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Techno-Economic Analysis of Hydrogen and Electricity Production by Biomass Calcium Looping Gasification

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Abstract: Combined cycle, biomass calcium looping gasification is proposed for a hydrogen and electricity production (CLGCC–H) system. The process simulation Aspen Plus is used to conduct techno-economic analysis of the CLGCC–H system. The appropriate detailed models are set up for the proposed system. Furthermore, a dual fluidized bed is optimized for hydrogen production at 700 °C and 12 bar. For comparison, calcium looping gasification with the combined cycle for electricity (CLGCC) is selected with the same parameters. The system exergy and energy efficiency of CLGCC–H reached as high as 60.79% and 64.75%, while the CLGCC system had 51.22% and 54.19%. The IRR and payback period of the CLGCC–H system, based on economic data, are calculated as 17.43% and 7.35 years, respectively. However, the CLGCC system has an IRR of 11.45% and a payback period of 9.99 years, respectively. The results show that the calcium looping gasification-based hydrogen and electricity coproduction system has a promising market prospect in the near future.

Keywords: calcium looping gasification; CO₂ capture; biomass; hydrogen; combined cycle; techno-economic analysis

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1. Introduction

Currently, the production of hydrogen from fossil fuels has experienced increased research attention because of the ecofriendly nature of hydrogen utilization. Hydrogen is widely used in different areas such as ammonia production, oil refining, and production of chemicals, but limited hydrogen utilization constrains its further use. The expected availability of hydrogen can replace gasoline fuel in transportation and thermal power generation, resulting in less emissions [1,2]. Globally, hydrogen production is 70 Mt/year; 23% is contributed by coal gasification and 76% by natural gas reforming through the steam methane reforming process [3]. In contrast, the remaining production comes from different methods such as electrolysis, solar, and biological processes. The amount of coal consumed for hydrogen production is 107 Mt (2% of global coal use), and natural gas accounts for 205 Gm³ (6% of global natural gas), generating 830 Mt CO₂ emissions, which are 2% of total global emissions [3]. Hence, an increase in hydrogen demand would lead to additional CO₂ emissions; however, without carbon capture and storage (CCS), the existing technology infrastructure cannot meet the 2 °C Paris Agreement goal for climate change [1,2,4].

Biomass gasification can be an alternate technology for hydrogen production, considering hydrogen's benefits in saving the environment and its economic feasibility [5]. The hydrogen produced from biomass gasification contributes a minor part of CO₂ emission, since biomass is a renewable resource that consumes CO₂ during its growth and releases CO₂ from the gasification process [6]. Many countries realized that the “H₂ economy” will be a potential solution to meet global energy demand and reduce fossil fuel use [7].

Market forecast shows that demand for hydrogen will increase in the near future with 5–10% growth per year, from 50 to 82 Mt by 2050. Globally, transportation (especially air and road) accounts for about a quarter of the net of carbon dioxide emissions [8–10]. Industrial activities by 2050 will increase the demand for H₂, resulting in H₂ production projected to rise, especially for steel and ammonia production, by about 2 EJ/a for each activity, while hydrogen-powered vehicles could produce approximately 22 EJ/a [11]. However, renewable resources contribute only 2% for hydrogen production; the main contribution comes from water electrolysis. There is no active use of biomass on an industrial scale [6,12], which creates an opportunity to convert biomass into a valuable source of hydrogen. Research has been carried out to assess the economic pros and cons of prevalent and upcoming technologies in terms of low CO₂ emissions resulting from hydrogen production, such as gas switching–reforming [13], membrane-assisted fluidized-bed reactors [14], chemical looping reform (CLR) [15–17], autothermal reform (ATR) with CCS [16], steam methane reform (SMR) with chemical looping combustion (CLC) [16], and SMR with amine scrubbing [15], to evaluate the economic performance for low-emission hydrogen production; the average cost of hydrogen was 1.6–3.2 USD/kg. Kathe et al. investigated the thermodynamic limitations and energy consumption of the natural gas CLR hydrogen generation process using an iron-based oxygen carrier (OC) [18]. Adiya et al. investigated using NiO on Al₂O₃ and NiO on CaO/Al₂O₃ as catalysts and OC in a packed bed reactor for shale gas chemical looping steam reforming for hydrogen production, which showed a high fuel conversion and yield under operating temperatures of 700 and 750 °C [19,20]. Techno-economics analysis of three hydrogen production processes was investigated by Chisalita and Cormos [21], which concluded that efficient hydrogen production via chemical looping is the most efficient with low maintenance and operating costs. Furthermore, by utilizing iron oxide (OC) and CaO as CO₂ adsorbents, a new integration of sorption-enhanced CLR and water splitting for hydrogen generation was proposed by Saithong et al. [22]. Thus, utilizing calcium looping technology for biomass to hydrogen and power production with in situ CO₂ capture is projected to improve thermodynamic process efficiency, provide economic advantage, and achieve negative carbon emission. A comprehensive investigation of the calcium looping mechanism is planned.

