



Full Length Article

Techno-economic analysis of integrated carbon capture and utilisation compared with carbon capture and utilisation with syngas production

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ABSTRACT

Currently, excessive CO₂ emissions have become a global challenge due to their influence on the climate. According to the Paris Agreement, global warming should be limited to 1.5 °C by 2100. Carbon capture and utilisation (CCU) are attractive as they can both reduce CO₂ content and utilise CO₂ as a carbon resource. However, in conventional CCU processes, CO₂ needs first to be extracted and purified for the following utilisation. In contrast, the recently reported Integrated Carbon Capture and Utilisation (ICCU) was designed to realise the overall process in one reactor, where CO₂ is captured by adsorbents (e.g., CaO) and utilised *in-situ* with the introduction of a reducing agent (e.g., H₂). This ICCU technology can promote CO₂ conversion with fewer intermediate steps, leading to a reduction in overall cost. Energy and economic analysis of ICCU are thus urgently required. According to several recent research, the operational cost of ICCU has been reported to be cheaper than that of CCU. However, a comprehensive view of ICCU is still expected due to further application. This paper focuses on comparing ICCU and conventional CCU processes based on Aspen simulations covering mass balance (i.e., CaCO₃ consumption, purge production, annual CO production), energy balance, the total annual cost and the CO cost, etc. Analysis shows that the ICCU process can produce more CO (1.20 Mt year⁻¹), less purge (0.21 Mt year⁻¹), and less consumption of CaCO₃ (0.62 Mt year⁻¹) with higher energy efficiency (37.1 %) than the CCU process. The results also show that the total annual cost of ICCU is \$867.07 million, corresponding to a total cost of CO of \$720.25 per tonne. In contrast, CCU has higher costs, with a total annual cost of \$1027.61 million and a total cost of CO of \$1004.53 per tonne. The Cost of CO₂ Avoided of ICCU (317.11\$/ton) is much lower than that CCU (1230.27 \$/ton). Therefore, ICCU was confirmed as a better choice for further industrial applications. In addition, H₂ is shown to have a significant influence on economic performance, which remains a challenge for further application.

1. Introduction

Carbon Capture and Storage (CCS) is the technology that removes CO₂ from carbon sources, compresses and transports it to a storage site (e.g., underground or ocean bedrock) without releasing it back into the atmosphere [1–4]. It has been researched intensively for its potential to reduce CO₂ emissions in the atmosphere [5]. CCS capacity by 2050 is estimated to be approximately 700 million tons annually, corresponding to 10 % of what is required [6]. However, CCS entails high initial capital investment and would drastically reduce power plant efficiencies, with potential negative environmental impacts and risk of accidental leakage

during long-term storage [7,8].

Carbon Capture and Utilisation (CCU) is complementary to CCS as it can be used to reduce CO₂ emissions from fossil resources by converting industrially emitted CO₂ into chemicals and fuels [9–11]. In various CCU processes reported, CO₂ is commonly absorbed in solvents such as amines, KOH, or methanol, followed by the regeneration of sorbents in a desorption step to obtain concentrated CO₂ [12–14]. The captured CO₂ can then be converted to chemicals such as carbonates [15], poly (carbonates) [16], carbamate derivatives [17], carboxylic acids [18,19] using a range of catalysts that include main-group metal complexes (e.g., Mg, Al, Ca, and In), transition-metal complexes (e.g., Zn, Fe, Cr, and Co),

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and organo-catalysts [20]. However, the CO₂ purification process increases CO₂-supply cost and greenhouse gas emissions [21]. Therefore, it is desirable to decrease the cost of the CO₂ desorption process along with increasing the environmental and economic benefits of CCU [22].

Aspen Plus® is widely used to design a new process, optimise operations of a full process, and predict the behavior of a process using basic engineering relationships (i.e. mass and energy balances) [23,24]. For example, researchers have reported simulations for the carbonate-looping steam cycle for a large coal-fired plant [25], a coal-fired power plant with CCS [26], CCUS for a coal-based power plant with the production of urea, methanol or sulfur [27]. Syngas is a mixture of H₂ and carbon monoxide (CO). It can be utilised to produce valuable fuels and chemicals via the Fischer-Tropsch process, especially in coal-to-liquid and gas-to-liquid processes [28,29]. The reverse water-gas shift (RWGS) reaction is one of the most established reactions to convert CO₂ into syngas [30,31]. Besides, the global CO market size was estimated to increase from \$11.3 billion in 2021 to \$13.15 billion in 2022, reaching \$23.19 billion in 2026 [32]. Steven *et al.* reported the techno-economic for the power-to-syngas (PtS) technology that sustainably utilises CO₂ from syngas into syngas [33]. By comparing a series of cases in this research to a referred syngas plant (the levelized syngas production cost of 6.94 \$/GJ), the levelized syngas production costs of PtS scenarios in this research range from 8.56 to 13.64 \$/GJ. Jeehoon *et al.* compared the CCU process (monoethanolamine-based chemical absorption and utilisation into syngas) with CO₂ emission from power plants and ironmaking/steelmaking plants, which showed that the minimum selling price of syngas from power plants was 19.31 \$/GJ and ironmaking/steelmaking plant was 16.02 \$/GJ with its market prices ranging from 7.82 \$/GJ to 23.25 \$/GJ [34]. Although these efforts have been made, the competitive costs of CO₂ to syngas are still an obvious challenge for further application.

Integrated Carbon Capture and Utilisation (ICCU) has been reported recently to improve CCU by utilising the captured CO₂ directly. Gassner and Leitner integrated conversion with capture through the hydrogenation of CO₂ to formic acid in aqueous amine solutions [35]. Lu *et al.* captured and converted CO₂ into formic acid using a photo-electrochemical system assisted by an aqueous-ionic liquid (1-amino-propyl-3-methylimidazolium bromide) solution [36]. Scott *et al.* demonstrated the conversion of saturated aqueous solutions (CO₂ overpressure 5–10 bar) of monoethanolamine (MEA) into the corresponding formate adducts [37]. In the ICCU processes, CO₂ was converted into methanol [38–42], oxazolidinones [43,44], urea [45–47], methyl formate [48], organic acids [49], syngas [50]. Iyer *et al.* reported an integrated process wherein CO₂ desorption and dry reforming of the CO₂-methane mixture simultaneously occurred in one reactor to produce syngas [50]. This was a partial integration process, which involved one separation column and one reactor. Luis *et al.* also reported an ICCU strategy that combined CO₂ capture and conversion in two reactors with unsteady-state operation under isothermal conditions to produce syngas [51], which also required two reaction systems.

