capital investment
FOC	Fixed operating cost
LPG	Liquefied petroleum gas
LSA	Local sensitivity analysis
MEA	Monoethanolamine
OPEX	Operating expenditure
TAC	Total annual cost
TEA	Techno-economic analysis
VGO	Vacuum gas oil
VOC	Variable operating cost

and water vapour stream for subsequent sequestration [7].

Heavy industries (iron and steel production, oil refineries, cement manufacturing, and petrochemicals) make up the majority of the remaining stationary CO₂ producers, even though the power sector (energy production) leads the pack ~78% [8]. Moreover, about 4–6% of the world's CO₂ emissions are from oil refineries [9], largely because they are the second-highest energy consumers among these industries. CO₂ emissions from oil refineries originate from several different units such as the topping tower, the utility production unit, the vacuum distillation unit, the steam methane reforming, and the fluid catalytic cracking (FCC) regenerator [10]. The FCC regenerator is responsible for about 25–35% of CO₂ emissions from a standard refinery [10]. As shown in Fig. 1, The traditional FCC process is implemented in petroleum refineries for the conversion of heavy oil fractions to lighter petroleum gas and gasoline via FCC catalyst, where coke is deposited over the catalyst surface. In order to clean the catalyst, the coke is therefore burned with

air in the regeneration unit. Burning of coke on the catalyst surface produces a significant amount of CO₂ which is the only CO₂ source in the FCC unit. In order to decarbonise the refineries, developing cost-effective CO₂ capture technologies for FCC units is significantly important. Considering the FCC unit characteristics and design, CO₂ released from the coke combustion in the regenerator can be captured by the integration of oxy-fuel and post-combustions [11,12].

The CO₂ from FCC unit can be captured by integration of post-combustion (3.1–4.2 GJ/t CO₂ of energy penalty and 75–110 €/t CO₂ of CO₂-avoiding cost) and integration of oxy-fuel combustion (1.8–2.5 GJ/t CO₂ of energy penalty and 55–85 €/t CO₂ of CO₂ avoided cost) [13–15]. In addition to these CO₂ capture processes, chemical looping combustion (CLC) has recently been proposed as an alternative CO₂ capture technology for the FCC unit and offers considerably lower energy penalties (*ca.* 0.2 GJ/t CO₂) [13]. Although post-combustion appears to be mature technology for the FCC unit thanks to its wide applications on industrial scales, the oxy-fuel combustion is preferred due to its cost-effectiveness [16]. However, oxy-fuel combustion requires further developments and improvements before its commercial implementation. Since this technology also faces challenges including advanced process equipment requirements and a high energy penalty [17]. Due to the nature of inherent combustion in CLC, the technology produce a concentrated CO₂ stream (as a flue gas) [18] from the coke combustion in the regenerator which eliminates the requirements of extensive CO₂ separation and purification processes unlike other technologies; a CO₂ separation unit for both post-combustion and an air separation unit for oxy-fuel combustion. Metal oxides are employed in the CLC process in place of air to provide oxygen for the fuel stream's combustion [19]. Güleç et al. [17,20] showed that the integration of CLC to FCC unit is an efficient and promising method for CO₂ capture from FCC unit. It was demonstrated that about 90% vol of coke combustion could be attained with a mixture of either CuO or Mn₂O₃ metal oxides. Nabipour and Iranshahi [21] explored the use of CLC as the source of heat for the residue FCC process using NiO18- α -Al₂O₃ as an oxygen carrier. Additionally, Güleç et al. [14] presented a comprehensive review of the status and progress of different CCS technologies applied to the FCC units.

Although several studies have demonstrated promising results in the integration of CLC with FCC, an investigation of the economic feasibility of the CLC-FCC concept is scarcely reported. The present study for the

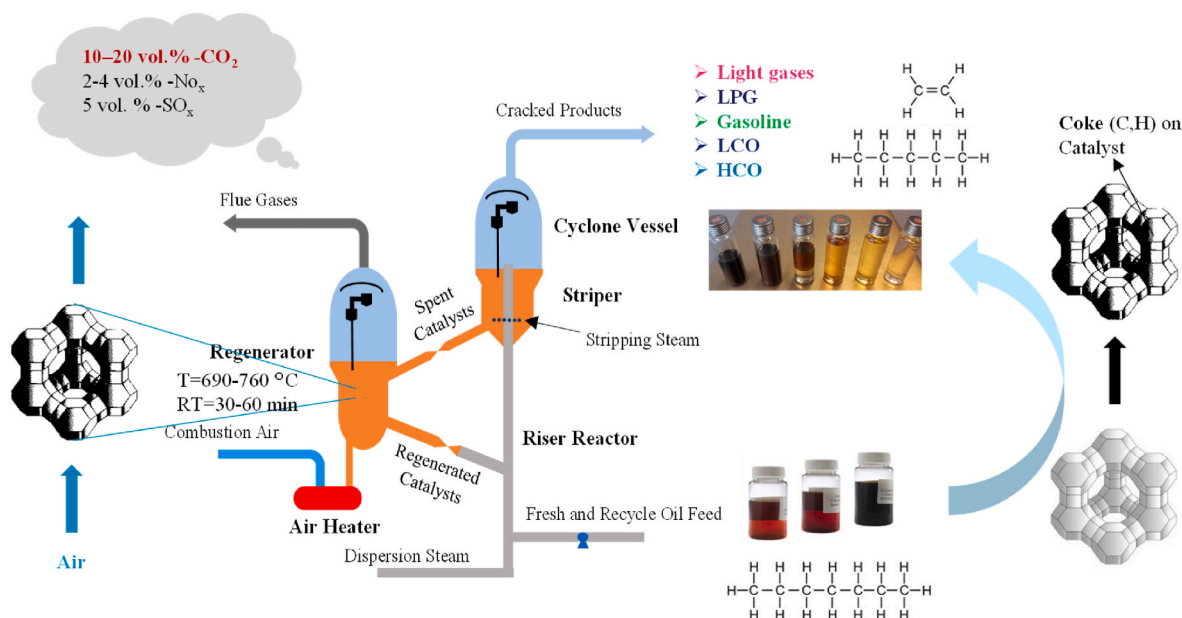


Fig. 1. General overview of conventional FCC unit.

first time explores the economic feasibility of integrating CLC with FCC unit using a case study with 50,000 bpd vacuum gas oil (VGO) cracking. A novel conceptual design of a potential CLC-FCC unit was proposed as well as a rigorous calculation to appraise the mass and energy balance on the CLC-FCC unit. In order to carry out the techno-economic evaluation, the total annual cost (TAC) of the CLC-FCC plant was determined by combining the annualized operating expenditure (OPEX) and capital expenditure (CAPEX). The CAPEX is converted into a constant yearly payment over the project's entire lifespan. To estimate the cost of CO₂ capture, the TAC is divided by the amount of captured CO₂ in the CLC-FCC unit. To evaluate the profitability, a cash flow analysis is conducted to assess the economic viability of the CLC-FCC design. Additionally, a sensitivity analysis is performed to determine the impact of various parameters on the CO₂ capture cost.

