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Process Simulation and Techno-Economic Assessment of Calcium Looping and Membrane Separation Technologies Applied to A Steel Plant in Thailand

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Abstract

A comparison was made of two simulated carbon capture technologies—calcium looping and membrane separation—used in the steelmaking process in Thailand with and without a coke production unit, including a techno-economic assessment. The study was based on a steel plant producing about 1.2 t of liquid steel per hour at >99% purity by weight. The proposed model used common raw materials and operations widely applied in Thailand. The results showed that the steel plant with a coke production unit was more profitable if the coke price exceeded THB 7.93/kg. Additional membrane separation technology with a coke production unit had a higher level of carbon capture purity (92.04% by weight) with the highest profitability (12.83%ROI, a payback period of 7.37 years, and 11.33%DCFRR).

Keywords: Carbon capture; Calcium looping; Membrane separation; Steelmaking process; Techno-economic assessment.
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1. Introduction

According to World Steel Association,[1] the total production of crude steel has increased substantially and continuously reaching about 1,878.5 Mt. in 2022. However, among heavy industries, this industry is ranked the first for carbon dioxide emission. In 2019, the iron and steel industries emitted about 2.6 Gt carbon dioxide annually, mainly from using coal to generate heat and produce coke.[2] To achieve climate goals, carbon dioxide emission should be reduced from 1.4 $t_{CO2}/t_{crude\ steel}$ to 0.6 $t_{CO2}/t_{crude\ steel}$ by 2050. Carbon capture technology plays a vital role in reducing carbon dioxide emissions.[3]

In 2022, crude steel consumption and total imported steel products in Thailand were 7.58 and 15.27 Mt, respectively, lower than the previous year because of the economic slowdown and inflation resulting from the COVID-19 pandemic. In 2023, it is expected that the domestic demand for steel products will likely recover by 8.6 percent, with expected steel consumption reaching 17.8 Mt. Thailand is the 11th largest importer of iron and steel products in the world, with the steel coming mainly from China. However, the Zero-COVID policy of the Chinese government resulted in higher steel selling prices and insufficient supply. Therefore, steel supply was inadequate to meet demand in Thailand.[4] Furthermore, Thailand has no upstream steel industries that can produce pig iron from iron ore as the raw material for midstream and downstream steel industrial processes. Thus, Thailand must import crude steel from abroad.[5]

The Thai government is aware of the greenhouse effect problem and supports achieving the objectives of the United Nations Framework Convention on Climate Change (UNFCCC). In 2007, the Thai government established the Thailand Greenhouse Gas Management Organization (Public Organization) (TGO) to analyze, follow, and promote the reduction of greenhouse gas emissions project. Additionally, TGO manages the marketing of greenhouse gas emissions.[6] Carbon capture technology is essential to control and reduce these emissions.

The overall process of steel production consisted of many units. Initially, iron ore, lime, quartzite, and coke are fed into a blast furnace unit for hot metal production. Then, this product is desulphurized using calcium carbide. Next, the pig iron is transferred to a basic oxygen furnace to produce liquid

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steel through ladle metallurgy and continuous casting to form a slab. After that, the slab is reheated and rolled to produce hot-rolled coil steel.[7] The post-combustion carbon capture technique is suitable for capturing steel plant flue gas. Calcium looping and membrane separation are post-combustion carbon capture techniques that were investigated in the current research study.[8]

Techno-economic assessment includes cost estimation, which consists of total capital investment (TCI) and total production cost (C). Total capital investment (TCI) is separated into two parts: fixed capital investment (FCI) and working capital (WC).[9] C is separated into two parts: cost of manufacturing (COM) and total general expenses (GE). COM includes raw materials and utilities. Four factors can be evaluated as part of the economic profitability analysis. First, return on investment (ROI) is the annual interest rate made by the profits from the original investment. Second, payback period (PB) is the time required for the annual net earnings to equal the original investment. Third, venture profit (VP) is the annual net earnings that exceed the minimum acceptable return on investment(i_{min}). Fourth, annualized cost (AC) is the sum of the production costs and a reasonable return on the original capital investment. Last, discounted cash flow rate of return (DCFRR) is the interest rate that results in a net present value of zero.[10]

2. Material and methods

2.1 Process modeling and simulation

The Aspen Plus V.12 software[11] was used for modeling and simulating the following processes in this research: iron-making in blast furnaces and steelmaking in basic oxygen furnaces with a carbon monoxide converter and carbon dioxide capture based on either calcium looping or membrane separation technologies. Furthermore, this research compared the profitability of different coke sources as fuel in the blast furnace from either outside purchase or from coal production as a part of the simulation.

This research compared six cases of hot-rolled coil steel production:

Case 1: hot rolled coil steel production by producing coke from coal without a carbon capture unit.

Case 2: hot rolled coil steel production using purchased coke without a carbon capture unit.

Case 3: hot rolled coil steel production by producing coke from coal with a calcium looping carbon capture unit.

Case 4: hot rolled coil steel production using purchased coke with a calcium looping carbon capture unit.

Case 5: hot rolled coil steel production by producing coke from coal with a membrane separation carbon capture unit.

Case 6: hot rolled coil steel production using purchased coke with a membrane separation carbon capture unit.

This research aimed to compare cases 1 and 2 as a part of the process simulation results to evaluate the content and purity of the obtained liquid steel and flue gas and as a part of the economic assessment of the profitability between produced and purchased coke. Furthermore, cases 3, 4, 5, and 6 were evaluated to determine the efficiency and purity of the different types of carbon capture technology and impact on profitability of installing carbon capture units.

2.1.1 Modelling and simulation of steel plant with coke oven batteries

Figure 1 presents the flow diagram for a steel plant process with coke oven batteries. Where the coke is produced from coal, initially, the coal is fed into the coke oven batteries to produce coke and coke oven gas for the iron making unit. Alternatively, where purchased coke is used as fuel, no coke oven batteries are required. The raw materials used in this process are iron ore, limestone, and quartzite from mining, coal, or coke depending on the type of fuel used, calcium carbide for desulphurization, and air for combustion and conversion. The main product of the unit was desulphurized liquid iron, with the by-products comprising carbon, lime, magnesium oxide, calcium silicate, calcium sulfide, aluminum oxide, silicon, silica, and ash. The flue gas contained carbon monoxide, carbon dioxide, nitrogen, oxygen, hydrogen, and water vapor.