Calcium looping (CaL) using CaO-based sorbents is a promising alternative to both oxy-combustion and chemical solvent absorption [52,53]. The mechanism of the CaL process includes carbonation of CaO to give CaCO₃ in a carbonator operating normally at 650 °C under atmospheric pressure, and calcination of CaCO₃ to give CaO at typically over 930 °C under a highly concentrated CO₂ environment (70–90 vol%) [54,55]. Compared to an amine-based post-combustion capture system where the heat of absorption cannot be recovered, the heat of the carbonation reaction in the CaL process can be recovered at high temperature by steam evaporation, superheating, or reheating [53]. It has already been demonstrated on a scale of up to 1.9 MWh [54,56].

The integration of the CaL process and RWGS reaction at the same temperature in a single reactor to achieve the capture and conversion of CO₂ by using CaO-based materials has been reported by our group [57,58]. The ICCU process with syngas production using only CaO as both sorbent and catalyst has been demonstrated with over 75 % CO₂

conversion efficiency at 600–700 °C [58]. This process can eliminate the energy requirement, corrosion, and transportation issues associated with CCS and CCU. Bin *et al.* have also reported combining the CaL process and RWGS, where CO₂ conversion efficiency reached nearly 90 %, and CO selectivity was close to 100 % by introducing transition metals (Co and Fe) into CaO [59]. These studies mainly focused on materials development and optimisation of process conditions, there is an urgent need for technical and economic performance analysis.

Currently, the techno-economic analysis of ICCU has been reported based on the capture with Zeolite 13X [50], MEA/K₂CO₃/ABS/amine-based resin [60], methanol [22], ionic liquids [61], MDEA and PZ solvent [62], etc. In addition, it is noticeable that ICCU is not always better than CCU in cost when ICCU is compared with CCU [22,59]. For example, it was estimated that ICCU based on CaL costed \$165/t CO₂ (only operational cost was considered) which was less than the simply combined process of CaL and RWGS conversion (\$393/t CO₂) [59]. In contrast, Jens *et al.* compared CCU and ICCU (using methanol to capture CO₂ from raw natural gas to produce methyl formate) to conclude that only if CO₂ in the input was 30 mol%, ICCU could be cheaper than CCU [22]. When CO₂ concentration was lower, ICCU required a higher cost than that of CCU, due to the higher heating demand of the separation of methyl formate and byproduct water from methanol. However, in this CaL and RWGS-based ICCU technology, the syngas product was shown high purity without purification [58].

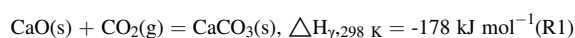
The primary aim of this study is to compare the CCU and ICCU processes based on the CaL process and RWGS reaction using the same reference, namely a coal-fired power plant (CFPP) [63]. In the CCU process, as shown in Fig. 1a, CO₂ is captured and concentrated by the CaL technology; then, the released CO₂ is used as a feedstock for the RWGS reaction reactor to produce syngas. On the other hand, in the ICCU process, as shown in Fig. 1b, the CaL reactor and the RWGS reactor are integrated, where the CO₂ capture, desorption, and utilisation occur isothermally in one reactor by switching the inlet gas between CO₂ sources (e.g., flue gas) and H₂. Therefore, this work is the first to present techno-economic analysis of ICCU with syngas production, by comparing CaCO₃ consumption, purge production, annual CO production, energy efficiency, the total annual cost and the CO cost as well as the Cost of CO₂ Avoided.

2. Process simulation and methodology

A coal-fired power plant with a net electrical power of 600 MW and a net efficiency of 40.6 % was used as a reference plant [63]. Software Aspen Plus® was used to develop the processes of CCU and ICCU to estimate material balances, energy and utility requirements. The reactors were simulated using the property method of the Peng–Robinson with Boston–Mathias modifications (PR–BM), which is suitable for gas processing and refinery applications and provides accurate results for hydrocarbon mixtures and light gases, such as H₂ and CO₂. The reactors were modeled as stoichiometric reactor blocks. The outlet flue gas in this power plant had a flow rate of 540.1 kg s⁻¹ and its composition is summarised in Table 1 [64].

2.1. CCU model description and assumption

The CCU process shown in Fig. 2 was designed ideally without adding CaO, which came instead from the calcination of CaCO₃. The specific assumptions are shown in Table 2. The following reaction was modelled in the carbonator:



CO₂ capture efficiency was assumed to be 85 % when the mole ratio of CaO to CO₂ was ca. 5 [63–66]. A steam generator extracted the excess heat (Q1) from the exothermic CO₂ capture reaction [63,64,66].