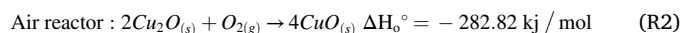
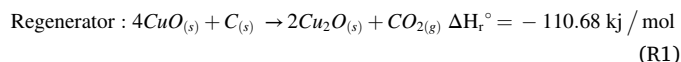
2. Proposed CLC-FCC concept

Fig. 2 shows the schematic of the CLC-FCC unit, and it is important to note that the proposed design has undergone experimental validation in previous studies [17,20]. The integrated CLC-FCC unit comprises three main units: an air reactor, a regenerator, and an FCC riser reactor. To incorporate CLC into the FCC unit, it is necessary to modify the FCC catalyst particles with an oxygen carrier such as CuO, Co₃O₄, or Mn₂O₃. Since coke deposition over the FCC catalyst is only around 1–2 wt%, a relatively small quantity of oxygen carriers is required for FCC catalyst modification [14,22]. Based on our previous studies, the FCC catalyst needs to be modified with approximately ~12 wt% of CuO, or ~18 wt% of Co₃O₄ or ~29 wt% of Mn₂O₃, due to the varying capacities of the oxygen in metal oxides [17,23].

These three interconnected units operate simultaneously as follows: in the FCC Riser Reactor, the modified FCC catalysts (designated as Me_nO_{m-1}/Cat) with the reduced form of oxygen carriers (Me_nO_{m-1}; e.g., Cu₂O, CoO, Mn₃O₄) are circulated from the regenerator to the FCC riser reactor. In the riser reactor, the cracking reaction over Me_nO_{m-1}/Cat leads to coke deposition (Coke/Me_nO_{m-1}/Cat), which is then transferred back to the regenerator for coke regeneration. In the Air Reactor, the Me_nO_{m-1}/Cat is circulated from the regenerator to the air reactor, where the reduced form of metal oxide (e.g., CoO to Co₃O₄, Cu₂O to CuO, or Mn₃O₄ to Mn₂O₃) (designated as Me_nO_m/Cat) is re-oxidized using the oxygen present in the air. In the Regenerator, by mixing the coke-deposited catalysts (Coke/Me_nO_{m-1}/Cat) with the oxidized oxygen carrier-modified catalysts (Me_nO_m/Cat), coke is oxidized to CO₂ with the oxygen in Me_nO_m/Cat. This process cleans the coke over the catalyst

(Coke/Me_nO_{m-1}/Cat → Me_nO_{m-1}/Cat) and reduces the oxidized oxygen carrier-modified catalysts (Me_nO_m/Cat → Me_nO_{m-1}/Cat). At the end of the reaction in the regenerator, the solid catalyst is expected to be in a reduced form, denoted as "Me_nO_{m-1}/Cat" ready for simultaneous circulation to both the riser and the air reactors (further details of the reaction is presented in Fig. 2).

The coke combustion with CuO are presented in equations (1)–(3). The new CLC-FCC unit was designed with the consideration of energy balance through chemical looping combustion, similar to the conventional FCC unit. CLC divides the combustion reaction into two steps: the oxidation of coke with metal oxide and the oxidation of reduced metal oxide with oxygen in the air. The total energy generated through coke combustion and metal oxide oxidation in the CLC-FCC concept are expected to be equivalent to the energy produced in the conventional FCC unit.



ΔH_o° and ΔH_r° are the standard heats of reaction for oxidation and reduction at 298 K and 1 atm.

The potential combustion reactions in the regenerator would be i) the soft coke may attack the solid oxygen carriers as a gas-solid reaction, ii) Oxygen release from oxygen carriers may attack soft coke (gas-gas) or hard coke (gas-solid) reactions, iii) the solid oxidized metal oxides (Me_nO_m/Cat) and coke deposited FCC catalysts (Coke/Me_nO_{m-1}/Cat) may also support solid-solid reaction, as they all operate in a fluidised bed reactor called a regenerator [14]. Coke combustion with metal oxide can be either endothermic or exothermic, depending on the specific metal oxide, while the oxidation of reduced metal oxide in the air reactor is exothermic. Regardless of the endothermic or exothermic nature of the regenerator process, the overall net energy/heat production remains unchanged. Heat transfer from the air reactor to the regenerator and the FCC riser reactor can be achieved through methods like employing an FCC feedstock preheater or utilising hot catalysts, or unique reactor designs. Although there is no study which has yet been conducted to optimise the design and heat/energy transfer between these units, the CLC-FCC concept is aimed at developing the next generation of cleaned net-zero FCC units.

In addition to inherent CO₂ capture, the CLC-FCC has advantages for

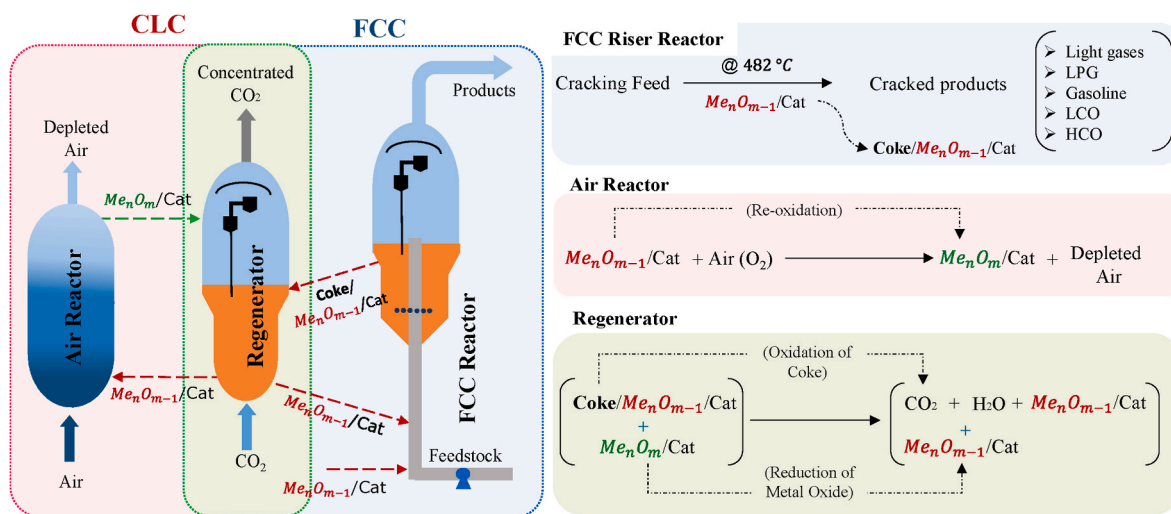


Fig. 2. Schematic representation of the integrated CLC-FCC unit (Me_nO_{m-1}/Cat: Reduced form Oxygen Carrier (Cu₂O, CoO, Mn₃O₄) modified FCC catalyst. Me_nO_m/Cat: Oxidized form Oxygen Carrier (CuO, Co₃O₄, Mn₂O₃) modified FCC catalyst).

lower NO_x and SO_x emissions compared to traditional combustion. Various chemical looping applications (CLC, iG-CLC, and CLOU) of coal and biomass resulted in lower SO_x, NO_x and N₂O emissions thanks to the unique oxygen supply mechanisms of chemical looping and the absence of atmospheric nitrogen in the combustion environment [24]. However, the SO_x and NO_x emissions strongly depend on the oxygen carriers in the chemical looping applications and operating conditions [25], as some of these metals could contribute to the oxidation of Sulphur and Nitrogen as catalysts.

The proposed CLC-FCC concept incorporates solid metal oxides, specifically CuO, Co₃O₄, and/or Mn₂O₃, modified with FCC catalyst to enable CO₂ capture with minimal energy loss [17,20]. However, the use of CuO and Co₃O₄ raises safety concerns, particularly in relation to their potential environmental impact [25–27]. On the other hand, Mn₂O₃ is generally considered safe for use as an oxygen carrier in the CLC-FCC process. It is crucial to consider the hazards associated with these metal oxides, especially when handling and disposing of the used catalysts.