Hosseini and Wahid [9] reported that supercritical water gasification of biomass is one of the most cost-effective thermochemical processes since wet biomass is fed directly into the gasifier without prior drying. Furthermore, generation at high pressure lowered the cost of hydrogen storage. Udomsirichakorn and Salam [23] studied calcium oxide in chemical looping of biomass gasification for hydrogen generation, and concluded that biomass gasification is a viable, environmentally benign, and long-term option for hydrogen generation, satisfying the environmental requirement. Adams and Barton [24] offered techno-economic studies for several technologies that were used to develop efficient polygeneration systems for processing coal and natural gas. Ahmed et al. [25] proposed an integrated model of coal gasification with SMR to enhance H₂ production by utilizing syngas heat and steam from the gasification process. Furthermore, Ahmed et al. [26] conducted a techno-economic analysis of both traditional coal and the integrated coal gasification and SMR process, establishing that the integrated model is more efficient. Moreover, Cormos [27] investigated techno-economic analysis of coal-fired combustion and gasification power plants having post-combustion CO₂ capture by utilizing a calcium looping cycle. It was concluded that calcium looping has good application potential in combustion-based power plants with 90% carbon capturing rate, low energy penalties, and high efficiency. Cormos [28] carried out a study on biomass direct chemical looping for cogeneration of hydrogen and power. This process provides high energy efficiencies and a carbon capture rate higher than 99%, which contrasts with various other CO₂ capture techniques (e.g., gas–liquid absorption). The economic factors revealed the reduction in CO₂ capture cost penalty for proposed technology in contrast to gas–liquid absorption. Jiang et al. [29] analyzed the thermodynamic and economic assessment of two chemical looping cycles schemes with conventional biomass gasification. The study provides a detailed comparison of chemical looping and

calcium looping with conventional biomass gasification in terms of capital cost, operating cost, and the levelized cost of energy. Calcium looping and chemical looping proved to be better options for the environment than conventional biomass gasification due to high carbon capturing efficiency.

In the literature, none of the studies have investigated the economic performance of the combined cycle of calcium looping gasification (CLGCC) with dual fluidized bed (DFB) in situ carbon capture or hydrogen and power generation by calcium looping gasification (CLGCC-H), which can offer near-zero or negative CO₂ emissions. Furthermore, all previous studies on the integration of chemical looping with IGCC and reforming processes were based on the additional use of steam and CO₂ to perform reforming processes. The current study mainly aimed to (1) propose and simulate a novel DFB, CLGCC-H, and CLGCC system with a CaO-based capture system, analyze the impact of different parameters on the system performance by Aspen Plus, and (2) to perform a comprehensive techno-economic data evaluation for both systems and to provide a reference framework for commercial operation.

2. Modeling and Simulation

2.1. Hydrogen and Electricity Production by Calcium Looping Gasification Process

The calcium looping gasification with the combined cycle for electricity generation (CLGCC) and calcium looping gasification with the combined cycle for hydrogen and electricity generation (CLGCC-H) schema mainly comprise flowing prime blocks in sequence, i.e., dual fluidized bed (gasifier and combustor), ASU, waste heat recovery (WHR), heat recovery steam generator (HRSG), gas cleaning (scrubber), acid gas removal unit (Selexol), elemental sulfur recovery unit (Claus), gas turbine (GT), and steam turbine (ST). The block diagram of CLGCC and CLGCC-H is shown in Figures 1 and 2. For hydrogen separation, the pressure swing absorption (PSA) is an additional unit after the selexol unit. Table 1 represent a design assumption for the CLGCC and CLGCC-H system.

Table 1. Design assumption for CLGCC and CLGCC-H.

Unit/Component/System	Modeling Unit	Parameters
Gasification Reactor	RYield, RGibbs (Reactor)	Biomass flow rate = 131.16 ton/h Temperature: 700 °C Pressure: 12 bar
Combustor Reactor	RGibbs (Reactor)	Temperature: 1000 °C Pressure: 12 bar
Air Separation Unit (ASU)	HeatX, Compr	O ₂ = 94.5% (vol) Power consumption: 235 kW/t
H ₂ S removal Section	RadFrac	Selexol solvent H ₂ S removal ~99.9% Absorber and regeneration stages: 12 each with thermal regeneration.
PSA	Sep2	Hydrogen separation ~99.99% Power consumption: 5.63 kW/ft ³ /m
Pump Efficiency	Pump	85%
Compressor Efficiency	Compr	82%
Steam Turbine Eff (Isentropic)	Compr	87.50%
Gas Turbine Eff (Adiabatic)	Compr	TIT (Turbine inlet temp) = ~1550 °C TOT (Turbine outlet temp) = ~570 °C Adiabatic efficiency = 85%
HRSG	HeatX, Compr	124/40/20/0.069 (bars) ΔT _{min} = 10 °C

Pine sawdust is used as fuel in this research; the chemical (ultimate and proximate) analysis results are recorded in Table 2. After preparation, fuel is fed to the gasifier, which reacts with the steam to produce hydrogen-rich syngas. The hydrogen-rich syngas is piped

to the waste heat recovery for steam generation. The unburned char from the gasifier and clean syngas after scrubber is combusted in a combustor with O_2 coming from ASU, which produces heat for gasifier needs and CaO regeneration, and CO_2 -rich flue gas produced in the combustor is piped to CO_2 storage after cooling. The flowsheet of the CLGCC is shown in Figure 3. After cleaning processes, the H_2 -rich syngas is routed for hydrogen and electricity through PSA or without PSA for power generation. The hydrogen separated is at 99.9 vol%, and the remaining syngas is combusted in a gas turbine after the GT steam cycle is used for power generation.