A cyclone (CYCLON1) was used to separate the solids from the

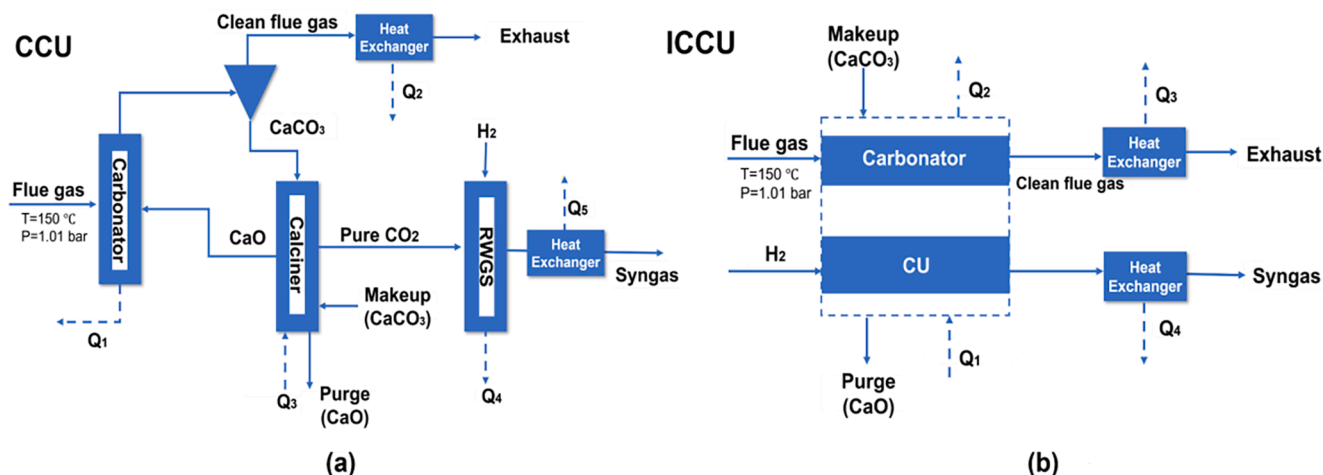


Fig. 1. Overview of (a) the carbon capture and utilisation (CCU) process and (b) the integrated carbon capture and utilisation (ICCU) process.

Table 1

Composition of the flue gas.

Component	Volume fraction (%)
CO ₂	12.0
N ₂	73.7
H ₂ O	8.3
O ₂	5.5
SO ₂	0.5

decarbonised flue gas, leaving the carbonator with a separation efficiency of 100 %. The flue gas stream was then cooled down to approximately 279 °C with the extracted heat Q2 [63], and the solid stream separated from the cyclone was transferred to the calciner. In the calciner, the temperature was maintained at ca. 900 °C yielding a 90 % conversion of CaCO₃. In order to make up for activity decay, the purge rate was assumed to be 4.6 %. It was reported that 5 % was set to keep the activity of the sorbent [63,65]. The heat for the endothermic calcination reaction (Q3) was provided by the combustion of natural gas using air.

Furthermore, a makeup stream (MAKEUP) of fresh limestone consisting of 100 % CaCO₃ was constantly fed into the calciner. The stream out of the calciner (CALCOUT) entered another cyclone (CYCLON2) with a separation efficiency of 100 %. The gas stream (CO₂) was separated from the solids (CaO) that were carried back to the carbonator. In the RWGS reactor, CO₂ was transformed into CO and H₂O, as shown in R2. The temperature was kept at ~650 °C with an excess of hydrogen

(H₂: CO₂ = 3: 1 M ratio) and 55 % conversion of CO₂ into CO [67]. After the reaction, the gas stream was cooled down to approximately 100 °C, providing heat output Q5.



2.2. ICCU model description and assumption

In practice, the ICCU process is conducted in one reactor. However, for illustration, the process in Aspen was simulated in two reactors with the same conditions (Fig. 3). CO₂ capture efficiency was assumed to be the same as the capture process in CCU, with 85 % CO₂ capture efficiency [63–66]. Heat Q1 was also obtained from the exothermic CO₂ capture reaction, which was set at 650 °C. A cyclone (CYCLON1) separated the solids from the decarbonised flue gas, leaving the carbonator with a separation efficiency of 100 %. After that, the flue gas stream was cooled down to around 279 °C with the extracted heat Q2. The solid stream separated from the cyclone was transferred to the reactor (CU), where CO₂ was converted into CO at 650 °C with the addition of excess hydrogen (H₂:CO₂ = 3:1 M ratio). The conversion is assumed to involve two steps, CaCO₃ into CaO and CO₂ into CO, with efficiencies of 55 % and 75 %, respectively [57]. Heat Q3 was generated in CU. After CU, the gas stream was separated by the CYCLON2 with a gas–solid separation efficiency of 100 %, and then cooled down to approximately 100 °C with heat Q5 and the solids (CaO) were carried back to the carbonator with a purge ratio of 1 %. The purge of ICCU is lower than CCU because in non-

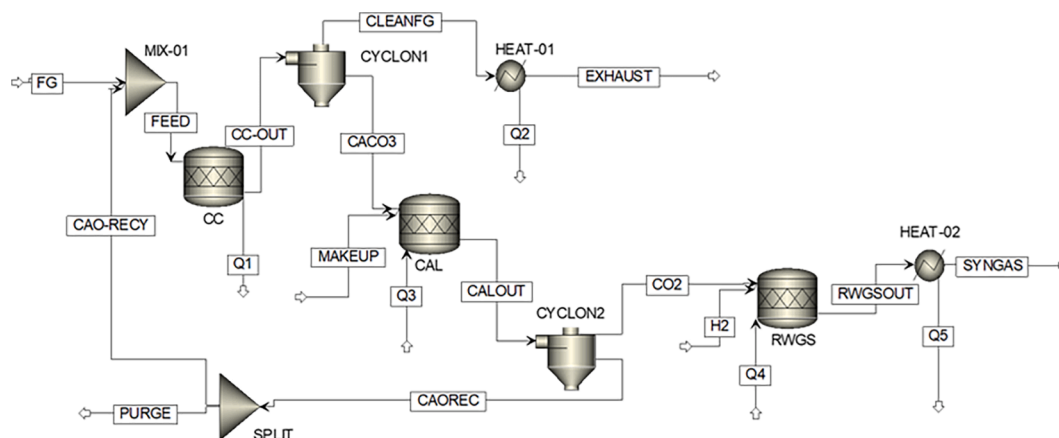


Fig. 2. Process model of the CCU process in ASPEN PLUS®.