3. Techno-economic analysis procedure

Fig. 3 shows the process of the techno-economic analysis (TEA) adopted in this study. The first step involves a rigorous calculation of the mass and energy balance with several assumptions and product distribution listed in Tables 1 and 2, respectively. The assumptions and data used to compute the mass and energy calculations as well as the experimental data were obtained from relevant literature. Details of the rigorous mass and energy balance can be found in the supplementary materials. The mass balance appraisal involves the definition of the FCC unit case study with 50,000 barrels feed per day (bpd).

The feed is a vacuum gas oil (VGO) and no recycle stream is processed. The FCC catalyst is modified with about 11.2 wt % of Cu₂O with the assumption that there is close to 72.28% conversion of VGO [28]. As shown in Table 1, VGO cracking reaction was assumed over Cu₂O-modified ECat and the product distributions reported earlier by Sadeghbeigi [28] were modified by experimental results presented over Cu₂O/ECat [17,20]. Our earlier research showed that the cracking reaction's conversion, yields, and product selectivity are unaffected by reduced metal oxide modification (Cu, Cu₂O, Mn₃O₄, and MnO) with ECat [17]. An insignificant decrease in the gasoline yield (–2.5%) and LPG yield (–1.5%) were observed while the amount of coke produced elevated by about +1.5% after n-hexadecane cracking over Cu/ECat.

Similar to the mass balance, the energy balance was manually computed by considering several thermodynamic information and assumptions listed in Table 3. The results from the energy balance helped in the estimation of the integrated process energy penalty. The energy balance was progressively computed for the FCC riser reactor, regenerator and air reactor with the overall energy balance determined from equation (1).

$$\sum Q_{CLC-FCC} = \sum Q_{FCC-Ris} + \sum Q_{Reg} + \sum Q_{AR} \quad (1)$$

where $Q_{CLC-FCC}$, $Q_{FCC-Ris}$, Q_{Reg} and Q_{AR} represent the heat balance in the integrated CLC-FCC unit, FCC reactor, regenerator, and air reactor respectively.

3.1. Equipment purchase cost

The equipment purchase cost (EPC) was estimated by combining the mass and energy balance information with literature values. Based on

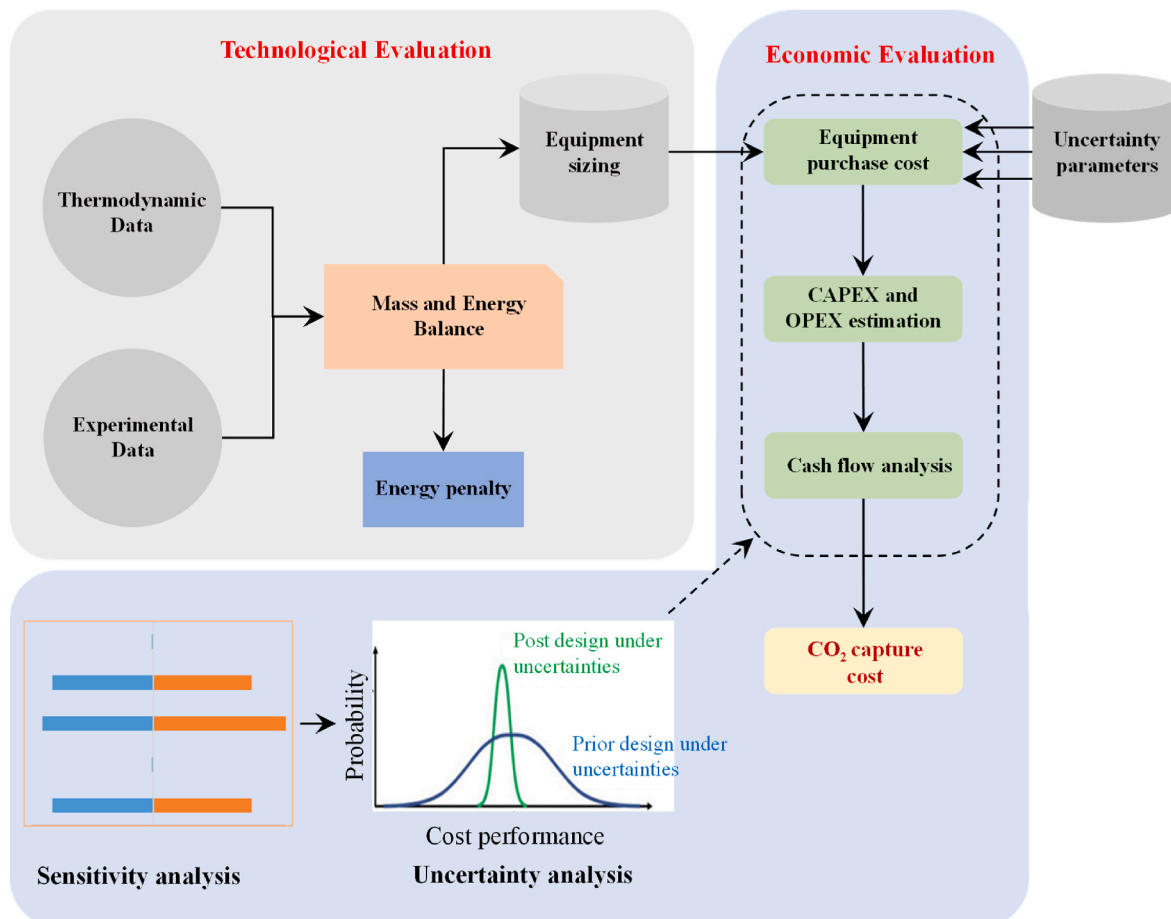


Fig. 3. Overview of the techno-economic analysis methodology.

Table 1
Assumptions for the operating conditions of the proposed CLC-FCC Unit.

Process Variable	Values	Reference
Assumptions for FCC Riser Reactor		
Reactor Feed Rate (VGO)	50000 bpd (299067 kg/h)	[28]
Feed Temperature	220 °C	[28,29]
Catalyst/Oil Ratio	5.0	[30]
Cracking Catalyst ^a	Cu ₂ O/ECat	[17,20,22]
Particle Diameter	150 µm	[22,31]
Reaction Temperature	520 °C	[30,31]
Steam for Atomizing	5% of VGO feed	[29]
Steam for Stripping	2% of VGO feed	[28,29]
Coke on Catalyst	1.12 wt %	[28]
Assumptions for Regenerator		
Regenerator Temperature	750 °C	[17,22,23]
Flue Gas Temperature	750 °C	[17,22,23]
Fluidisation Gas (CO ₂) Temperature	200 °C	Assumption
Oxidation material ^b	CuO (modified on ECat)	[17,22,23]
CuO/Coke ratio ^c	1.0	[17,22,23]
Combustion Efficiency	96 vol %	[17,22,23]
Fluidisation Gas ^d	CO ₂	[17,22,23]
Assumptions for Air Reactor		
Oxidation Temperature	700 °C	[32]
Fluidisation Gas (Air) Temperature	200 °C	Assumption
Air/Cu ₂ O flowrate ratio ^e	1.0	[33]

^a Cracking catalyst is an Equilibrium Catalyst (ECat), which is modified 11.2% of Cu₂O.

^b The combustion of coke on ECat requires the stoichiometric amount of CuO and Mn₂O₃.

^c Coke deposited on Cu₂O/ECat can be combusted with the stoichiometrically required amount of CuO (12%) modified with ECat. Therefore, the molar ratio of oxygen (released during the reduction of CuO to Cu₂O) to coke was kept at 1.0.

^d The volumetric flow rate of CO₂ for fluidisation is assumed similar to the volumetric flow rate of nitrogen as if air was used for the combustion of coke in the regenerator.