Figure 1. The calcium looping gasification with the combined cycle for hydrogen and electricity generation (CLGCC-H).

Table 2. Chemical analyses of pine sawdust.

Pine Sawdust		
Ultimate analysis	C	52.53
	H	6.08
	O	39.04
	N	0.09
	S	0.55
	A	1.70
Proximate analysis	V	85.15
	FC	13.15
	A	1.70
	LHV (kJ/kg)	19,853
	Moisture	2.07

Figure 2. The calcium looping gasification with the combined cycle for electricity generation (CLGCC).

2.1.1. Dual Fluidized Bed Calcium Looping Gasification

Figure 4 depicts the layout of the simplified process for H_2 production by DFB biomass steam gasification, consisting of gasification and combustor reactors. The carbonous fuel is fed into a fluidized bed that uses steam for fluidization. The gasification occurred at a temperature of $700\text{ }^{\circ}\text{C}$, with 10 bar pressure, and recirculated CaO was used as a CO_2 absorbent and formed CaCO_3 . Residual char and CaCO_3 from the gasifier after cyclone enter into the combustor, where char and dehydrated syngas from the gasifier (if required) is combusted with oxygen to reform CaO from CaCO_3 , producing CO_2 -rich flue gas at a combustion temperature of $1000\text{ }^{\circ}\text{C}$ and 12 bar pressure. Additionally, CO_2 is captured in DFB by the calcium looping process, based on Le Chatelier's Principle. Moreover, enhanced hydrogen production reactions, such as water–gas, water–gas–shift, and steam methane reforming, are all based on the same principle.

The energy required for gasification and the rate at which the bed material circulates influences the temperature differential between the combustion and gasification zones. The quantity of unburnt char that exits the gasifier with the bed material and the gasification temperature are two more factors that have an energy impact. This system is intrinsically auto-stabilizing because the decrease in the gasification temperature results in a higher quantity of residual char and, as an outcome, the combustion reactor has more fuel. Thus, it feeds more energy into the gasification zone, which adjusts the temperature accordingly. The process produces two distinct gas streams, H_2 -rich syngas from the gasifier reactor and CO_2 -rich flue gas from the combustor reactor, at high temperatures. The following are the significant reactions that occur in a dual fluidized bed.

Figure 3. The flowsheet of the calcium looping gasification with the combined cycle for hydrogen and electricity generation (CLGCC-H).

Figure 4. Basic concept of calcium looping gasification with CaO as bed material.

Gasifier Reactor (600–700 °C),
Water gas:

Hydro-gasification or methanation:

Methane steam reforming:

Water-gas shift:

Hydro-carbon steam reforming:

Hydro-carbon dry reforming:

Carbonation:

Combustor Reactor (830–930 °C),
CO oxidation:

Partial oxidation of carbon:

Total oxidation of carbon:

Calcination:

2.1.2. Air Separation Unit

The main purpose of the air separation unit (ASU), being a large-scale modern plant, is to feed highly pure O₂ to the gasification unit employing combined distillation and cryogenic air separation [30]. In practice, initially, the compressed liquefied air at 5–6 bar, is cooled using an expander to achieve liquefaction. Then, it is fed into the oxygen–nitrogen separation tower, having two parts, i.e., upper and lower. The upper tower's bottom is used to distill oxygen, the separation of O₂ from the N₂ and Ar, taking place at the base of the lower tower. The combustor unit employs O₂ (94.5 mol%) as an oxidant, while as stripping gas highly pure N₂ is utilized and N₂ is used for syngas dilution in the selexol unit and gas turbine, respectively. To thermodynamically simulate the ASU model, the

Peng–Rob EOS method was opted, whereas to model the separation tower, a RadFrac block was employed.

2.1.3. Heat Recovery and Water Scrubbing

The heavy particles are separated in the cyclone; heat is then recovered in HRSG from hot H₂-rich syngas, rich in fine particles. After heat recovery, syngas is fed to water scrubbing to eliminate fine particles. MHeatX and Flash2 models of Aspen Plus software are employed to simulate the heat recovery and fine particle trapping, assuming that the efficiency of fine particle separation is 100%. Then the clean and cool syngas, with a temperature of 45 °C, is fed to the selexol unit, while CO₂-rich gas at 100 °C flows to the CO₂ compression unit for subsequent storage.