In cases 1, 3, and 5, a coke production unit was installed. Australian bituminous coal was set as the raw material for coke production in Thailand, with its proximate and ultimate composition and size available elsewhere.[12] Coke oven batteries were represented by the RYield model. The decomposition (pyrolysis) of coal occurred inside the oven.[13] The yield equations were based on Neto *et al*. research.[14] The temperature inside the oven was set at 1,100 °C. The composition of products (coke, coke oven gas (COG), tar, and BTX) were taken from other work.[15] The products were separated. The coke was fed into the RACEWAY reactor. The raw coke oven gas was sent to the treatment unit, then sent to the RACEWAY reactor. Other gases were discharged. From a feed input of 493.9 kg/hr of coal, the coke oven batteries unit produced coke and COG at 353.26 and 80.88 kg/hr, respectively.

Australian hematite iron ore was used as the raw material for the steel production in Thailand, with its composition and size distribution referred to works in Li *et al*.[16] The limestone and quartzite were from India, with their compositions indicated in IEAGHG.[7] Iron ore, limestone, and quartzite at

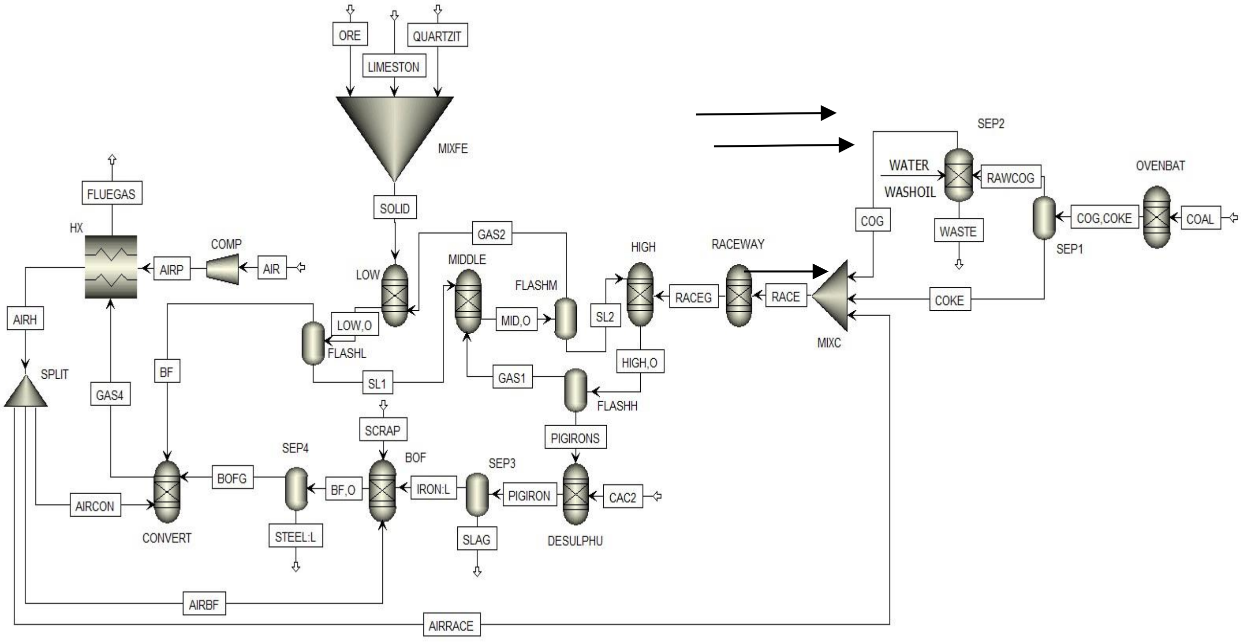

Fig. 1 Process flow diagram of steel plant with coke oven batteries.

1,585.68, 216.65, and 129.52 kg/hr, respectively, were fed into the blast furnace.

The blast furnace (BF) was the main unit in the primary iron production process, feeding solid raw materials into the top section of the furnace, based on a counter-current model from hot gas fed in the RACEWAY section. The simulation considered sub-reactors representing the reaction zone at different temperatures. Coke was fed in at the bottom of the furnace model and combusted in the RACEWAY section; then, hot gas flowed into the top of the furnace. From top to bottom, the model contains four reactors using Gibbs's reactor model to represent low-, middle- and high-temperature, and raceway reactors.[17] For the blast furnace as the RGibbs reactor model at a temperature range of 800–1,600 °C, most ferric oxide (Fe_2O_3) and ferrous oxide (FeO) reacted with carbon monoxide and carbon to produce iron. Consequently, carbon from the coke was combusted and formed carbon monoxide and carbon dioxide. This flue gas from the furnace was sent to the additional reactor to oxidize carbon monoxide and form carbon dioxide. The flue gas was sent to the carbon capture unit.[18] Fuel combustion occurred in a part of the raceway section in the blast furnace at a typical temperature of around 2,000–2,300 °C.[19,20]

In the low temperature (LOW) reactor, the temperature was set at 900°C, where hematite (ferric oxide) and magnetite (ferrous ferric oxide) were converted to wüstite (ferrous oxide) in the presence of carbon monoxide, as shown in chemical equations (1) and (2):

$$3Fe_2O_3 + CO \longrightarrow 2Fe_3O_4 + CO_2 \tag{1}$$

$$Fe_3O_4 + CO \longrightarrow 3FeO + CO_2 \tag{2}$$

In the mid temperature (MIDDLE) reactor, the temperature was set at 1,200 °C. As a result, some wüstite was converted to iron in the solid phase by carbon monoxide, as shown in the chemical equation (3):

$$FeO + CO \longrightarrow Fe + CO_2 \tag{3}$$

In the high temperature (HIGH) reactor, the temperature was set at 1,550 °C. As a result, all the wüstite was converted to iron in the presence of carbon monoxide and coke from the RACEWAY reactor, as shown in chemical equations (3) and (4):

$$FeO + C \longrightarrow Fe + CO \tag{4}$$

Furthermore, the iron melted into liquid iron because the reactor temperature was higher than iron's melting point.