^e The air flow rate in the Air Reactor is assumed to be equal to the flow rate of Cu₂O transferred from regenerator unit to air reactor. Under this condition, the oxygen supplied through the Air is 2 times higher than the stoichiometrically required oxygen for the oxidation of Cu₂O to CuO. The oxidation of Cu₂O is possible once the ratio of Air flowrate to Cu₂O flowrate is kept at 0.34.

the information obtained from relevant literature and the mass and energy balance results, the six-tenths-factor rule was implemented in the final EPC appraisal (Equation (2)).

$$\frac{C_a}{C_b} = \left(\frac{F_a}{F_b} \right)^n A_m A_T A_p A_M \quad (2)$$

where C_a and C_b represent the approximate cost of the equipment with the required capacity and known cost respectively. Similarly, F_a and F_b are the size factor of the equipment with the required capacity and

Table 2
Cracking reaction products under 50000 bpd fresh VGO feed and updated product distribution for the CLC-FCC unit.

Product distribution	Case study ^a		CLC-FCC unit ^b		Product Distribution Differences
	Flowrate (kg/h)	Distribution (wt. %)	Flowrate (kg/h)	Distribution (wt. %)	
Fresh Feed ^c (50000 bpd)	299067.0		299067.0		
Products					
Light gases	9540.4	3.2	9540.4	3.2	0
LPG	45218.9	15.1	40718.9	13.6	- 1.5
Gasoline	148626.0	49.7	140226.0	47.2	-2.5
LCO	62122.1	20.8	62122.1	20.8	0
HCO + Slurry oil	21173.2	7.1	28773.0	9.6	+2.5
Coke	12386.5	4.1	16686.5	5.6	+1.5
Total Hydrocarbon (kg/h)	299067.0	100	299067.0	100	
Conversion	72		70		

^a The feed and product distribution presented in a cracking case study [28].

^b Updated cracking products based on previous works on metal oxidized modified ECat [17,20].

^c The kilograms (vacuum gas oil) per hour unit number 5.98 kg/h converts to 1 bbl/d, one barrel vacuum gas oil per day.

known cost respectively. A_M, A_T and A_P are correlation factors due to the manufacturing materials, operating temperature and pressure respectively. The correlation coefficient was assigned a value of 1.0 based on the assumption in the previous study [34]. 'n' is the cost exponent for size/capacity correction. The value of 'n' varies from 0.3 to 0.84, a value of 0.6 was assumed in this study based on the assumptions from a previous study related to CO₂ capture via post-combustion based on MEA-solvent absorption [35].

Since the EPC is sensitive to time changes over the years, the cost must be adjusted to the current year. In the present study, the year 2022 was considered, therefore the EPC was updated to 2022 using the Chemical Engineering plant cost index (CEPCI) via Equation (3).

$$EPC_{current} = EPC_{ref} \left(\frac{CEPCI_{current}}{CEPCI_{ref}} \right) \quad (3)$$

EPC_{ref} and EPC_{current} are the equipment purchase cost of the reference year and current year, respectively. CEPCI_{ref} and CEPCI_{current} represent the chemical engineering plant cost index of the reference and

Table 3
Assumptions for the energy balance on the CLC-FCC unit.

Variable	Value	References
Heat Capacity of VGO at 80 °C, Cp _{VGO,80}	2.67 kJ/kg°C	[36,37]
Heat Capacity of VGO at 220 °C, Cp _{VGO,220}	3.30 kJ/kg°C	[36,37]
Heat Capacity of Catalyst, Cp _{cat}	1.12 kJ/kg°C	[29,37]
Heat Capacity of Cu ₂ O, Cp _{Cu2O}	0.56 kJ/kg°C	[38]
Heat Capacity of CuO, Cp _{CuO}	0.70 kJ/kg°C	[38]
Heat Capacity of CO ₂ at 200 °C, Cp _{CO2}	0.99 kJ/kg°C	[38]
Heat Capacity of CO ₂ at 750 °C, Cp _{CO2}	1.24 kJ/kg°C	[38]
Heat Capacity of Air at 200 °C, Cp _{Air}	1.02 kJ/kg°C	[38]
Heat Capacity of Air at 750 °C, Cp _{Air}	1.14 kJ/kg°C	[38]
Heat of Vaporisation of VGO, ΔH _{vap}	156.00 kJ/kg	[36]
Heat of Cracking reaction of VGO ^b , ΔH _{Crack}	376.56 kJ/kg	[28]
Heat of saturated Steam at 100 °C, ΔH _{s,100}	2675.43 kJ/kg	Steam Tables
Heat of Steam at 520 °C, ΔH _{s,520}	3533.15 kJ/kg	Steam Tables
Heat of Coke adsorption ^b , ΔH _{C-ads}	3372.7 kJ/kg	[28]
Heat of CuO reduction, ΔH _{Red}	288 kJ/mol	-
Heat of Cu ₂ O oxidation, ΔH _{Oxd}	288 kJ/mol	-
Heat of Coke combustion, ΔH _{Comb}	393 kJ/mol	[29]
Heat Losses ^c	5%	Assumption

^a Depending on conversion level, catalyst type, and feed quality, the heat of reaction can vary from 120 BTU/lb to 220 BTU/lb [28].

^b The adsorption of coke on the catalyst is an exothermic process. The heat associated with the adsorption is assumed to be the same as the desorption of coke in the regenerator (ΔH_{C-des} = ΔH_{C-ads}).

^c It was assumed that 5% of the heat coming through the catalyst circulation from the regenerator to the FCC Reactor is losing. Similarly, 5% of heat loss was assumed from the supplied heat through the combustion of coke with CuO. Finally, another 5% of heat loss was assumed from the supplied heat through the oxidation of Cu₂O.

current years, respectively.

3.2. Estimation of operating expenditure (OPEX) and capital expenditure (CAPEX)

The methodology for evaluating the operating expenditure (OPEX) and capital expenditure (CAPEX) in this is presented in Fig. 4. Most of the assumptions used for CAPEX and OPEX appraisal have been documented in the following literature [35,39,40]. CAPEX is often determined as a fraction of the EPC and it includes cost components such as the bare module cost, process contingencies, engineering construction and management. In contrast, the OPEX include fixed and variable operating cost. The latter includes the utilities and raw materials costs. The total cost of labour and supporting facilities was adopted based on the recommendation by Turton et al. [41] while the process and project contingencies were set at 50% of the bare module cost (BMC). The increase in the percentage of contingencies is due to the low technology readiness level of the proposed design. The equipment sizing including the regenerator, air reactor, and FCC riser reactor were estimated based on mass and energy balance. The fixed operating cost (FOC) is calculated as a fraction of the FCI, and the labour cost as shown in Fig. 4. While the variable operating cost (VOC) comprises of the raw material and utility costs. The main raw materials for the proposed CLC-FCC process are the catalyst, VGO oil, compressed air and Cu₂O oxygen carrier. In contrast, the cost of utilities comprises of electrical energy cost used up by pumps, the compressor, blower, cooling water cost, regeneration energy cost and air reactor energy cost.