2.1.4. Selexol Unit

Chemical and physical solvents are commonly employed in the desulfurization process on a commercial scale to remove the acid gas, particularly for H₂S. Since the physical absorption process is more energy-efficient at a very high concentration of H₂S [31], physical solvents, viz., dimethyl ether of polyethylene glycol (Coastal AGR[®] or DEPG; Selexol[™]), methanol (Rectisol[®]), n-methyl-2-pyrrolidone (NMP, Purisol[®]), and propylene carbonate (Fluor Solvent[™]) are advantageously preferred over the chemical solvents [32]. Specifically, as the Rectisol[®] is a complex process, it therefore has a higher operational cost. Further, the Fluor Solvent[™] process is preferred due to having little or no H₂S in the syngas [32]. In the selexol unit, DEPG is adopted as a solvent to remove H₂S.

The complex configurations of H₂S absorbers and the stripper that generates DEPG, causing a comparatively slow convergence speed, Sep block model is employed, representing the single-stage Selexol[™] process to remove H₂S from the water scrubbed clean syngas. The perturbed-chain statistical associating fluid (PC-SAFT) property method is chosen [31,33]. The concentration of H₂S-rich gas and the clean gas is employed by Field et al. [32]. Then, to generate elemental sulfur, acid gas-rich H₂S is fed to a Claus/SCOT plant [34]. The Claus process allows for a split-flow configuration with a portion of the H₂S supply going to the furnace and the rest bypassing it to mix in the first catalytic reactor. In the Claus process, sulfur condensers are used to eliminate sulfur, allowing the Claus equilibrium reaction to occur in the converter reactor. After two stages of condensers and converters, the remaining SO₂ is converted back to H₂S in the Tail Gas Thermal Unit (TGTU), allowing the tail gas to be recycled back into the selexol unit. Below is a summary of the responses [35,36].

2.1.5. Pressure Swing Adsorption (PSA) Unit

As a modern gas separation technology, comparatively, PSA is extensively utilized to achieve hydrogen purification, employing a physical adsorption technique [37]. It functions at an average temperature and varied pressure levels to remove impurities in hydrogen through an adsorbent. The PSA hydrogen production, an operationally stable process, is accomplished in three stages: high-pressure adsorption, low-pressure desorption, and pressure boosting. It helps to remove various impurities from the mixed gas simultaneously to produce a highly pure product. Firstly, the H₂-rich mixed gas is introduced into the adsorbent bed from the bottom at high pressure, making adsorption of impurities, viz., CO, CO₂, and CH₄ are weakly adsorbed components, resulting in hydrogen separation. Secondly, based on the efficiency of the adsorbed components, the adsorbent is reproduced by either product hydrogen purging or reverse pressure relief method to maintain the quality of the products. Following this, the pressure in the adsorbent bed is readjusted. Commonly, at least two or more adsorbent beds are utilized for sustainable production of the hydrogen cycle. For modeling the PSA, RK-Soave was adopted owing to its thermodynamic nature, whereas adsorption tower was simulated by adopting Sep block.

2.1.6. The Gas Turbine (GT) and Steam Turbine (ST)

Both the gas and steam turbine models are set up by using Aspen Plus. The clean syngas from selexol/PSA is directed to the combustion chamber. The exhaust gas is used in the gas turbine to generate electricity. Compressed air is also introduced to accomplish the combustion process. Additionally, separated nitrogen is mixed with the clean syngas to constrain the temperature at the gas turbine inlet to 1550 °C and to avoid the formation of nitrous oxides. Then, the exhaust from the gas turbine is fed into the heat recovery steam generator (HRSG), with additional heat being recirculated from different parts of the plant for steam production. A steam cycle occurs with the subcritical power generation technology, as the temperature of exhaust gas from the gas turbine is comparatively low (~570 °C). This superheated steam is used to drive the steam turbine, which generates additional electricity for the grid. Table 3 shows the specific parameters of the combined cycle (GT and ST). For the material property analysis, the PR-BM property package is utilized for analysis. In contrast, STEAM-TA is adopted for water and steam accessories including pumps and steam turbines.

Table 3. Specific parameters for combined cycle (GT and ST).

Facility	Parameter	Unit	Value
Air Compressor GT	Outlet Pressure	bar	21.2
	Inlet Temperature	°C	1550
	Outlet Temperature	°C	570
Steam Turbine	Inlet Temperature of HP	°C	520
	Inlet Pressure of HP	bar	124
	Reheating Temperature	°C	520
	Reheating Pressure	bar	40
	Inlet Temperature of IP	°C	373.55
	Inlet Pressure of IP	bar	20
	Outlet Pressure of LP	bar	0.069
Pump	Outlet Pressure	bar	124

3. Techno-Economic Evaluation

The production of hydrogen and electricity from biomass by calcium looping gasification process was realized using Aspen Plus; techno-economic evaluation was carried out using the simulation-based results.