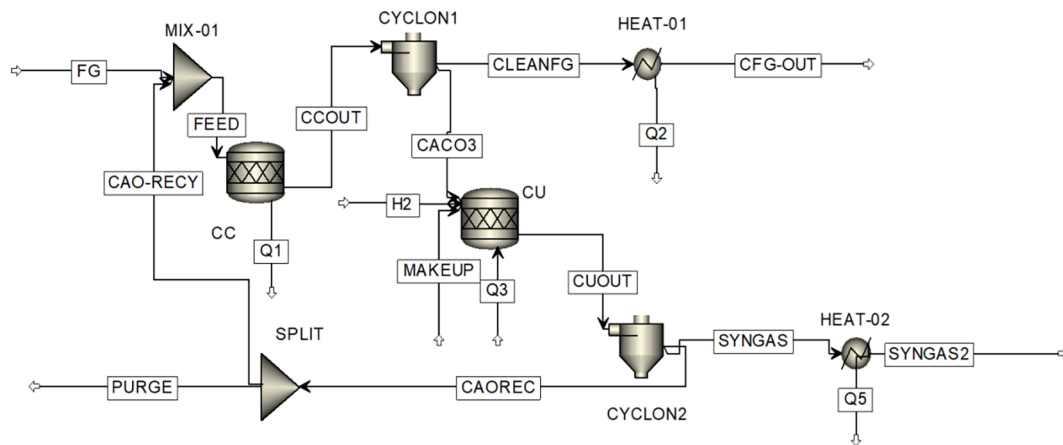


Fig. 3. Process model of the CCU process in ASPEN PLUS®.

isothermal CCU the CaCO₃/CaO stability is much less than that in isothermal ICCU [58].

All the operating conditions of the ICCU process are shown in Table 2.

2.3. Energy efficiency

Energy efficiency for the power plant with CCU/ICCU was defined as the ratio of the total output to input energy:

$$\eta_{\text{energy efficiency}} = \frac{\sum E_{\text{out}}}{\sum E_{\text{in}}} \times 100\% \quad (1)$$

where $\sum E_{\text{out}}$ is the sum of the electricity from the coal-fired power plant

Table 2
Process parameters of CCU and ICCU [25,67–72].

Parameter	CCU	ICCU	
Flue gas	Pressure (bar)	1.01	1.01
	Temperature (°C)	150	150
	Mass flow (kg s ⁻¹)	540.10	540.10
H ₂	Pressure (bar)	1.01	1.01
	Temperature	150	150
	Mass flow (kg s ⁻¹)	13.85	13.85
Makeup (100 % CaCO ₃)	Pressure (bar)	1.01	1.01
	Temperature (°C)	25	25
	Mass flow (kg s ⁻¹)	52.68	21.50
Carbonator	Temperature (°C)	650	650
	Pressure (bar)	1.01	1.01
	CO ₂ capture efficiency	85 %	85 %
	CaO:CO ₂ mole ratio	6	6
Calcination	Temperature (°C)	900	
	Pressure (bar)	1.01	
	Calcination efficiency	90 %	
RWGS reactor	Temperature (°C)	650	
	Pressure (bar)	1.01	
	Conversion	55 %	
	H ₂ /CO ₂ mole ratio	3	
CU reactor	Temperature (°C)		650
	Pressure (bar)		1.01
	CaCO ₃ -to-CaO Conversion		55 %
	CO ₂ to CO Conversion		75 %
Purge ratio		4.6 %	1 %

(600 MWe) and the heat from CCU/ICCU. $\sum E_{\text{in}}$ is the sum of the energy required for the coal-fired power plant (1478.33 MW) and the CCU/ICCU. To convert the heat into electricity, herein the efficiency of generating electricity was about 35.3 % [73].

2.4. Assumptions and method of economic evaluation

In these two cases, total annual capital cost (ACC) and operation and maintenance (O&M) costs were considered. The sum of ACC and O&M was the total annual cost (TAC), as shown in Eq. (2).

$$TAC = ACC + O\&M \quad (2)$$

The O&M costs mainly constituted two parts, namely, fixed and variable O&M costs. Fixed costs included four contributions: annual maintenance cost, direct labor cost, property taxes and insurance as well as administrative, support and overhead cost. The variable costs were associated with the cost of catalyst, H₂, natural gas, and CaCO₃.

ACC was a combination of capital recovery factor (CRF) and total capital investment costs (C_{total}), which is expressed in equation Eq. (3).

$$ACC = C_{\text{total}} \times CRF \quad (3)$$

The equipment costs were assumed equal to the overnight costs, without considering any scaling effect, due to the modularity and the simplicity of installation of both these technologies. And C_{total} was representative of the total capital investment cost, involving the total cost of equipment, owner's cost and contingency and land permitting and surveying costs during plant construction. Total costs of equipment are listed in Table 3.

The capital cost of each component (C_m) was empirically estimated using the scaling factor exponent, through Eq. (4) [74,76–78]. The sum of the equipment cost was defined as C.

$$C_m = C_r \left(\frac{S}{S_r} \right)^f \quad (4)$$

where the C_r and f represent the reference cost (with reference size S_r) and equipment scaling factor exponent, respectively.

Table 3
Parameters of the scaling function for capital cost estimation.

	Cr	Scaling parameter	S _r	f	Ref.
Carbonator	354	Heat duty, MW	1027	0.98	[74]
Calciner	14	Outlet, m ³ /s	839	0.98	[74]
RWGS reactor	32	Input, t/h	43	0.65	[75]
Heat exchanger	187	Heat duty, kW	150	0.78	[75]
Steam turbine, generator and auxiliaries' cost	246	Net power, MW	569	0.69	[74]

The plant was assumed to have 8,000 operating hours annually, a 25-year plant lifetime (n in Eq. (5)), and an interest rate of 8 % (i in Eq. (5)). The CRF was defined as the ratio of constant annuity to the present value at a period with a certain interest, as shown in Eq. (5) [79].

$$CRF = \frac{i(1+i)^n}{(1+i)^n - 1} \quad (5)$$

To evaluate the technology of ICCU and CCU systems, the cost of CO (\$/t CO) was also calculated according to Eq. (6).

$$\text{Cost of CO} = \frac{TAC}{\text{AnnualCOproduction}} \quad (6)$$

Parameters for capital cost estimation with their reference values are summarised in Table 4. The costs of other components such as water pumps, splitters, mixers and separators were not included.