3.3. Total annual cost (TAC) estimation and cash flow analysis

The total annual cost (TAC) expressed in US\$/year of the CLC-FCC plant is estimated as the sum of annualized OPEX and CAPEX (sum of fixed and variable operating costs). However, the CAPEX is transformed into a constant yearly payment over the entire life of the project. The yearly CAPEX (CAPEX_{annual}) is estimated from equation (4) [35].

$$CAPEX_{annual} = CAPEX \left(\frac{i(1+i)^n}{(1+i)^n - 1} \right) \tag{4}$$

The cost of CO₂ capture ($Capture_{cost}$) is determined using the TAC and the captured CO₂ (equation (5)).

$$Capture_{cost} = \frac{TAC}{CapturedCO_2} \tag{5}$$

In order to determine the prominent profitability, a cash flow analysis was conducted and evaluate the economic viability of the proposed CLC-FCC design. Following the cash flow evaluation, in order to determine the impact of several parameters on the $Capture_{cost}$ a sensitivity analysis was performed.

4. Results and discussion

4.1. Mass and energy balance

Fig. 5 shows the mass balance of the novel CLC-FCC unit with a Sankey Diagram. The amount of catalyst injected into the FCC Reactor is about 4–6 times higher than the cracking feed rate. Assuming that the amount of catalyst is 5.0 times higher than the cracking feed rate, then the regenerated catalyst would have 11.2% of reduced oxygen carriers. As illustrated in Fig. 5, the amount of coke is about 16,686 kg/h, which is deposited on the catalyst (299,067 kg/h) surface in the cracking reaction of VGO with the production of 286,628.7 kg/h cracking products. A large amount of coke deposition on the catalyst surface leads to challenges such as catalyst deactivation and active site blockage.

The spent catalyst is transferred to the FCC-CLC regenerator, in which coke is combusted (burnt off) with oxidized form metal oxides. The mass balance results in the regenerator reactor show that for every 1,700,622.3 kg/h of spent catalyst and 3,820,536 kg/h of oxidized oxygen carrier modified catalyst added, about 3,772,480.18 kg/h of catalysts are regenerated for the FCC unit. It should be mentioned that 96 vol% of coke combustion was achieved using stoichiometric amount of

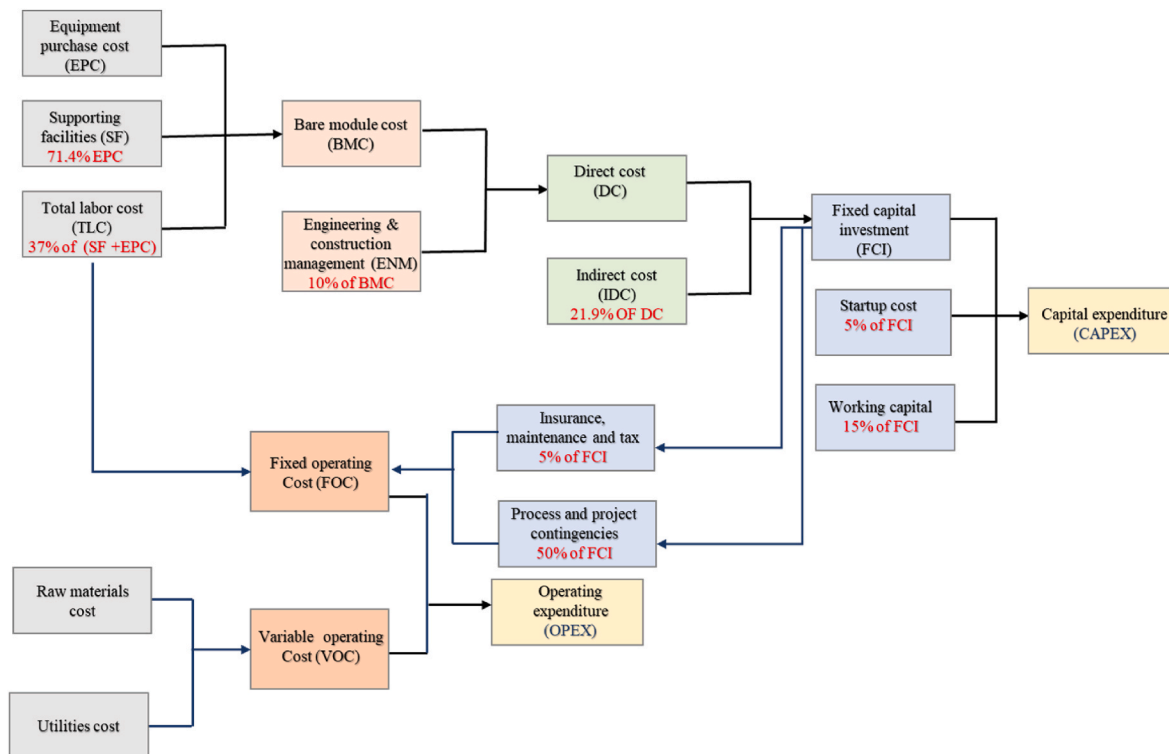


Fig. 4. Methodology for estimating the CAPEX and OPEX for the conceptual integrated CLC-FCC plant.

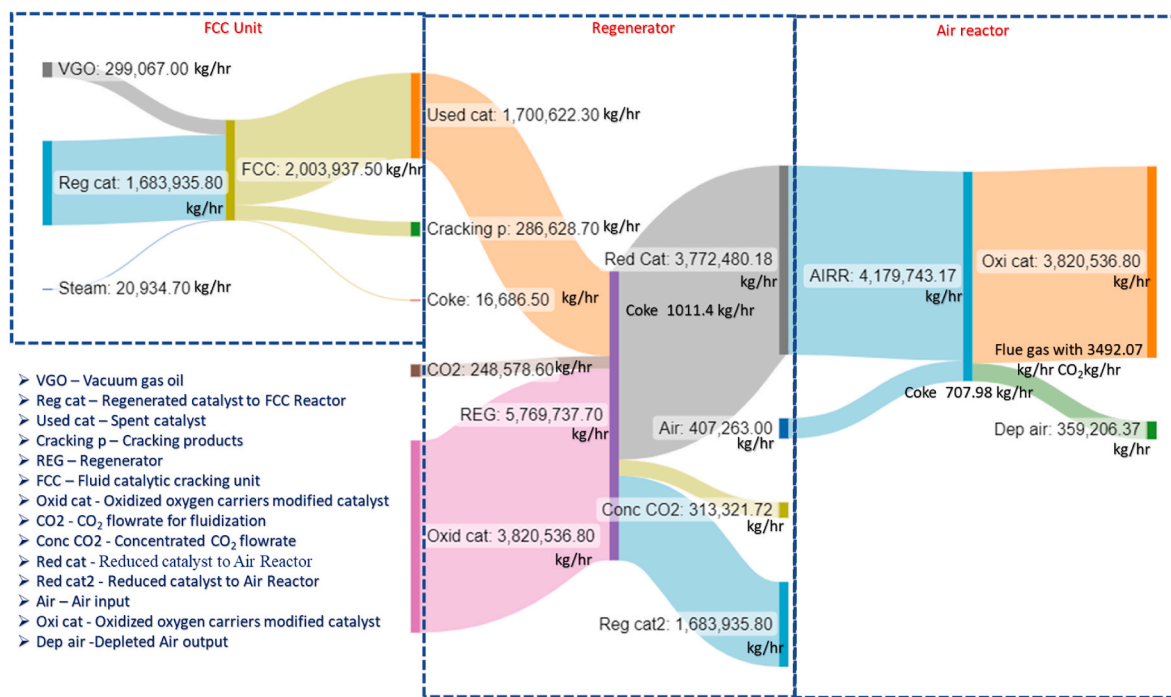
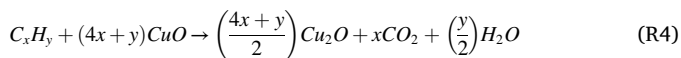


Fig. 5. Sankey diagram representing the mass flow in the proposed CLC-FCC unit.