3.1. Thermodynamic Evaluation

The first law of thermodynamics is used to determine the energy efficiency, while exergy efficiency is evaluated by the second law of thermodynamics. Energy and exergy efficiencies are simultaneously applied to investigate the hydrogen and electricity production by calcium looping gasification.

$$\eta_{\text{CLGCC-H}} = \frac{E_{\text{net product,tot}}}{E_{\text{in,tot}}} \times 100 = \frac{m_{\text{Biomass}} \text{LHV}_{\text{H}_2} + E_{\text{GT}} + E_{\text{ST}} - E_{\text{AUX}}}{m_{\text{Biomass}} \text{LHV}_{\text{Biomass}}} \times 100 \quad (13)$$

$$\varepsilon_{\text{CLGCC-H}} = \frac{Ex_{\text{net product,tot}}}{Ex_{\text{in,tot}}} \times 100 = \frac{Ex_{\text{H}_2} + Ex_{\text{GT}} + Ex_{\text{ST}} - Ex_{\text{AUX}}}{Ex_{\text{Biomass}}} \times 100 \quad (14)$$

$$\eta_{\text{CLGCC}} = \frac{E_{\text{net product,tot}}}{E_{\text{in,tot}}} \times 100 = \frac{E_{\text{GT}} + E_{\text{ST}} - E_{\text{AUX}}}{m_{\text{Biomass}} \text{LHV}_{\text{Biomass}}} \times 100 \quad (15)$$

$$\varepsilon_{\text{CLGCC}} = \frac{Ex_{\text{net product,tot}}}{Ex_{\text{in,tot}}} \times 100 = \frac{Ex_{\text{GT}} + Ex_{\text{ST}} - Ex_{\text{AUX}}}{Ex_{\text{Biomass}}} \times 100 \quad (16)$$

For these equations, $\varepsilon(\%)$ and $\eta(\%)$ represent the exergy and energy efficiencies, respectively; m_i denotes the flow rate of species i , and LHV_i is lower heating value of material i . E_{GT} (MW), E_{ST} (MW), and E_{AUX} (MW) represent the energy of net power generation

from the gas turbine, steam turbine, and power consumed by auxiliary equipment in the whole plant. Finally, Ex_{H_2} (MW), Ex_{GT} (MW), Ex_{ST} (MW), and Ex_{AUX} (MW) represent the exergy of hydrogen, net power generation from the gas turbine, steam turbine, and power consumed by auxiliary equipment in the whole plant. The exergy of biomass is calculated by Szargut's correlation [38], exergy of hydrogen is determined by Rant's equation [39].

The CO_2 capture efficiency η_{ccs} of CLGCC-H and CLGCC system is defined as:

$$\eta_{ccs} = \frac{X_{c \text{ in } CO_2\text{-rich flue gas}}}{X_{C \text{ in biomass feed}}} \times 100 \% \quad (17)$$

where $X_{c \text{ in } CO_2 \text{ rich gas}}$ is carbon mole flow in CO_2 -rich flue gas out from the combustor and $X_{C \text{ in biomass feed}}$ is carbon mole flow through biomass feed.

3.2. Economic Evaluation

The fixed capital investment (FCI), payback period, and internal rate of return (IRR) are used to evaluate and highlight the economic efficiency of CLG-hydrogen and electricity, considering the cost of associated facilities, input materials, and resulting products. The FCI is determined by the scaling-up method, promoted by Sweny et al. [40].

$$FCI = \sum_{a=0}^b \left[I_a V_{r,a} \left(\frac{C_a}{C_{r,a}} \right)^{S_a} \right] \quad (18)$$

where I_a , C_a , and S_a represent installation factor, scale, and scale factor of equipment in the calculated scale, respectively. $V_{r,a}$ and $C_{r,a}$ denote the reference cost and scale of equipment respectively, with b as the total number of devices. The price of every piece of equipment is calculated with reference to the cost index as defined by the thermal power engineering limit design [41].

$$\text{Cost in current year} = \text{cost in year } x \times \frac{CEPCI_{\text{current}}}{CEPCI_{\text{year } x}} \quad (19)$$

The effect of inflation is reported by employing the chemical engineering plant cost index (CEPCI), as given by Equation (19). The annual CEPCI for 2011, 2014, and 2017 is 585.7, 576.1, and 567.5, respectively, whereas the CEPCI for 2019 (607.5) is considered the current year CEPCI [42].

The feasibility of engineering projects is commonly evaluated based on the IRR, expressed as [43,44]:

$$\sum_{t=0}^n \frac{C_t}{(1 + IRR)^t} = 0 \quad (20)$$

where C_t denotes the cash flows of the year t , whereas n represents the calculation years.