In addition, according to Eq. (7), a cost index was used to estimate the cost from year m to year n . Herein, n was selected as 2020; thus, the cost in the previously built or current period (year m , C_m) was used to calculate C_n . The ratio of cost index value in year m (I_m) and year n (I_n) multiplied by C_m gives C_n (Chemical Engineering Plant Cost Index, CEPICI: 650) [83–86]. Costs were calculated in US dollars at an exchange rate €/€ of 1.126 and A\$/€ 0.770.

$$C_n = (I_n/I_m) * C_m \quad (7)$$

The cost of CO₂ avoidance (CAC) was obtained by comparing the TAC and the CO₂ emission rate to the reference plant with and without CCU/ICCU, shown in Eq. (8).

$$CAC = \frac{TAC_{CCU/ICCU} - TAC_{ref}}{e_{ref} - e_{CCU/ICCU}} \quad (8)$$

2.5. System boundaries

System boundaries define the elements included in economic analysis, which influence the cost of CO₂ avoided and production [87]. The value chain of CCU/ICCU involves the following stages: the capture of CO₂, the conversion of CO₂ into the syngas, the energy to drive the capture and conversion process, and heat recovery during the capture and utilisation process (Fig. 4). In this research, it did not include the transport or distribution of the final product and the H₂ source. In addition, as CO was the main product and the key element of syngas, the final indicator to compare CCU and ICCU was set as CO cost.

2.6. Sensitivity analysis

Sensitivity analysis (SA) is acknowledged as a standardized way to

Table 4
Parameters for economic analysis [59,74,79–81].

Quantity	Unit	Value
Plant lifetime	years	25
Annual operational time	hours	8000
Process utilities and offsite unit costs	25 % of the equipment cost	
Owner's cost and contingency cost	15 % of the installation cost	
Land, permitting, surveying, etc.	5 % of the installation cost	
Total operation & maintenance cost		
Direct labor	Persons	30
Average annual direct labour cost	\$/person	5000
Administrative, support and overhead cost	35 % direct labour cost	
Annual maintenance costs	3.5 % of the installation cost	
Property taxes and insurance	3 % of the install cost	
Natural gas price	\$/KW h	0.045
Catalyst ^a	\$/ton	22,000
Hydrogen	\$ t ⁻¹	1400
CaCO ₃	\$ t ⁻¹	14.83
Carbon tax		None

^a The amount of catalyst is referred to as the reference (each tonne of CO requires 1.06 kg catalyst) [82].

evaluate the influence of a certain parameter on the TEA result [88]. It is essential to conduct a sensitivity analysis as there are both inevitability and uncertainty surrounding the final results [89,90]. According to The DOE/NETL Quality Guidelines for Energy System Studies [91] and the European Best Practice Guidelines for Assessment of CO₂ Capture Technologies [92], sensitivity analysis can be researched based on the variations in the primary data elements from the input data, financial assumptions, and state of technology development, which are diverse timely or spatially in different cases [93,94]. The parameters tested in this paper were the H₂ cost, interest rate and plant life. However, similar to other techno-economic analysis for CCS/CCU, the sensitivity analysis here is relatively superficial, which could facilitate more detailed investigations and research in the future [24,93].

3. Mass and energy balance

3.1. Mass balances

Table 5 presents the mass balance for all four CCU and ICCU processes evaluated in this study. With the same input of flue gas (540 kg s⁻¹) and H₂ (13.85 kg s⁻¹), the CaCO₃ makeup of CCU (4.6 %) was higher than that of ICCU (1 %). The CaCO₃ makeup of CCU was 1.52 Mt year⁻¹, while that of ICCU was 0.62 Mt year⁻¹. As a result, more CO (1.20 Mt year⁻¹) was produced in the ICCU process than in the CCU process (1.02 Mt year⁻¹ of CO produced). In addition, the lower purge ratio of ICCU led to less purge for ICCU (0.21 Mt year⁻¹) than CCU (0.86 Mt year⁻¹) due to the improved stability of sorbents in ICCU compared to CCU.

3.2. Energy balances

In the CCU process (Fig. 2), the energy was produced from the carbonator CC (Q1) and the two heat exchangers (Q2 and Q5), while being consumed by the calcination reactor CAL (Q3) and the RWGS reactor (Q4). As shown in Table 6, the net heat requirement was 742.65 MW in the CCU process, while it was 575.05 MW in the ICCU process. On the other hand, 525.90 MW of heat was output in the CCU process and for the ICCU process, 455.89 MW was produced. Electricity production from CCU and ICCU was 185.65 MWe and 160.93 MWe, respectively. Considering the electricity production and requirement of the power plant, the energy efficiency of ICCU (37.1 %) was higher than that of CCU (35.4 %). ICCU process is isothermal with extremely excellent cyclic and stable performance, which means less energy demand. Besides, the CCU requires the regeneration and transportation of sorbent, which is energy intensive and with heat loss.

Table 5
Summary of mass balance for the CCU and ICCU.

	CCU	ICCU
Input		
Flue gas (kg s ⁻¹)	540.10	540.10
CaCO ₃ makeup (kg s ⁻¹)	52.68	21.50
H ₂ feed (kg s ⁻¹)	13.85	13.85
Output		
CO produced (kg s ⁻¹)	35.52	41.80
Purge produced (kg s ⁻¹)	29.98	7.27
Clean flue gas (kg s ⁻¹)	461.28	461.28
Annual results		
Annual CO production (Mt year ⁻¹)	1.02	1.20
Annual CaCO ₃ consumption (Mt year ⁻¹)	1.52	0.62
Annual purge production (Mt year ⁻¹)	0.86	0.21

Table 6
Summary of energy balance for basic models of CCU and ICCU.

		CCU	ICCU
The heat from the carbonator (Q1)	MW	-174.83	-50.59
The heat from clean flue gas (Q2)	MW	-184.99	-184.99
The heat for the calciner (Q3)	MW	627.19	-
The heat for the CU (Q3)	MW	-	575.05
The heat for the RWGS reactor (Q4)	MW	115.46	- ^a
The heat from the syngas (Q5)	MW	-166.08	-220.31
Net energy input ^b	MW	742.65	575.05
Net energy output ^c	MW	525.90	455.89
$\sum E_{in}$ ^d	MW	2220.98	2053.38
$\sum E_{out}$ ^e	MWe	785.65	760.93
$\eta_{energy\ efficiency} (\sum E_{in} / \sum E_{out})$	-	35.4 %	37.1 %

^a In ICCU, the calciner and RWGS reactor are combined in the CU reactor.