In the RACEWAY reactor, the temperature was set at 2,200 °C. Complete and incomplete combustion occurred using air based on two reactions, as shown in chemical equations (5) and (6):

$$C + O_2 \longrightarrow CO_2 \tag{5}$$

$$2C + O_2 \longrightarrow 2CO \tag{6}$$

In the DESULPHU reactor, the temperature was set at 1,550 °C, with 6 kg/hr of calcium carbide being added to the reactor. The carbon in the calcium carbide captured the sulfur in the stream and converted it into calcium sulfide. Therefore, ferrous sulfide was converted and increased the amount of iron based on chemical equation (7):

$$FeS + CaC_2 \longrightarrow Fe + CaS + C_2 \tag{7}$$

Then, pig iron was sent to the separator (SEP1). Finally, the liquid iron was separated from the slag in the solid phase. The liquid iron was sent to the basic oxygen furnace (BOF) unit which is the main unit of the steel plant that produces products with low concentrations of carbon.[21] The liquid iron increases the iron content by adding scrap and contamination in the liquid iron is decreased by oxygen, with oxide compounds from contamination forming. This unit causes flue gas called basic oxygen flue gas (BOFG) which entered the treatment unit afterward.[22] In the current study, 200 kg/hr steel scraps with the same composition of liquid steel and air were fed to this unit for contamination elimination and to increase the iron content. Inside the BOF, chemical equation (6) occurred. Then, the product stream from the BOF unit was separated. The gas product, BOFG, was mixed with the blast furnace gas stream, while the solid product was sent to the metallurgical ladle unit, the continuous slab caster unit, and the hot rolling mill unit, respectively. Comparison of steel plants with and without coke oven batteries and the main solid and liquid phase stream resulting from the steel plants are presented in Table 1.

The main product, liquid steel, had a high iron content. This composition produced a hot rolled steel coil with a low carbon content. In case 1, the liquid steel had a mass percentage composition of 99.85% Fe, 0.11% C, and 0.04% Si. In case 2, the liquid steel had a mass percentage composition of 99.87% Fe, 0.12% C, and 0.01% Si.

In a part of the gas phase, carbon monoxide was important in reducing iron oxide to iron inside the blast furnace. This gas mainly occurred in the RACEWAY reactor due to the incomplete combustion of coal with oxygen in the AIRRACE stream. However, this process produced carbon dioxide from the complete combustion of coal inside the RACEWAY reactor and as the by-product of iron oxide reduction. Therefore, inside the blast furnace, the amount of carbon monoxide decreased while the quantity of carbon dioxide increased.

Then, the gas phase was sent to the plate heat exchanger (HX) as a hot stream for heating the AIRP stream that was compressed to 5 bars using the centrifugal compressor (COMP). Next, the air stream was separated into three streams. About 1,041.11 kg/hr (Case 1) or 1,046.5 kg/hr (case 2) of air was fed to the RACEWAY reactor as a hot blast. About 290 kg/hr (Case 1) or 465 kg/hr (case 2) of air was fed to the BOF reactor to eliminate carbon in the pig iron.

When the GAS4 stream exited the blast furnace, the stream was mixed with the BOFG stream from the BOF and fed into the CONVERT reactor (RGibbs model). In this reactor, the oxidation of carbon monoxide to carbon dioxide occurred due to the presence of oxygen from the AIRCON stream at 1,500 °C, as shown in chemical equation (8):

$$2CO + O_2 \longrightarrow 2CO_2 \tag{8}$$

The leftover air stream was fed to the CONVERT reactor for oxidation of carbon monoxide. Lastly, flue gas was sent to the carbon dioxide capture unit for cases 3–6. The steel plants with and without coke oven batteries and the main gas phase stream results for the steel plants are compared in Table 2. Case 1 needed a higher air content, especially in the CONVERT reactor, which resulted in a higher amount of flue gas with a lower carbon dioxide content.

Table 1. Main solid and liquid phase stream results (case 1 vs. case 2) for steel plant simulation.

	Case 1			Case 2		
Stream	PIGIRON	IRON:L	STEEL:L	PIGIRON	IRON:L	STEEL:L
Temperature (°C)	1,550	1,550	1,600	1,550	1,550	1,600
Pressure (bar)	5	5	5	5	5	5
	Mass Flows (kg/hr)					
Fe:L	1,000.85	1,000.85	1200.54	1,000.85	1,000.85	1,200.59
Fe:S	0	0	0	0	0	0
C	58.87	58.87	1.32	85.06	82.52	1.45
SiO_2	98.41	0	0	100.7	0	0
MgO	4.43	0	0	4.43	0	0
$CaSiO_3$	250.67	0	0	247.71	0	0
CaS	4.61	0	0	6.45	0	0
Al_2O_3	55.84	0	0	55.84	0	0
Si	0.42	0.42	0.5	0.06	0.06	0.07
Ash	40.17	0	0	56.17	0	0
Total Mass Flows	1,514.27	1,060.14	1202.36	1,557.26	1,083.42	1,202.11

Table 2. Main gas phase stream results from the steel plant (case 1 vs. case 2).

	Case 1				Case2			
Stream	AIR	BF	BOFG	FLUEGAS	AIR	BF	BOFG	FLUEGAS
Temperature (˚C)	25	900	1,600	783.3	25	900	1,600	703.15
Pressure (bar)	1	5	5	4.93	1	5	5	4.93
	Mass Flows (kg/hr)							
CO	0	337.26	131.33	1.81	0	357.7	189.61	1.58
CO_2	0	535.11	0	1,274.08	0	774.05	0	1,640.83
H_2O	0	356.82	0	637.42	0	113.83	0	179.44
H_2	0	31.42	0	0.04	0	7.35	0	0.01
O_2	866.57	0	0	4.68	733.69	0	0	8.9
N_2	2,483.43	771.03	214.98	2,489.26	2,416.31	792.54	356.69	2,419.37
CH_4	0	0.08	0	0	0	0	0	0
Total Mass Flows	3,350	2,031.73	346.32	4,407.29	3,150	2,045.48	546.31	4,250.13

2.1.2 Modeling and simulation of carbon capture based on calcium looping methodology

Carbonation is the reaction of metal oxide and carbon dioxide. This technique separates carbon dioxide from flue gas to form metal carbonate. In the current study, the metal carbonate was calcined to capture carbon dioxide while metal oxide was regenerated in a looped system. Calcium was the primary metal for this technique.[23] The calcium loop model concept had two central units, namely the two reactors that represent the reaction inside the model. Flue gas from the furnace was sent to the carbonator reactor. Lime (CaO) fed from the regenerator reactor combined with CO_2 in the flue gas to produce limestone ($CaCO_3$), which was combusted in the regenerator reactor, resulting in its decomposition into CO_2 and lime. CO_2 that was sent to the storage unit. Furthermore, the lime was recycled back to the carbonator reactor as raw material to combine with CO_2 from the flue gas.[24]

The flue gas was fed into the reactor (CARBO) type RCSTR model at 25 kg/hr lime from an external source and recycled inside the calcium looping unit. This reactor was set at 650 ˚C (Fig. 2). The carbonation reaction occurred, as shown in the chemical equation (9):

$$CaO + CO_2 \longrightarrow CaCO_3 \qquad (9)$$

Carbon dioxide from flue gas mainly reacted with the lime to form limestone, based on the particle size distribution of lime[25] and coarse limestone.[26] Therefore, the flue gas was