The net cash flow C_t is calculated as per Equation (21) [44].

$$C_t = C_P - ((CRF(1 + \alpha) + OAM)FCI + C_F + C_M) \quad (21)$$

where C_P , C_F , and C_M denotes the annual income of products, fuel cost, and material cost, respectively. OAM represents the annual operation and management cost ratio to FCI, which is 4% of FCI. Further, α symbolizes the interest rate during the construction period, while the CRF denotes the average annual investment ratio [45]. Table 4 summarizes the detailed information about the capital cost of all facilities.

$$CRF = \frac{i}{1 - (1 + i)^{-n}} \quad (22)$$

The payback period is calculated as reported in [46]:

$$\sum_{n=1}^{P_t} C_t = 0 \quad (23)$$

where P_t denotes the payback period and n represents time.

Table 4. Plant equipment primary capital cost data.

Components	Basic Cost (10 ⁶ USD in 2019) *	Basic Scale	Scale Factor	Installation Factor	Reference
Biomass preparation (kg s ^{−1})	21.11	23.15	0.7	1	[47]
Gasifier (dual fluidized bed) biomass input (MW)	559.27	1198.03	0.6	1	[28]
ASU (O ₂ kg s ^{−1})	134.93	53.36	0.6	1	[47]
Selexol (t d ^{−1} sulfur input)	46.91	81	0.7	1	[34]
Claus/SCOT (t d ^{−1} sulfur input)	32.51	81	0.7	1	[34]
H ₂ PSA (kmol H ₂ h ^{−1})	17.79	16,616	0.65	2.56	[44]
Heat exchanger (MW _{th})	13.34	138	0.6	5	[48–50]
Power island					
Gas turbine (net MW)	118.36	276	0.75	1	[44]
Steam turbine and cycle (gross MW)	57.54	275	0.67	2	[48–50]
CO ₂ compression unit (m ³ h ^{−1})	76.76	430,850.77	0.67	1	[51]

* 1 USD = 6.91 RMB on average exchange rate in 2019 [52].

4. Results and Discussion

4.1. System Validation and Optimization

The biomass CLGCC–H and CLGCC models were verified using experimental data from our 10 kW system [53] and compared with other literature [54–56]. The model's conditions were found to be the same in both simulation and experiment. The simulated syngas compositions are acceptable compared with experimental data by the root mean square error (RMSE) method. In principle, the WGS and methane reforming process are kinetically restricted; larger plants have longer residence times of gases and solids within the furnace, contributing to a syngas that is closer to the final equilibrium result. The RMSE for H₂, CO, CO₂, and CH₄ are observed to be 0.025, 0.43, 0.33, and 0.6, respectively. However, simulation results obtained from this current study revealed no major difference from the previous study [53].

4.2. Effect of Gasifier Temperature on Hydrogen Yield and System Performance

The hydrogen production depends on gasifier temperature; Figure 5 shows that as the temperature rises from 650 to 750 °C, the hydrogen generation increases from 9594.1 to 10,970.6 kg/h, reflecting that the higher temperature is favorable for higher hydrogen yield. The key reason for increment in hydrogen yield is because of hydro-carbon reforming reactions (Equations (6) and (7)), methane reforming reaction (Equation (4)), and WGS reaction (Equation (5)) at elevated temperatures, resulting in a lower fraction of CO and CH₄. The result matched the previous findings [57,58]. In addition, the high temperature promotes tar cracking and endothermic reaction, which is advantageous in producing more H₂ [58,59]. High temperature also enables CO₂ production due to enhancement of biomass pyrolysis and water gas shift reaction. The CaO effectively absorbs the CO₂ produced (carbonation process (Equation (8))), which is enhanced by moving equilibrium of CaO carbonation towards forward direction, a function of temperature [53]. The system efficiency is increasing first and then decreasing, but the reduction in efficiency is not considerable. The reason is that with the increase in hydrogen production and separation by PSA, the remaining syngas quality decreases, resulting in decrease in the syngas heat output, which in turns produces less electricity by the combined cycle. The effect of temperature on calcium looping gasification limits hydrogen production; thus, it significantly affects the system efficiency.

Figure 5. The effect of gasifier temperature on hydrogen yield, net electricity generation, and system performance for CLGCC–H system.

Now we turn to the CLGCC system and analyze the effect of gasifier temperature on electricity generation. This situation is depicted in Figure 6, fairly reflecting an analogous behavior to that of the CLGCC–H system. One observes an increase in electricity generation as temperature increases for the temperature range 650–750 °C, reaching its maximum of 391.96 MWe corresponding to 700 °C. Beyond this point, a gradual decrease is witnessed, and eventually settles to 391 MWe at 750 °C. The underlying processes involves an increment in syngas quality with the inclusion of hydrogen content and hence an improved electricity generation vis-a-vis temperature. However, figuratively speaking, the corresponding values for electricity generation and system efficiency turn out to be low compared with the CLGCC–H system.

Figure 6. The effect of gasifier temperature on net electricity generation and system performance for CLGCC system.

4.3. Effect of Gasifier Pressure on Hydrogen Yield and System Performance

Another critical variable in the CLG process is pressure. Pressurized CLG process has some advantages over traditional atmospheric CLG process, e.g., increase in the rate of char gasification [53,60], the power reduction for compressing syngas to high pressure before using a gas turbine [61], and improvement in heat and mass transfer in fluidized bed [62].