^b Net energy input for CCU is the sum of Q3 and Q4, while net energy input for ICCU is Q3.

^c Net energy output for CCU is the sum of Q1, Q2 and Q5, while net energy output for ICCU is the sum of Q1, Q2 and Q5.

^d $\sum E_{in}$ is the sum of the energy input for the coal-fired plant (1478.33 MW) and the required energy for CCU or ICCU.

^e $\sum E_{out}$ is the sum of the electricity produced from the coal-fired plant (600 MWe) and the electricity from CCU or ICCU.

3.3. Process performance analysis

To find optimum process parameters in CCU and ICCU, the CaO/CO₂ ratio, conversion of CaCO₃, and conversion of RWGS were investigated with regard to CO production and energy efficiency.

As shown in Fig. 4a & b, the increase of CaO/CO₂ ratio leads to an increase in annual CO production and a decrease in energy efficiency. When the CaO/CO₂ ratio increased from 2 to 10, the annual CO production of CCU changed from 0.87 Mt year⁻¹ to 1.15 Mt year⁻¹, and that of ICCU increased from 1.12 Mt year⁻¹ to 1.28 Mt year⁻¹. In terms of energy efficiency, the ICCU process decreased from 37.1 % to 35.9 %, while for the CCU process it declined from 36.9 % to 34.3 %. It is indicated that ICCU produced more CO with higher energy efficiency than CCU when the same CaO/CO₂ ratio was applied. The CaO/CO₂

ratio means less CaCO₃ input. Based on the experiment in the lab, ICCU happens with a longer cycle and better stability of sorbents, so the fewer sorbents input can also operate well [58].

The influence of CaCO₃-to-CaO conversion efficiency on annual CO production and energy efficiency is demonstrated, but its influence was very slight. With CaCO₃-to-CaO conversion changed from 50 % to 90 %, the annual CO production (CCU: 0.98 → 1.02 Mt year⁻¹, ICCU: 1.20 → 1.21 Mt year⁻¹), and the energy efficiency (CCU: 35.59 % → 35.37 %, ICCU: 37.14 % → 36.48 %) remained almost unchanged.

4. Economic evaluation results

Table 7 summarises the key economic performances of CCU and ICCU. The total capital costs (C_{total}) for CCU and ICCU were around 624.65 and 311.26 M\$, respectively. The capital cost of CCU consists of these main contributions: the carbonator for carbon capture, calcination for regeneration of sorbents and CO₂ desorption, RWGS reactor for syngas production, heater exchangers, and the steam turbine, generator and auxiliaries. Compared to CCU, all the reactions in ICCU occurred in one reactor in the lab as reported [57,58], but two reactors were still included and calculated as one more reactor can be used to increase the efficiency of carbon capture up to 100 % [95]. Although the cost of two reactors was considered, the results still implied that the ICCU process was cheaper than CCU (the total capital costs of ICCU were 50 % of that of CCU).

The total O&M cost, including fixed and variable O&M costs, was 969.09 M\$ year⁻¹ for CCU, and 837.91 M\$ year⁻¹ for ICCU (Table 7). H₂ cost dominated the O&M cost, occupying 58 % and 67 % of the total O&M cost for CCU and ICCU, respectively. Similar research has indicated that the H₂ price had the most significant influence on the O&M cost [59]; thus, the influence of other factors can be discussed without the H₂ price influence. In terms of fixed O&M costs, CCU (34.04 M\$ year⁻¹) was twice that of ICCU (17.06 M\$ year⁻¹), as the cost for property taxes, insurance, and annual maintenance of CCU is higher due to their direct relationship to the capital cost. As far as the variable O&M costs are concerned, the cost for natural gas for CCU was higher than that for ICCU, as CCU required more energy due to the separate reaction. Apart from these, both CCU and ICCU shared the same direct labour cost,

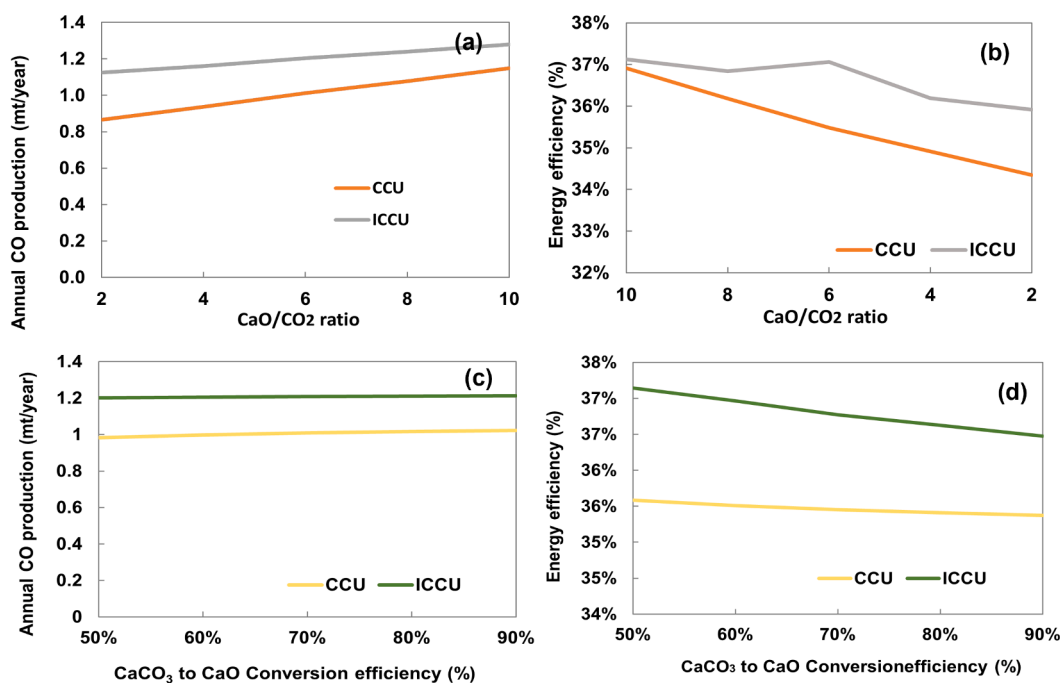


Fig. 4. The makeup/CO₂ ratio effects on annual CO production (a) and energy efficiency (b). The effects of CaCO₃ to CaO efficiency on annual CO production (c) and energy efficiency (d).