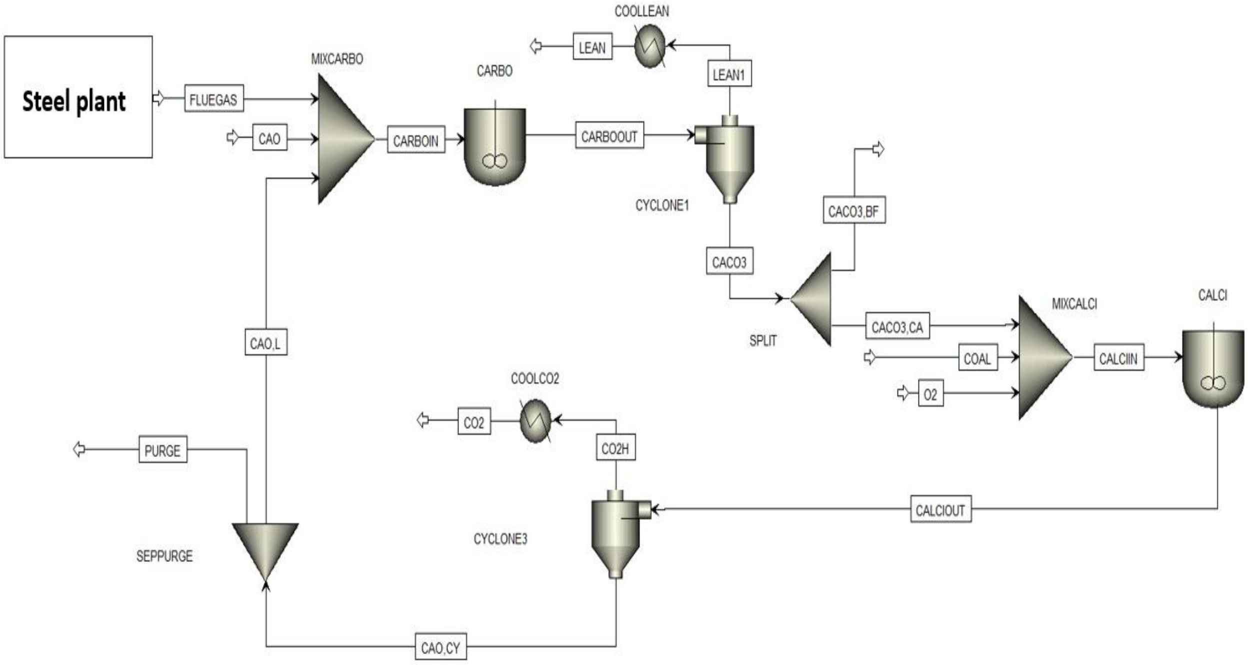

Fig. 2 Process flow diagram of carbon capture based on calcium loop methodology.

lean in carbon dioxide. The carbonation reaction was a first-order reaction with an activation energy of 28.3 kJ/mol and a pre-exponential factor of 2.22 × 10^{-4} mol/m^2-s.[27]

The product from the carbonator reactor was fed to the cyclone, wherein the solid and gas phase products were separated. The gas phase product (lean carbon dioxide) was cooled using a heat exchanger and discharged from the carbon capture unit. On the other hand, the solid phase product was separated into two streams. The CACO3, BF stream was fed to the blast furnace unit for use as raw material, while leftovers were fed to the reactor (CALCINE) type RCSTR model. This reactor was set at 900°C. Limestone decomposed to lime and carbon dioxide, as shown in the chemical equation (10):

$$CaCO_3 \longrightarrow CaO + CO_2 \qquad (10)$$

Additionally, coal and oxygen were fed in for combustion. The calcination reaction was a first-order reaction with an activation energy of 203.8 kJ/mol and a pre-exponential factor 1.1 × 10^8/s.[28]

The calciner reactor products were fed into the cyclone to separate the solid and gas phases. Carbon dioxide was discharged from the carbon capture unit, while the solid phase product (lime) was purged from looping by 40% because of the presence of ashes and contaminants in the stream and the decreasing amount of accumulating lime in the calcium looping. Furthermore, this purge stream could be used in steel plants as raw material. The remaining lime was fed to the carbonator reactor to separate carbon dioxide from flue gas in a looped system. The details of the mainstream results of calcium looping methodology in cases 3 and 4 are compered in Table 3. With the same conditions and lime feeding, the carbon dioxide of flue gas in case 3 was higher than in case 4, causing a lower carbon capture rate.

2.1.2 Modeling and simulation of carbon capture based on membrane separation methodology

For membrane separation, the membrane must have high CO_2 permeance, with as low a permeance as possible for other gases. A polymeric membrane is the conventional material for membrane CO_2 separation in post-combustion capture.[29] The transport of gases through polymer membranes occurs due to diffusion from the side with higher pressure to the side with lower pressure. The rate of mass transfer depends on the partial pressure gradient. It is customary to use the permeance Q of the membrane. The mass flux of the permeated gas is calculated from Fick's law equation.[30]

In the current study, the flue gas from the steel plant was fed to the membrane separation unit (Fig. 3). This gas was compressed and cooled using a propeller fan (COMP1) and a finned double pipe heat exchanger (COOL1) to 5 bar and 50°C, which are appropriate values for the membrane. Then, the flue gas was fed to a polyvinyl amine membrane type spiral-wound with a co-current model (MEMB). The membrane properties can be referenced elsewhere.[31,32] An essential characteristic for stream evaluation is the specific gas species permeance unit. The CO_2, CO, O_2, H_2, N_2, and H_2O permeances were 1, 1, 0.02, 0.006, 0.005, 0.02, and 2.25 × $10^{-9}\ \frac{m^3}{bar\cdot hr\cdot m^2}$ at STP, respectively. MEMB was represented by the user2 model, which transferred data to the Microsoft Excel software for calculation. The membrane area was

Table 3. Mainstream results of calcium looping methodology (case 3 vs. case 4).