CuO impregnated on ECat at 750 °C, for 45 min (reaction 4) [17,20].



The required amount of CO₂ for fluidisation in the regenerator was about 248,578.6 kg/h. The value is determined with the assumption that the volumetric CO₂ flow rate for fluidisation is similar to the volumetric flow rate of inert nitrogen as if conventional FCC regenerator operations.

In the air reactor, about 3,772,480.18 kg/h of reduced catalysts are oxidized with 407,263 kg/h of air to produce oxidized oxygen carrier-modified catalysts that are sent back to the regenerator. The reduced Cu₂O can be oxidized to CuO under an air atmosphere at about 700 °C in minutes. The reduced catalyst flow rate (3,772,480.18 kg/h) consists of 11.2 wt % of Cu₂O, which is about 422,517.8 kg/h.

The stoichiometric amount of oxygen for the Cu₂O oxidation to CuO was estimated and the air input was assumed to be 2 times higher than the stoichiometrically required air flow rate (359,206.37 kg/h). Due to the 96 vol % of coke combustion efficiency, approximately 4 wt% of coke (1011.4 kg/h) may be circulated to both FCC and air reactors with the reduced oxygen carriers modified FCC catalysts flow rate. Based on flowrates, 70% of uncombusted coke (707.98 kg/h) goes to the air reactor and 30% of uncombusted coke (303.42 kg/h) goes to the FCC reactor (which is equal to 0.018% of coke on the circulating catalyst to FCC reactor). Therefore, the air reactor flue gas contains 3492.07 kg/h of CO₂ due to the combustion of 707.98 kg/h of uncombusted coke transferred from the regenerator to the air reactor. The proposed CLC-FCC concept is therefore reach approximately 95 vol % of CO₂ capture (56,386.9 kg CO₂/h). However, the CO₂ capture ratio can reach above 99% with an additional oxy-combustion of the remaining coke, which is neither experimentally demonstrated or included in this study.

The energy balance of the integrated CLC-FCC process is computed and compared with the conventional FCC process. That way the energy loss due to CLC can be determined. Details of the mass and energy balance have been meticulously described in Supplementary A of the supplementary materials. The heat loss from the FCC Reactor; (assuming that 5% of the heat coming through the catalyst circulation between regenerator and riser reactor) is 20.47 GJ/h. In contrast, heat loss from the regenerator is higher (34.91 GJ/h). Regenerator heat loss was also

calculated with the assumption that 5% of the supplied heat was through the combustion of coke with CuO. The air reactor only loses 21.26 GJ/h based on the assumption that 5% of the supplied heat is through the oxidation of Cu₂O.

The energy penalty for the novel CLC-FCC integration is compared with an FCC unit that employs oxy-fuel combustion and post-combustion capture. Table 4 shows the comparison of energy penalties. The energy penalty of the proposed CLC-FCC unit (0.21 GJ/ton CO₂) is relatively low compared to the post-combustion unit via amine solvent (3.1–4.2 GJ/ton CO₂) depending on amine solvent and oxy-fuel combustion unit (1.8–2.5 GJ/ton CO₂) based on the purity of supplied oxygen (from 95 to 99.5%) [13,15,42]. Although the energy penalty of post-combustion interaction to FCC unit was provided to be as low as 0.5 GJ/t CO₂ due to the lower net power consumption than oxyfuel combustion [15,16,43], there is not a distinct or apparent reason for why this situation exists.

In order to provide the heat balance on the new CLC-FCC unit as in the conventional FCC unit, the process was designed considering the energy balance by applying chemical looping combustion. As provided in the supplementary, in this new concept, riser reactor requires –139.80 GJ/h, regenerator requires –381.25 GJ/h, and air reactor produces 579.54 GJ/h. The net energy balance is 58.50 GJ/h. Although the net energy balance shows a positive energy, it is significantly important to identify how the energy produced in the air reactor transfer to regenerator and riser reactor. The units in the CLC-FCC must be designed considering the maximum energy transport between air reactor to riser reactor and regenerator. For example, instead of three independent reactors configuration, CLC-FCC concept can be designed as shell-and-tube concept with two stages, in which the air reactor will be the centre and heat transfer to the surrounding shell tubes which could be riser reactor and regenerator. Considering the variety of design configurations in the conventional FCC unit [28], CLC-FCC concept could also be designed and operated with maximum heat transport operation between these units.

Table 4
Technical evaluation of the applicable CCS technologies for FCC unit.

	Post-combustion capture			Oxy-fuel combustion capture			CLC
	Amine	Amine	Amine	95% O ₂	97% O ₂	99.5% O ₂	CuO
Composition of flue gas (regenerator)							
CO ₂ (mol %)	17.7	13.5	16.3	84.3	–	89.4	98.06
N ₂ (mol %)	77.5	72.9	74.3	1.7	–	0.1	–
O ₂ (mol %)	1.2	2.7	1.04	2.8	–	2.8	–
H ₂ O (mol %)	3.6	10.0	7.3	6.9	–	6.9	1.9
Summary of technical results							
CO ₂ capture (%)	74.0	90.4	85.5	90.5	–	99.9	95.6
CO ₂ purity (vol. %)	–	99.9	–	95.2	–	96.1	99.0
CO ₂ product (t/h)	62.9	101.7	55.22	101.6	58.33	112.3	56.4
Net Power Consumption (GJ/h)	–	56.88	28.23	256.32	123.2	266.40	12.17
Energy Penalty (GJ/t)	–	0.56	0.56	2.52	2.11	2.37	0.21
Reference	[12]	[16]	[15]	[16]	[15]	[16]	This study

4.2. Economic analysis

4.2.1. Equipment purchase cost (inside battery limits)

Details of the EPC including the cost contribution of each piece of process equipment are presented in Fig. 6(a). The evaluation of EPC is based on the presupposition that all the capital equipment has been acquired recently, rather than being adapted or modified from existing installations. It should be mentioned that the EPC cost presented is the

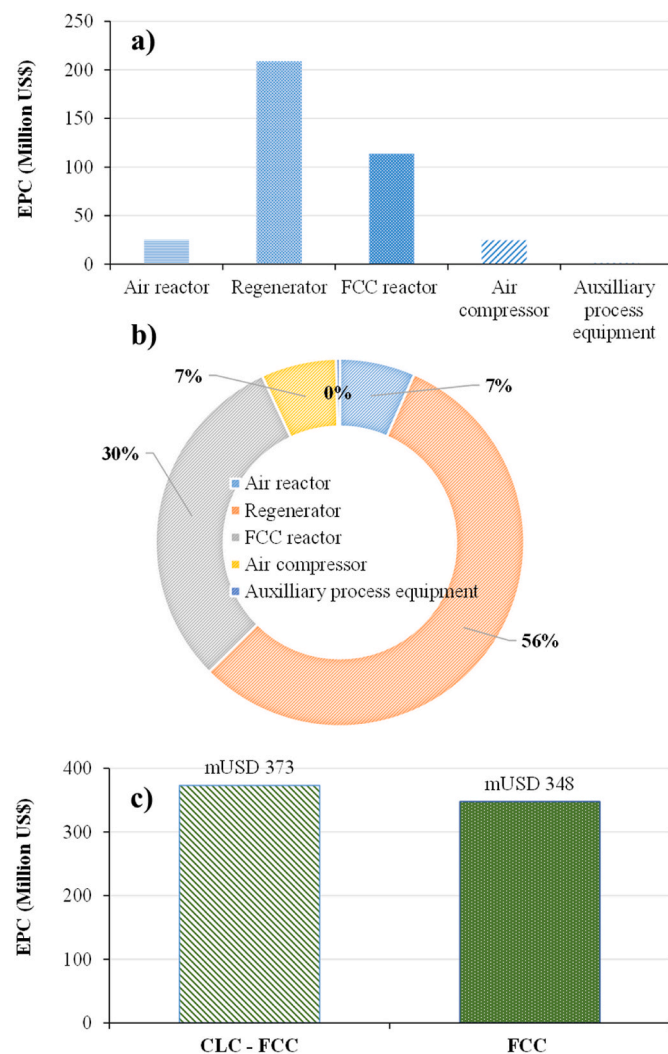


Fig. 6. (a) Overview of the equipment purchase cost (b) Percentage contribution of each equipment (c) EPC comparison between CLC-FCC and FCC unit.