Figure 7 shows that from 1 to 6 bar pressure, the hydrogen yield and other product gases remain constant; however, after 6 bar pressure, the hydrogen yield increases first and then decreases. This trend is because from 1 to 6 bar pressure carbonation reaction (Equation (8)) cannot be realized at calculated temperature (700 °C), after 6 bar pressure, the CO₂ absorption started and gradually increases with the increase of pressure. The partial pressure of CO₂ decreases due to its adsorption, which promotes water gas shift reaction to convert CO and H₂O in the gas phase to produce more H₂ accordingly. Further, it is observed from Figure 7 that at approximately 12 bar pressure, the hydrogen yield achieved a maximum value of 11,082.1 kg/h. In addition, at higher pressure (above 12 bar), the steam methane reforming reaction started to work in a reverse direction to produce CH₄ and H₂O, which in turn caused reduction in hydrogen yield. After 25 bar pressure, the results of the current study cannot be observed, and no study found in the literature reported any results above this pressure. However, at high-pressure, the CH₄ mole fraction in the gas phase increases with the favor of reverse steam methane reforming reaction (Equation (4)). Enhanced reverse reaction (Equation (4)) delivers more energy to maintain the gasifier temperature, increases carbon conversion and cold gas efficiency continuously under pressured conditions, and requires less char to be burned. The same aforementioned phenomena are also observed in the CLGCC system. Unlike the temperature case, the impact of gasifier pressure has a radically distinguished character for both electricity generation and system efficiency, depending upon whether one considers the CLGCC or CLGCC-H system. As shown in Figure 8, electricity generation shows an increment for the pressure range 6–12 bar, reaching its peak and eventually decreasing to its minimum of 365 MWe at 20 bar pressure. Compared to the CLGCC-H system, considerable increase is reported due to improved syngas quality. However, concerning system efficiency, one notes no considerable difference in terms of overall behavior, though the numbers differ. We can deduce from the results that gasification with high-pressure operation is a preferable alternative for energy conversion. Still, it may not be a suitable option for hydrogen production without providing costly and sophisticated downstream reformers. After a thorough assessment, it is concluded that the optimum pressure should be set between 12–20 bars.

4.4. System Energy Conversion Efficiency

The dual fluidized bed (CaO–CaCO₃ recirculation) is a crucial part of calcium looping gasification for hydrogen and electricity production. For calcination and carbonation, it is necessary to operate the gasifier and combustor at appropriate temperature and pressure to fulfil thermodynamic effectiveness. The gasifier temperature is controlled at 700 °C and the combustor at 1000 °C for 12 bar operating pressure. The experiments set the steam/C ratio and CaO/C ratio as 2 and 1 for optimized hydrogen yield [53,63]. The biomass treatment capacity of the CLGCC-H and the CLGCC power plant is 131.16 t/h. Table 5 compares the thermodynamic performance of the CLGCC-H and CLGCC power plants. In the CLGCC-H system, clean syngas from selexol is fed to PSA for hydrogen separation, which separates 10,284.57 kg/h of hydrogen. After hydrogen separation, the remaining syngas is combusted in GT to produce electrical power. In addition, the waste heat from different parts of the plant and flue gases produced by the GT enters into the HRSG to produce steam for power generation through the Rankine cycle. The CLGCC-H plant can realize the coproduction of hydrogen energy of 342.54 MW and the electricity of 147.55 MWe. The system energy and exergy efficiency of the CLGCC-H system can reach 64.75% and 60.79%, which is 10.56% and 9.57% higher than the CLGCC system. From the perspective of power

generation of the CLGCC system, the output of GT is 317.76 MWe, and the ST the total power generation is 95.17 MWe, with a net power output of 391.96 MWe.

Figure 7. The effect of gasifier pressure on hydrogen yield, net electricity generation, and system performance for CLGCC–H system.

Figure 8. The effect of gasifier pressure on net electricity generation, and system performance for CLGCC system.

Table 5. Thermodynamic comparisons of CLGCC and CLGCC–H with biomass and coal.