	Case 3					Case 4				
Stream	FLUEGAS	CAO,L	LEAN	CACO3	CO2	FLUEGAS	CAO,L	LEAN	CACO3	CO2
Temperature (°C)	750.5	900	50	650	50	703.15	900	50	650	50
Pressure (bar)	4.91	1	1	1	1	4.93	1	1	1	1
	Mass Flows (kg/hr)									
$CaCO_3$	0	0.04	26.86	2,640.24	0	0	0.05	34.3	3,363.76	0
CaO	0	1,773.92	16.93	1,664.61	29.88	0	1,789.28	12.99	1,274.37	30.15
CO	1.81	0	1.81	0	0	1.58	0	1.58	0	0
CO_2	1,274.08	0	101.34	0	1,331.11	1,640.83	0	146.68	0	1,697.14
N_2	2,489.26	0	2,489.26	0	0.92	2,419.37	0	2,419.37	0	1.15
O_2	4.68	0	4.68	0	45.26	8.9	0	8.9	0	56.8
H_2O	637.42	0	637.42	0	3.75	179.44	0	179.44	0	4.71
H_2	0.04	0	0.04	0	3.75	0.01	0	0.01	0	4.71
S	0	0.25	0	0.25	0	0	0.32	0	0.32	0.01
C	0	0.18	0	0.18	0	0	0.22	0	0.21	0
Ash	0	21.22	0.17	21.05	0.34	0	26.79	0.21	26.58	0.42
Total Mass Flows	4,407.29	1,795.62	3,278.51	4,326.33	1,415.02	4,250.13	1,816.66	2,803.49	4,665.24	1,795.09

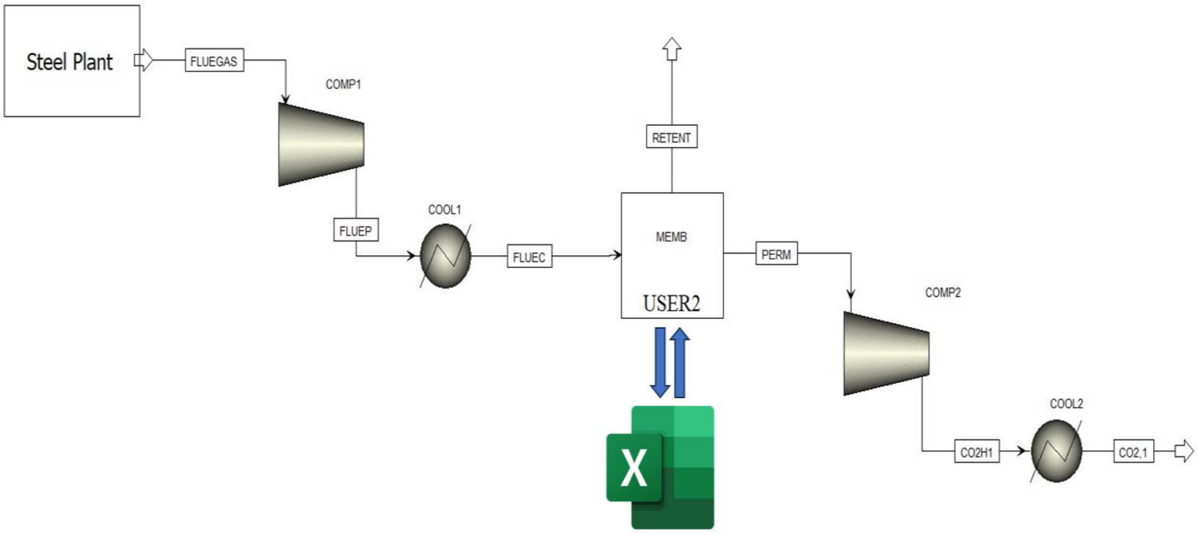

Fig. 3 Process flow diagram of carbon capture based on membrane separation methodology.

calculated using the Fick's law equation [the mass flux of the permeated stream (J) = Q(P_2 – P_1)],[30] where the membrane properties are specified by the permeance of gas (Q) and the differential partial pressure (P_2 - P_1) is calculated using the difference between the feeding partial pressure and the permeated partial pressure of carbon dioxide. By specification, the mass flowrate of carbon capture was the same as for calcium looping for comparison; thus, the area of the membrane could be calculated based on the mass flowrate divided by the mass flux. The areas of the membrane in cases 5 and 6 were 853.69 and 661.24 m^2, respectively, with the pressure ratio equal to 100. Then, permeated gas (0.05 bar pressure) was pressurized and cooled using the blower (COMP2) and the finned double pipe heat exchanger (COOL2) to 1 bar and 50°C. Next, the permeated gas was sent to the storage unit for carbon dioxide. The mainstream results based on the membrane separation methodology in cases 5 and 6 are compared in Table 4.

2.2 Techno-economic assessment

The Aspen Process Economic Analysis V.12 software[33] was used for equipment cost estimation along with techniques from the reference textbook for blast furnaces,[10] for convertors, carbonators, and calciners.[34] For comparison, membrane cost estimation data were sourced from standard information.[32]

The current research studied hot-rolled coil steel production in Thailand based on a lifetime of 20 years. Therefore, cost estimation was based on the Thai unit of currency, which is the baht (THB). The raw materials (iron ore, coal, and coke) used Australian sources, while the other raw materials used Indian sources. Aspen Economic Analyzer evaluated the amounts and types of operational utilities. The prices of utilities were specified according to public utility prices in Thailand. The electricity prices referred to the Provincial Electricity Authority in Thailand, based on type3 (industry, using electricity less than 250,000 (kW/h)/month.)

Table 4. The mainstream results of membrane separation methodology (case 5 vs. case 6).

	Case 5			Case 6		
Stream	FLUEGAS	RETENT	CO_2	FLUEGAS	RETENT	CO_2
Temperature (˚C)	783.3	50	50	703.14	50	50
Pressure (bar)	4.93	4.75	1	4.93	4.75	1
	Mass Flows (kg/hr)					
CO_2	1,274.08	101.34	1,172.74	1,640.83	146.68	1,494.15
N_2	2,489.26	2,487.71	1.55	2,419.37	2,418.04	1.33
O_2	4.68	4.56	0.12	8.9	8.7	0.2
CO	1.81	0.15	1.66	1.58	0	1.43
H_2O	637.42	637.42	0	179.44	179.44	0
H_2	0.04	0.04	0	0.01	0.01	0
Total Mass Flows	4,407.29	3,231.22	1,176.06	4,250.13	2,753.02	1,497.11

with a 22–33 kV voltage.[35] The water prices were taken from the Provincial Waterworks Authority in Thailand, based on type 3 (industry).[36] The product prices for hot-rolled coil steel with a low carbon content were based on the selling price in Thailand. The carbon credit prices in Thailand were referenced to information from the Thailand Greenhouse Gas Management Organization.[37] The raw material, utility, and product prices used are shown in Table 5.

Table 5. Raw material, utility, and product prices.