inside battery limit cost that includes purchasing and installing costs for all process equipment. The regenerator, FCC and air reactor account for the majority of the EPC. The catalyst regenerator cost accounts for 56% of the total EPC, while the FCC reactor accounted for 30% of the total EPC. Compared to the regenerator and FCC reactor, the purchase cost for the air reactor is lower (7% of the EPC) (Fig. 6(b)). Auxiliary equipment such as heat exchangers, blowers, feed pumps and boilers only constitute 1% of the EPC.

The regenerator plays a crucial role in optimizing the overall profitability of the CLC-FCC process by helping in restoring the catalyst activity and improving the heat balance in the reactor. Designing an efficient regenerator that could withstand high temperatures with enhanced coke burning rate and prolonged operating cycle requires an advanced reactor material. The increased cost of the FCC regenerator could be attributed to the extra cost of units such as spent catalyst and air distributor, cyclones, catalyst buffer and baffles. These units are important for the efficient functioning of the FCC regenerator. The EPC for the novel CLC-FCC unit is compared with that of an FCC unit without CO₂ capture and presented in Fig. 6(c). The EPC for the CLC-FCC unit is 25.33 million US\$ (1.1 times) higher than the conventional FCC unit due to the requirement of extra process equipments, which is also similar for CO₂ capture from FCC with amine scrubbing technology [12]. The authors noted that the EPC of the amine CO₂ capture FCC plant is 1.25 times higher than a conventional FCC unit [12].

4.2.2. Capital expenditure (CAPEX) and operating expenditure (OPEX)

The raw materials and utilities costs are the major component of OPEX, therefore they are first estimated and compared with an FCC unit integrated with oxy-fuel combustion and post-combustion capture technologies. The results of the raw materials and utilities costs are

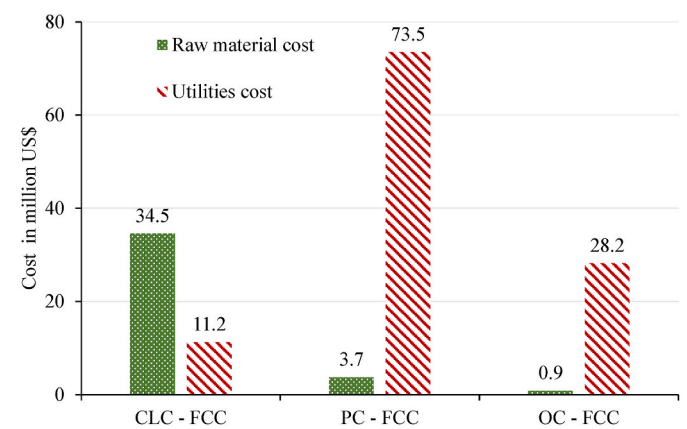


Fig. 7. Total raw material cost and utilities cost between different capture methods integrated into FCC units. Data for the post-combustion (PC) and oxy-fuel combustion (OC) were obtained from Ref. [13].

presented in Fig. 7. The solvent (amine) based CO₂ capture (post-combustion) had the highest cost of utilities (mUS\$73.5). The rise in utility costs was ascribed to the necessity for low-pressure steam needed for the application of amine in order to capture CO₂ from regeneration, as well as an extra demand for water in the cooling tower. The raw material cost increases in the following order: oxy-fuel combustion (mUS\$0.9) < post-combustion (mUS\$3.7) < CLC (mUS\$34.5). The high cost of raw materials for the CLC-FCC concept is attributed to the cost of the oxygen carrier (metal oxides). As oxygen carriers, CuO is prone to natural decay resulting from reduction-oxidation cycles and are often replaced over time, thereby increasing the cost throughout the process's lifetime. The post-combustion unit also showed a moderately high raw material cost due to the need for MEA solvent, corrosion inhibitor, sodium carbonate to reclaim MEA, activated carbon for hydrocarbon removal as well as the disposal costs [14]. These additional raw material costs are not required for the oxy-fuel combustion capture system.

A breakdown of the CAPEX and OPEX as well as a detailed calculation is presented in Table 5. It should be emphasized that the CAPEX and OPEX reported herein is relatively higher than most CO₂ capture plants. As mentioned before, the economic evaluation was carried out on the premise that the capital equipment would be acquired through purchase rather than retrofitting or configuring existing equipment. Also, the EPC is for the year 2022 in which inflation has a role to play. Moreover, the proposed CLC-FCC unit is a novel technology without only a few experimental verifications. Since there is no adequate commercial or pilot scale plant, project and process contingencies that is 50% of the fixed capital investment (FCI) was assumed, thereby contributing significantly to the increasing CAPEX and OPEX.

4.2.3. Cash flow analysis

A detailed cash flow analysis was conducted to evaluate the CO₂ capture cost for the CLC-FCC with the assumption that the plant has a lifetime of 20 years and a 10% discount rate [35]. While the discount rate was selected based on prior study assumptions, to assess the effect of different discount rates on the cost of CO₂ capture, a sensitivity analysis was also performed. Through the cash flow analysis, the economic implications of integrating CLC into FCC units can be determined. Details of the cash flow analysis calculations can be found in Supplementary B of the supplementary materials. The cash flow analysis results reveal a yearly basis average CO₂ capture cost of 0.0106 US\$/kg of CO₂ (~10.6 US\$/ton of CO₂) for the integrated CLC-FCC unit (Fig. 8). The CO₂ capture cost of a CLC-FCC unit (~10.6 US\$/ton of CO₂) is very similar to

Table 5
Breakdown of the CAPEX and OPEX calculations.

Cost component	Factor	Cost, (mUS \$)
Equipment purchases cost (EPC)	1%	373.0
Supporting facilities (SF)	71.4% EPC	266.3
Total labor cost (TLC)	37% (SF + EPC)	236.6
Bare module cost (BMC)	EPC + SF + TLC	639.4
Engineering, construction and management (ENM)	10%(BMC)	63.9
Direct cost (DC)	BMC + ENM	703.3
Indirect cost (IDC)	21.9%DC	154.0
Fixed capital investment (FCI)	DC + IDC	857.4
Working capital (WC)	15%FCI	128.6
Start-up cost (SUC)	5%FCI	42.9
CAPEX	FCI + WC + SUC	1028.8
Raw material cost (RMC)	RMC	34.5
Utilities cost (UC)	UC	11.2
Variable operating cost (VOC)	RMC + UC	45.8
Contingencies (PPC)	50%FCI	428.7
Insurance, maintenance and tax (IMT)	5% FCI	42.9
Fixed operating cost (FOC)	TLC + PPC + IMT	708.1
OPEX	FOC + VOC	753.9