Table 7
Economic evaluation summary of CCU and ICCU.

Quantity	CCU	ICCU
Capital cost (M\$)		
Carbonator	75.78	22.48
Calcliner	4.29	–
RWGS reactor	202.97	–
Carbonator and CU	–	64.76
Heat exchanger	86.79	97.04
Steam turbine, generator and auxiliaries	291.14	263.81
Total equipment cost	416.43	207.51
Utilities and offsite units	104.11	51.88
Total install costs (excl. contingency)	520.54	259.38
Land permitting, surveying etc. costs	26.03	12.97
Owner's cost and contingency	78.08	38.91
Total Capital cost	624.65	311.26
Annualized capital cost (M\$/year)	58.52	29.16
Fixed O&M Cost (M\$/year)		
Annual maintenance cost	18.22	9.08
Direct labour cost	0.15	0.15
Property taxes and insurance	15.62	7.78
Administrative, support, & overhead cost	0.053	0.053
Total fixed O&M costs	34.04	17.06
Variable O&M cost (M\$/year)		
Catalyst	23.79	27.98
Hydrogen	558.43	558.43
Natural gas	330.29	225.23
CaCO ₃ make-up	22.55	9.20
Variable O&M cost	933.71	820.84
Total fixed and Variable O&M cost (M\$/year)	969.09	837.91
Total annual cost (M\$/year)	1027.61	867.07
Cost of CO (\$/t CO)	1004.53	720.25

administrative, support, & overhead cost, as well as H₂ cost.

The TAC of CCU was higher than that of ICCU, with costs of 1027.61 and 867.07 M\$ year⁻¹, respectively (Table 7). H₂ cost was the main factor with 54 % and 64 % of the TAC in CCU and ICCU, respectively. For comparison, Steven *et al.* proposed a CCU process with syngas production in three scenarios [33]. When the CO₂ and H₂ feeds were 71.2 kg/s and 3.3 kg/s, respectively, the TAC was 2038.43 M\$ year⁻¹. Therefore, CCU and ICCU can be used to save capital costs without the separation, storage and transportation equipment.

CO cost is another crucial indicator for decision-makers to choose the closely appropriate case for power generation. As shown in Table 7, the CO cost of CCU was 1004.53 \$ ton⁻¹, while that of ICCU was 720.25 \$ ton⁻¹. Compared to the market price of CO of 660 ton⁻¹ and the CO cost of 1394 \$ ton⁻¹ in the reference [60], both CCU and ICCU systems were higher than the market price but lower than the reference cost (Fig. 5). The difference between CO price from CCU and the market price of CO was much higher than for ICCU. The advantage of ICCU in terms of the total cost of CO derives from high CO production and low TAC.

The CO₂ emission intensity between CCU and ICCU is compared in

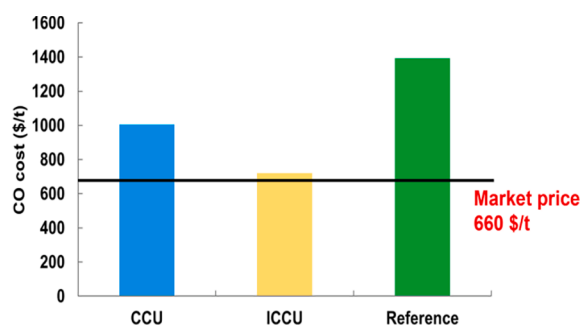


Fig. 5. CO cost comparison between CCU, ICCU and reference with the market price.

order to determine the more environmental-friendly scenario. There are two sources of CO₂ emissions coming from utilities (indirect emission) and released CO₂ after CO₂ capture in the carbonator (direct emission). When the conversion rate of natural gas during combustion is assumed as 100 %, the amount of CO₂ indirect emission is 50.40 kg/s and 39.02 kg/s for CCU and ICCU, respectively (Table 8). Direct emission of CO₂ in both CCU and ICCU are the same as they share the same temperature in the process of carbon capture (13.91 kg/s, Table 8). In total, the CCU process release more CO₂ than ICCU, with 64.31 kg/s and 52.93 kg/s respectively (Table 8). Therefore, the cost of CO₂ avoided by CCU (1230.27 \$/ton) is more than the triple amount of ICCU (317.11 \$/ton), when they perform the same carbon capture capability. These results indicate that ICCU is a better choice in terms of CO₂ emissions. In addition, the cost of CO₂ avoided of the MEA-based capture reached about 301.73 \$/ton, which shows the potential for the application of ICCU (including both capture and utilisation) [96].

5. Sensitivity analysis

Sensitivity analysis clarifies how uncertainties in the input parameters (i.e. raw materials, fuel price, labour, construction, land, etc.) influence the TEA result/s, which can be used to judge the ability of the project to bear risk. [97–99]. However, in this case, the price of H₂ was shown to be the most important factor for the economic results; thus, it was used to perform a sensitivity analysis on the total cost of CO for both CCU and ICCU processes. From Fig. 6a, with the H₂ price increasing from 500 to 3500 \$ ton⁻¹, the total CO cost of CCU increased from 668.62 \$ ton⁻¹ to 1865.26 \$ ton⁻¹, while that of ICCU climbed from 508.07 \$ ton⁻¹ to 1704.71 \$ ton⁻¹. The gap between CCU and ICCU dropped from 24 % to 9 %, with the H₂ price ranging from 500 to 3500 \$ ton⁻¹. Thus, the development of the technology to produce H₂ with less cost will affect the development of the carbon economy in this case. From Fig. 6b, when plant life increased from 5 years to 30 years, the CO cost of CCU decreased from 1100.26 to 1001.56 \$ ton⁻¹ while that of ICCU decreased from 760.79 to 719.00 \$ ton⁻¹. The interest rate and plant life also directly influence CO cost, as shown in Fig. 6c. When the interest rate increased from 0.02 to 0.12, the CO cost of ICCU changed from 709.27 to 729.00 \$ ton⁻¹, while the CO cost of CCU increased from 978.60 \$ ton⁻¹ to 1025.18 \$ ton⁻¹. These minor changes indicate a small influence on interest. Overall, in Fig. 6b and c, CCU is shown to have a higher CO cost than ICCU in all scenarios. CO cost climbed with the increasing interest rate but declined with longer plant life. It is reasonable that increasing interest and short plant life lead to higher CO costs.