Item	Price per unit (THB/kg)	Reference
Coal	4.71	38
Coke	8.11	39
Iron ore	3.66	40
Limestone	0.73	41
Quartzite	0.34	42
Calcium carbide	23.63	43
Lime	0.42	44
Scrap	6.00	45
Electricity	Depending on amount	35
Water	Depending on amount	36
Hot rolled coil steel	25.80	46
Carbon credit	0.11	37

Total capital investment was evaluated based on the percentage of delivered-equipment cost, a preliminary estimate technique. This technique estimated the total direct cost, total indirect cost, and working capital as a percentage of the purchased equipment cost depending on the phase type of the processing plant. Steel plants are specified as solid-fluid processing plants. Based on this assumption, the total direct cost, total indirect cost, and working capital were 302, 126, and 75% of purchased equipment costs, respectively.[9]

As a part of the total production cost, the raw material cost differed depending on the case. However, in cases 3 and 4, which used calcium looping as the carbon capture technology, limestone was used as an intermediate product. Therefore, limestone from the carbon capture unit to the iron-making unit was used as raw material in these two cases. Furthermore, the scrap used in the basic oxygen furnace could be received from other units in the steel plant, such as the continuous slab caster unit and the hot rolling mill unit. About 88.56 kg/hr of internal scrap was recycled back to the steel plant and purchased from external sources.

The utilities required in a steel plant with a carbon capture unit are electricity and water. The electricity and water consumption details for the six study cases are presented in Fig. 4.

In the operations (labor-related), direct wages and benefits were specified at THB 354.00/operator-h based on the steel plant at Rayong in Thailand, according to the Ministry of Labour.[47] The number of operators was assumed to be 3 persons per process. Regarding land and building taxes, the steel plant was at Rayong in Thailand. The plant area was 0.083 km^2 with a land appraisal price of THB 1,125 M/km^2. The land and building taxes were calculated from the summation of the land value and total direct cost with the land and building tax rate based on the Fiscal Policy Office in Thailand.[48] The total production cost was estimated based on the cost sheet outline[10] adapted with assumptions for Thailand costing.

The main product of the steel plant in the simulation results was hot-rolled coil steel. The carbon-credit sale value depended on the carbon dioxide capture rate, as shown in the simulation results. Tax was evaluated according to the Revenue Department in Thailand using a juristic tax rate of

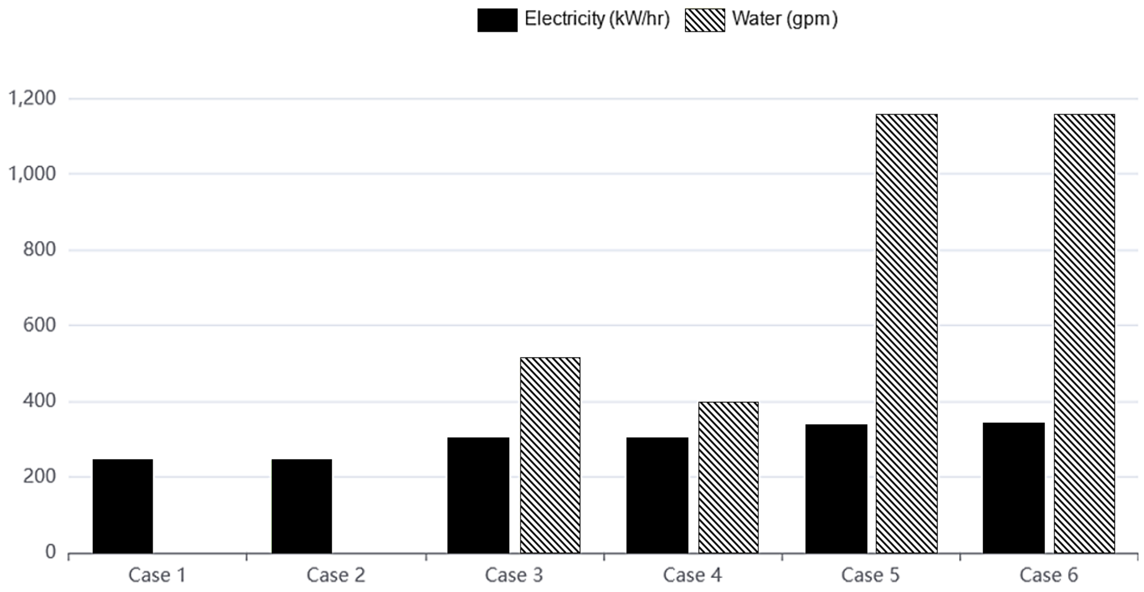

Fig. 4 The electricity and water consumption of six cases.

20% of profit.[49] The steel plant reinvestment period was 20 years.[2] The total capital investment, raw material cost, utility cost, total production cost, sales, tax, return on investment, payback period, venture profit, annualized cost, and the discounted cash flow rate of return for the 6 cases are provided in Table 6. These results showed that the payback period for all cases was lower than 20 years, indicating that in all cases full repayment would occur before reinvestment was required.

3. Results and discussion

3.1 Process modeling and simulation

The stream results of the steel plant indicated that the iron content and carbon dioxide concentration increased along with the duration in the furnace. An increase in temperature promoted the reduction reaction for the iron oxide compound using carbon and carbon monoxide as reactants, resulting in a higher amount of carbon dioxide in the gas phase product. In the steelmaking section, increasing the iron content, which produced better quality steel, resulted in more flue gas from the oxidation of carbon inside the liquid iron. Furthermore, coke production from the coke oven batteries produced more flue gas in coke oven gas form. Therefore, these flue gases contained carbon dioxide, a pollutant that needed treatment or processing in the separation unit. The volume of mixed flue gas, containing COG, BF, and BOFG, in the case with coke oven batteries (4,407.29 kg/hr) was higher than without coke oven batteries (4,250.13 kg/hr) because of the presence of the coke oven gas with a greater air stream needed. The flue gas in cases 1 and 2 mainly consisted of nitrogen at 2,489.26 and 2,419.37 kg/hr, respectively, at 56.48 and 56.92% wt. of flue gas, respectively. In addition, for these cases, the carbon dioxide flow rates in the flue gas stream were 1,274.08 and 1640.83 kg/hr, respectively, at 28.91 and 38.61% wt. of flue gas, respectively.

Pig iron from the DESULPHU reactor, at 1,550°C, was separated into a hot metal liquid phase (IRON:L) and a solid phase (SLAG) by tapping. The liquid iron stream had a production capacity of 1,000.85 kg/hr with 94.41% wt. and 1,000.85 kg/hr with 92.38% wt. because the case without coke oven batteries had much more carbon in solid form and was contaminated with liquid iron. This caused the steelmaking section to purify liquid iron. The liquid steel had a production capacity of 1,200.54 kg/hr with 99.85% wt. and 1,200.59 kg/hr with 99.87% wt. Therefore, there was no substantial difference between steel plants with or without coke oven batteries regarding the productivity, purity, and quality of liquid steel products. However, there were effects on the cost and carbon capture unit.