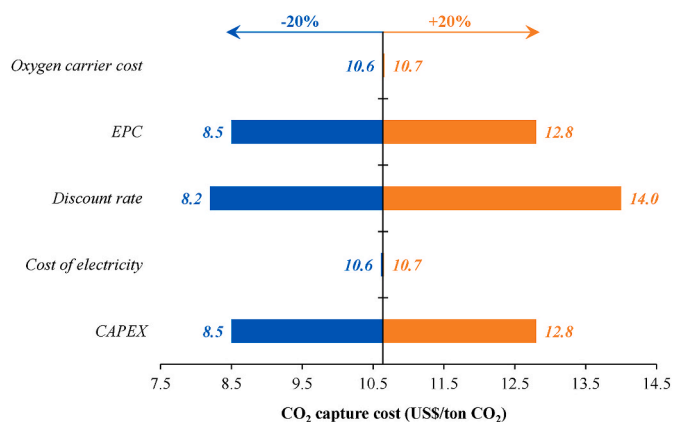


Fig. 8. Sensitivity analysis of Oxygen carrier cost, EPC, Discount rate, Cost of electricity, and CAPEX on the CO₂ capture cost.

the CO₂ capture cost of the application of CLC to solid fuel power plant in an EU project (10–40 €/t CO₂). Considering that there are relatively few recent studies on the economic feasibility of CO₂ capture plants integrated with FCC units, the CO₂ capture cost of CLC-FCC unit was lower than the reported FCC-post-combustion combination (75–110 €/ton CO₂) and FCC-oxy combustion combination (55–85 €/ton CO₂) [13,15,42]. Furthermore, Nwaoha et al. [44] reported a capture cost of 0.10 US\$/kg CO₂ from an amine post-combustion capture plant implemented in a 1.2 million metric tonne per annum cement plant. Another study reported a capture cost of 0.015 US\$/kg of CO₂ with an indirect carbonation CO₂ capture process in the cement industry [45]. Overall, the presented capture cost reported in this study is lower when compared to the post-combustion and oxy-fuel combustion costs for other FCC and cement industries.

The CO₂ avoidance cost was estimated from equation (6).

$$C_{avoidance} = \frac{CC_{ref} - CC_{considered}}{E_{ref} - E_{considered}} \quad (6)$$

Where $C_{avoidance}$, CC_{ref} and $CC_{considered}$ represents the CO₂ avoidance cost, capture cost of the reference plant and capture cost of the FCC-CLC plant respectively [25]. E_{ref} and $E_{considered}$ are values of electricity generation in reference plant and FCC-CLC plant respectively. The FCC-oxy combustion capture was used as a reference plant with a capture and electricity cost of 126.9 US\$/ton CO₂ and 34.2 MW respectively [26]. The cost of CO₂ avoided for FCC-CLC process is 96.92 US\$/tonCO₂. This value is quite low compared to FCC- post combustion (122.9 US\$/ton CO₂) and FCC-oxy combustion (159.7 US\$/ton CO₂) processes [26].

Sensitivity analysis provides the results to evaluate the impact of several economic factors on CO₂ capture cost. Factors such as CAPEX, cost of electricity, EPC, oxygen carrier cost, and discount rate are presumed to impact the CO₂ capture cost. A local sensitivity analysis (LSA) was performed to selectively identify and rank the parameters that had the most impact on the capture cost. LSA analysis involves the varying of one input parameter with ±20% of their nominal values and assessing the impact of the variation on the capture cost while the other parameters are kept at a constant value.

Fig. 8 shows that the oxygen carrier cost and cost of electricity have no significant impact on the CO₂ capture cost. There is no quantifiable change in the CO₂ capture cost with a 20% increase or decrease in both parameters. In contrast, parameters such as discount rate, EPC, and CAPEX had a significant impact on the capture cost. For instance, the CO₂ capture cost increased by 20.2% (12.8 US\$/ton) with a 20% elevation in the EPC. In the same way, a decline in the EPC by 20% led to a 20.2% decrease (8.5 US\$/ton) in CO₂ capture cost. The discount rate had the greatest influence on the CO₂ capture cost with almost 31.5% elevation in the capture cost with a 20% rise in the discount rate. Reducing the CAPEX by 20% also led to a 20.2% decrease in the capture

cost.

5. Conclusions

For the first time, a techno-economic feasibility and sensitivity analysis for a novel CO₂ capture process – CLC integrated FCC unit – were performed based on a case study with an FCC feed rate of 50,000 barrels per day. This study provides a novel CLC-FCC conceptual design with a rigorous calculation to appraise the mass and energy balance.

- The proposed CLC-FCC unit provides a relatively low energy penalty (0.21 GJ/ton CO₂) compared to the other two alternatives: the post-combustion unit via amine solvent (3.1–4.2 GJ/t CO₂) and oxyfuel combustion unit (1.8–2.5 GJ/t CO₂).
- Integration of CLC to FCC unit increased the EPC by 25 mUS\$ (373 mUS\$ for CLC-FCC and 348 mUS\$ for conventional FCC).
- The requirement of oxygen carriers in the CLC-FCC concept increased the raw material cost in the following order: oxy-fuel combustion (0.9 mUS\$) < post-combustion (3.7 mUS\$) < CLC (34.5 mUS\$). However, the CLC-FCC concept provides the lowest utility cost; CLC (11.2 mUS\$) < oxy-fuel combustion (28.2 mUS\$) < post-combustion (73.5 mUS\$).
- CAPEX and OPEX for the CLC-FCC concept were determined as 1028.8 mUS\$ and 753.9 mUS\$, respectively.
- The CLC-FCC concept shows the lowest CO₂ capture cost (~10.6 US\$/ton of CO₂) compared to the other two alternatives; the post-combustion unit via amine solvent (75–110 €/ton CO₂) and oxy-fuel combustion unit (55–85 €/ton CO₂).

Considering the importance of industrial decarbonisation and the contribution of oil-refining to CO₂ emissions, this study demonstrates that CLC is a promising technology and will potentially play a crucial role in the industrial decarbonisation for refineries thanks to its low energy penalty and low CO₂ capture cost. However, the process requires extensive experimental and modelling works to validate the results and identify the optimum process conditions. All mass and energy balance were performed rigorously and presented in the supplementary materials. Furthermore, a sensitivity analysis was performed to estimate the effect of economic indicators on the CO₂ capture cost. An error analysis in this case would be in the form of a detailed uncertainty analysis using monte Carlo simulation. This is currently beyond the scope of this study as the authors focused on preliminary economic evaluation. However future studies would explore detailed uncertainty analysis, development of a comprehensive dynamic process simulation and environmental assessment.

CRedit authorship contribution statement

Fatih Güleç: Conceptualization, Project administration, Formal analysis, Funding acquisition, Investigation, Visualization, Supervision, Writing – original draft, Writing – review & editing. **Jude A. Okolie:** Conceptualization, Methodology, Investigation, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Ahmet Erdogan:** Conceptualization, Methodology, Investigation, Writing – original draft, Writing – review & editing.

Declaration of competing interest

I would like to confirm that there are no known conflicts of interest associated with this publication and there has been no significant financial support for this work that could have influenced its outcome. I confirm that the manuscript has been read and approved by all named authors and there are no other persons who satisfied the criteria for authorship but are not listed. I further confirm that the order of authors listed in the manuscript has been approved by all of us.

I confirm that we have given due consideration to the protection of intellectual property associated with this work and that there are no impediments to publication, including the timing of publication, with respect to intellectual property. In so doing we confirm that we have followed the regulations of our institutions concerning intellectual property.

We further confirm that any aspect of the work covered in this manuscript that has involved either experimental animals or human patients has been conducted with the ethical approval of all relevant bodies and that such approvals are acknowledged within the manuscript.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.energy.2023.128663>.

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