6. Conclusion

Process simulation has yielded important outcomes for process flexibility and efficiency improvement. In the ICCU process, more CO (1.20 Mt year⁻¹) can be produced (CCU: 1.02 Mt year⁻¹), while less purge (0.21 Mt year⁻¹) is produced (CCU: 0.86 Mt year⁻¹) with less consumption of CaCO₃ (CCU: 1.52 Mt year⁻¹, ICCU: 0.62 Mt year⁻¹). In terms of energy efficiency, ICCU performs better than CCU, at 37.1 % and 35.4 %, respectively. The better performance of ICCU, compared to CCU, can be ascribed to the integrated design of carbon capture and utilisation in one reactor and correspondingly improved stability of sorbents in ICCU compared to CCU. The CaO/CO₂ ratio and conversion

Table 8
CO₂ emissions and Cost of CO₂ avoided.

	CCU	ICCU
CO ₂ in flue gas (kg/s)	92.74	92.74
Indirect CO ₂ emission (kg/s)	50.40	39.02
Direct CO ₂ emission (kg/s)	13.91	13.91
Total CO ₂ emission(kg/s)	64.31	52.93
Cost of CO ₂ avoided (\$/ton)	1230.27	317.11

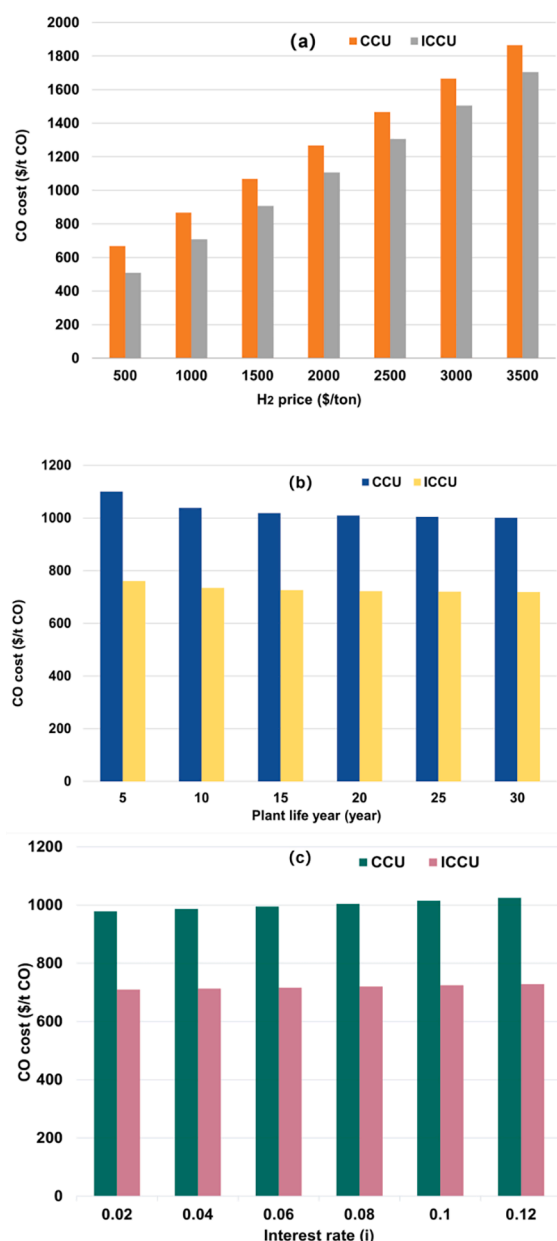


Fig. 6. Sensitivity analysis: (a) H₂ to the total cost of CO; (b) interest rate *i* and (c) plant life to the total cost of CO.

of CaCO₃ into CaO were investigated to evaluate the optimal conditions for high CO production and energy efficiency for the system. In all cases, higher CO production results from larger CaO/CO₂ ratio and higher conversion of CaCO₃ into CaO. In contrast, the larger CaO/CO₂ ratio and higher conversion of CaCO₃ into CaO lead to lower energy efficiency. It is also shown that ICCU always performs better than CCU.

Economic analysis has also been performed according to the indicators such as total annual cost and CO cost. From the results, ICCU performs better than CCU in all cases. The total annual cost and the CO cost of the ICCU process are estimated to be \$867 million and \$720.25 per tonne, respectively. In contrast, CCU requires a higher total annual cost (\$1027.61 million) and cost of CO production (\$1004.53 per tonne). The Cost of CO₂ Avoided by ICCU (317.11 \$/ton) is much lower than that of CCU (1230.27 \$/ton). Therefore, ICCU is confirmed to be a better choice for further industrial applications. H₂ cost was the main contributor to the total annual cost (TAC) of CCU and ICCU, at 54% and 64%, respectively. For further application, H₂ cost remains a challenge

to be addressed.

CRedit authorship contribution statement

Yuanting Qiao: Writing – original draft, Software, Formal analysis, Data curation, Investigation. **Weishan Liu:** Validation. **Ruonan Guo:** Validation. **Shuzhuang Sun:** Investigation. **Shuming Zhang:** Project administration, Resources. **Josh J. Bailey:** Writing – review & editing. **Mengxiang Fang:** Conceptualization, Supervision. **Chunfei Wu:** Methodology, Funding acquisition, Supervision.

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

The data that has been used is confidential.

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