For model validation, a comparative study involving coke oven batteries and a blast furnace with coke oven batteries in the solid and gas phases was based on information report elsewhere.[7,17] The results of the model validation are presented in Table 7.

Table 6. Economics evaluation for 6 cases.

	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6
TCI (THB million)	237.154	212.273	293.851	271.495	252.865	226.704
Raw mat. (THB million/yr)	75.726	89.169	81.883	96.238	75.726	89.169
Utilities (THB million/yr)	6.313	6.311	7.905	7.883	8.908	9.038
TPC (THB million/yr)	180.817	186.501	202.708	211.639	188.908	195.919
Sales (THB million/yr)	228.456	228.408	229.609	229.878	229.472	229.702
Tax (THB million/yr)	9.528	8.382	5.38	3.648	8.113	6.756
ROI (%)	16.07	15.79	7.32	5.37	12.83	11.92
PB (yr)	5.95	6.02	12.39	16.18	7.37	7.86
VP (THB million/yr)	33.368	29.281	15.644	9.161	27.393	22.492
AC (THB million/yr)	185.56	190.746	208.585	217.069	193.966	200.454
DCFRR (%)	15.11	14.79	3.95	0.7	11.33	10.22

Table 7. Results of model validation of coke oven batteries (kg/t $_{Coke}$) and blast furnace with coke oven batteries in solid phase (kg/t $_{Liquid\ iron}$) and gas phase (kg/t $_{BF}$).

Coke Oven Batteries	Coal	Coke	COG
IEAGHG 2013 [7]	1,396.9	1,000	195.4
Case 1	1,398.12	1,000	228.95
%Error	0.07	0	17.17

Blast Furnace (Solid)	Fe	C	Si	Mn	SiO_2	$CaSiO_3$	Al_2O_3	CaO	CaS	MgO	MnO
Suzuki 2015 [17]	949.29	45.04	5.3	2.5	93.31	0	37.84	126.74	0	18.72	1.38
Case 1	944.07	55.53	0.4	0	92.83	236.45	52.67	0	4.35	4.18	0
%Error	0.55	23.26	92.45	100	5	100	46.79	100	100	71.45	100

Blast Furnace (Gas)	CO	CO_2	H_2O	H_2	N_2	CH_4
Suzuki 2015 [17]	195.83	320.07	14.87	2.79	465.98	0.48
Case 1	166	263.38	175.62	15.46	379.49	0
%Error	15.23	17.71	100	100	18.56	100

The validation results showed different concentrations between the coke oven batteries and the blast furnace solid and gas streams because of the different compositions of iron ore, coal, and other raw materials. Additionally, the effect of using a reactor in the RYield model with the coke oven batteries, which used the yield equation from other research[24] and the RGibbs model in blast furnace, depended on the Gibbs's free energy potential.

Calcium looping in the case 3 simulation results indicated that the carbon dioxide captured flow rate that formed into limestone was 1,145.74 kg/hr, with a carbon capture rate of 92.04% wt. Some carbon dioxide did not react with the lime to form limestone. Some of the limestone was sent to the iron-making unit as raw material, while the leftover limestone was calcined in the calciner using coal as the fuel and complete combustion occurred. This produced more carbon dioxide in the outlet stream. The carbon dioxide flow rate and purity were 1,331.11 and 94.07% wt., respectively. Furthermore, the carbon dioxide purity depended on cyclone performance, which could be adjusted in the design to improve the solid-gas separation performance. On the other hand, in case 4, the carbon dioxide captured flow rate was 1,494.15 kg/hr, with a carbon capture rate of 91.06% wt. Therefore, for the same conditions and lime feeding, the captured efficiency slightly decreased when the concentration of carbon dioxide increased. The carbon dioxide flow rate and purity were 1,697.14 and 94.54% wt., respectively, indicating that the carbon dioxide purity for case 4 was higher than for case 3 because of the coal that was needed more for the combustion of limestone in case 4, which was more than in case 3. In conclusion, using the calcium looping methodology based on the carbon capture technique (case 3) produced less carbon dioxide and a greater carbon dioxide captured rate. However, case 4 caused had a greater level of carbon dioxide purity.

The simulation results from membrane separation as applied in case 5 indicated that the CO_2 captured flow rate was 1,172.74 kg/hr, with a carbon capture rate of 92.04% wt. Some carbon dioxide could not permeate the membrane because of inadequate membrane area or pressure. Therefore, the remaining carbon dioxide was discharged with the nitrogen from the carbon captured unit in the RETENT stream. The carbon dioxide purity was 99.72% wt., with a small amount of contaminated gases, since most of the nitrogen and water from the flue gas stream was retained. Consequently, the permeated stream mostly contained carbon dioxide of high purity. On the other hand, in case 6, the carbon dioxide captured flow rate was 1,494.15 kg/hr, with a carbon capture rate of 99.8% wt. However, the carbon dioxide captured rate and purity could be adjusted by changing the membrane area and pressure of the stream. This research specified the same carbon capture rate for cases 3 and 5 and for cases 4 and 6, based on the same iron and steelmaking process. A comparison of the results between the different cases of carbon capture is presented in Table 8.

In cases 3 and 5, the same quantity of carbon dioxide (1,274.08 kg/hr) was sent to two different carbon capture units. Both methodologies set the pure carbon dioxide flow rate at a lean gas level of 101.34 kg/hr and the carbon dioxide captured rate at 92.04% wt. The results indicated that the membrane separation methodology had a lower total flow rate at the output stream than the calcium looping methodology (1,172.74 vs. 1,331.11 kg/hr). In addition, the membrane separation methodology had a higher carbon dioxide percentage than the calcium looping methodology (99.72 vs. 94.07% wt.).

In cases 4 and 6, the same quantity of carbon dioxide (1,640.83 kg/hr) was sent to two different carbon capture units. Both methodologies set the pure carbon dioxide flow rate at a lean gas level of 146.68 kg/hr and the carbon dioxide captured rate at 91.06% wt. The results indicated that the membrane separation methodology had a lower total flow rate at the output stream than the calcium looping methodology (1,494.15 vs. 1,697.14 kg/hr). In addition, the membrane separation methodology had a higher carbon dioxide percentage than the calcium looping methodology (99.8 vs. 94.54% wt.).

The comparison indicated that two techniques in 4 cases could capture carbon dioxide with high efficiency (>90% wt.), with membrane separation being slightly more efficient. Thus, economic profitability would be more crucial in deciding which carbon dioxide capture technique was more worthwhile.

3.2 Techno-economic assessment

The comparison between cases 1 and 2 indicated the effect of installing a coke oven battery unit for coke production. In case 1, the coal was decomposed and fed as coke and coke oven gas to the iron-making unit, while case 2 used purchased coke, with the same amount of coal being used as in case 1, for the iron-making unit instead. Thus, in case 1 the total capital investment was higher than in case 2 because of the coke oven batteries. Case 2 used purchased coke instead of purchased coal, which increased the raw material costs because the price for coke was higher than for coal. This affected the total production cost of case 2, which was higher than that of case 1. With slightly different sales, Case 1 had a higher annual profit after tax than Case 2. Considering profitability, the results showed that overall, case 1 (with coke oven batteries) was more worthwhile than case 2 (without coke oven batteries) because case 1 had a higher ROI, lower PB, higher VP, lower AC, and a lower AC than case 2. However, if the coke price fell below THB 7.93/kg, purchased coke would be more expensive than the coke produced from the installed coke oven batteries.

The comparison between cases 3 and 4 involved the same process of iron and steel making as for cases 1 and 2, respectively, using calcium looping carbon captured methodology. However, in case 4, the amount of flue gas was higher than in case 3, which required higher purchased equipment costs for the calcium looping unit. In addition, the inclusion of the coke oven batteries required even more investment in total capital costs. Furthermore, case 4 had higher raw material and total production costs due to the higher amount of flue gas than case 3. With slightly different sales, Case 3 had a higher annual profit after tax than Case 4. Considering profitability, the results showed that overall, case 3 was more worthwhile than case 4 because case 3 had a higher ROI, lower PB, higher VP, lower AC, and higher DCFRR than case 4.

The comparison between cases 5 and 6 involved the same process of iron and steel making as for cases 1 and 2, respectively, using membrane separation carbon captured

Table 8. Comparison of results between different cases of carbon capture.

Stream/Parameter	Carbon Dioxide Capture Methodology			
	Calcium Looping		Membrane Separation	
	Case 3	Case 4	Case 5	Case 6
Input Stream				
Pure CO_2 flow rate at input stream (kg/hr)	1,274.08	1,640.83	1,274.08	1,640.83
Output Stream				
Pure CO_2 flow rate at lean gas (kg/hr)	101.34	146.68	101.34	146.68
Total flow rate at output stream (kg/hr)	1,415.02	1,795.09	1,176.06	1,497.11
Pure CO_2 flow rate at output stream (kg/hr)	1,331.11	1,697.14	1,172.74	1,494.15
Parameter				
CO_2 capture rate (% wt.)	92.04	91.06	92.04	91.06
CO_2 capture purity (% wt.)	94.07	94.54	99.72	99.8

methodology. However, in case 6, the amount of flue gas was higher than in case 5, due to the higher purchased equipment costs for the calcium looping unit. In addition, the coke oven batteries required more investment in total capital costs. Furthermore, case 6 had a higher total production cost than case 3. With slightly different sales, Case 5 had a higher annual profit after tax than Case 6. Considering profitability, the results showed that overall, case 5 was more worthwhile than case 6 because case 5 had a higher ROI, lower PB, higher VP, lower AC, and higher DCFRR than case 6.

Installing the carbon capture unit greatly increased TCI, especially with calcium looping (>THB 55 M), while the membrane process only increased the TCI by about THB 15 M. Considering TPC, the membrane unit increase was a relatively small amount (about THB 9 M/yr), whereas the calcium looping unit represented a much greater increase (THB >22 M/yr), because the membrane unit only added to the utility cost with a constant raw material cost. Therefore, TPC was mainly affected by the calcium looping cost, while TCI was affected by the membrane cost.

The comparison between the calcium looping and membrane separation carbon capture technologies indicated that membrane separation was considerable better than calcium looping. This indicated that the recycling of limestone in the calcium looping unit to the iron and steelmaking unit was not sufficient to increase the project's profitability. Additionally, calcium looping had higher total capital and total production costs compared to membrane separation.

Thus, case 1, (iron and steelmaking using coke oven batteries) was the most worthwhile. Considering carbon capture technology, case 5, (membrane separation) was more worthwhile as it the membrane separation applied to iron and steelmaking with coke oven batteries decreased the ROI, VP, and DCFRR by about 3.24%, THB 5.975 M/yr, and 3.78%, respectively, and increased the PB and AC by about 1.42 yr and THB 8.406 M/yr, respectively.

4. Conclusions

This study investigated, the simulation of iron and steelmaking with and without coke production and carbon dioxide capture based on calcium looping and membrane separation technology regarding production and CO_2 capture feasibility. The different processes were analyzed using the AspenPlus One V.12 industrial simulation software.

In the steel plant, liquid steel and iron purity production capacity with and without coke production had similar values (about 1,200 kg/hr and 99.8% wt., respectively). However, the amount of carbon dioxide in the flue gas stream without coke production was higher than in the case with coke production by about 360 kg/hr. At the same carbon capture rate of about 91–92%, the membrane separation methodology had about 5 % higher carbon captured purity than from calcium looping.

In terms of economic profitability, the steel production with the coke production unit was more worthwhile than the purchased coke case. The coke production case had a higher %ROI and %DCFRR than from purchasing coke, by about 0.3%. With an installing carbon capture unit, membrane separation technology was considerably more worthwhile than the calcium looping technology. Steel plants with membrane separation technology had a higher TCI of about THB 15.7 M and lower values for %ROI and %DCFRR of 3.24 and 3.78%, respectively, with a longer payback period of 1.42 years.

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Conflict of Interest

There is no conflict of interest.

Supporting Information

Applicable. The supplementary information, containing the techno-economic analysis, is available at the shared drive: https://drive.google.com/drive/folders/1683DqYcYkcV_l3Hay3syZDRmSebyGL4R?usp=sharing.

Abbreviations

AC	Annual cost, THB/yr
C	Total production cost, THB/yr
COM	Cost of manufacturing, THB/yr
DCFRR	Discounted cash flow rate of return, %
FCI	Fixed capital investment, THB
GE	Total general expenses, THB/yr
i_{min}	Minimum acceptable return on investment, %
J	Mass flux, $\frac{kg}{m^2 \cdot hr}$
P	Partial pressure, bar
PB	Paback period, yr
Q	Permeance, $\frac{m^3}{m^2 \cdot bar \cdot hr}$
ROI	Return on investment, %
TCI	Total capital investment, THB
VP	Venture profit, THB/yr
WC	Working capital, THB

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