Facility	Parameter	CLGCC–H	CLGCC
Fuel input	Input pulverized biomass (kg/s)	36.43	36.43
	LHV (MJ/kg)	19.85	19.85
	Thermal input (MW)	723.31	723.31
Gasifier	Outlet pressure (bar)	12.00	12.00
	Outlet temperature (°C)	700.00	700.00
	Steam (kg/s)	26.25	26.25
	Syngas mass flow (kg/s)	23.21	23.21
	Syngas LHV (MJ/kg)	24.26	24.26
	Thermal output (MW)	562.98	562.98
Combustor	Outlet pressure (bar)	12	12
	Outlet temperature (°C)	1000	1000
	Oxygen mass flow (kg/s)	14.91	14.91
	CO ₂ -rich mass flow (kg/s)	53.79	53.79
Scrubber column	Pressure syngas (bar)	11.7	11.7
	Outlet temperature syngas (°C)	45	45
H₂S absorber	Outlet temperature syngas (°C)	11.92	11.92
	Mass flow syngas (kg/s)	12.8	12.8
	Syngas LHV (MJ/kg)	41.13	41.13
PSA	Hydrogen (kg/h)	10,284.57	-
	Hydrogen LHV (MJ/kg)	119.90	-
	Hydrogen energy output (MW)	342.54	-
Power block	Air compressor discharge (bar)	20	20
	Combustion chamber (bar)	19.02	19.02
	Gas turbine inlet temperature (°C)	1502	1502
	Gas turbine pressure ratio	18.65	18.65
	Gas turbine power output (MWe)	107.82	317.76
	Steam cycle power output (MWe)	39.73	95.17
Power output	Gross power output (MW)	490.09	412.93
	Auxiliary load (MW)	21.76	20.97
	Net power output (MW)	468.33	391.96
	Net efficiency (%)	64.75	54.19
	Exergy efficiency (%)	60.79	51.22
	η_{CCS} (%)	83.12	83.12

4.5. Economic Analyses

This section reports an economic analysis of the CLGCC–H system and compares it against the CLGCC system. Before the economic analysis, the baseline data for each part are presented in Tables 4 and 6, which present the assumptions for total production cost. In addition, Table 7 shows the baseline economic data, the economic comparison of the CLGCC–H and the CLGCC systems is carried out, as shown in Table 8.

Table 8 shows that the FCI of the CLGCC–H system is approximately USD 689.66 million, and the FCI of the CLGCC is USD 770.99 million, which is about USD 81.32 million higher than that of the CLGCC–H system. This is due to the higher investment in power island and heat recovery during electricity production. Low investment is required for the CLGCC–H system power island, as most of the hydrogen from H₂-rich syngas is separated through PSA for the downstream process (i.e., fuel cell, transport fuel, or oil upgradation) and less syngas is available for electricity production. According to the economic calculations, the production cost of H₂ is calculated to be 2.37 USD/kg; with this hydrogen price the IRR of the CLGCC–H system with dual products (hydrogen and electricity) reaches 17.43%, while that of the CLGCC system is only 11.45%. The CLGCC–H system payback period is 7.35 years, which is 2.64 years less than the CLGCC system. Owing to high IRR and short payback period, the CLGCC–H system is deemed to be promising.

Table 6. Assumptions of the total production cost.

Component	Basis
(A) Raw material	Pine sawdust 65.12 USD/t
(B) Utilities	Water 0.7 USD/t; electricity 0.07 USD/kWh
(C) Operating and maintenance	
(1) Operating labor + welfare	200 laborers (45,000 USD/laborer/year) + 10% operating labor
(2) Direct supervisory and clerical labor	20% of operating labor
(3) Maintenance and repairs	2% of fixed capital investment
(4) Operating supplies	0.7% of fixed capital investment
(5) Laboratory charge	15% of operating labor
(D) Depreciation	Salvage value: 4%, life period: 30 years
(E) Interest and surcharge	According the landing schedule
(F) Plant overhead cost	60% of (1) + (2) + (3)
(G) Administrative cost	2% of total production cost
(H) Distribution and selling cost	2% of total production cost
(I) Total production cost	(A) + (B) + (C) + (D) + (E) + (F) + (G) + (H)

Table 7. The basic economic assumptions.

Items	Value	Price	Value
Discount rate	8%	Biomass	65.12 USD/t
Plant life	30 years	Water	0.76 USD/t
Yearly running time	8000 h	Electricity	0.07 USD/kWh
Establishment period	3 years	Sulfur	115 USD/t
Tax	25%	Limestone	19.97 USD/t
Interest	7.53%		

4.6. Sensitivity Analysis

The IRR is a cost analysis method for determining the profitability of a process. The IRR is a discount rate that makes the H₂ processing plant's net present value (NPV) zero. This might bring the plant's cash input and outflow into balance: the higher the IRR discount rate, the lower the NPV and longer the payback period. The plant will be more successful if the IRR discount rate is lower, the NPV is larger, and the payback period is shorter [64]. Sensitivity to IRR is an important factor in determining the annual growth rate of plant investment, and it emphasizes the payback period.

Referring to the economic sensitivity analysis of the power plants with CCS [4,47], a sensitivity analysis was performed by varying main economic parameters leads, e.g., biomass price, electricity price, hydrogen price yearly running hours, and capital expenditure (fixed capital cost). The economic parameters are identified by variation from −50% to 50%.

Figure 9 shows the economic sensitivity of the CLGCC–H. It is observed that the IRR increases up to 25.14% corresponding to the 50% increase in hydrogen price. On the other hand, the IRR decreases 8.87% when the hydrogen price decreases by 50%. Similarly, the IRR can increase up to 23.83% when the fixed capital cost decreases to 50%. Apparently, the IRR is sensitive to the variation of hydrogen selling price and fixed capital investment. The other parameters such as biomass price and electricity price have a sensitivity effect on the IRR, but the degree of sensitivity is unlike the hydrogen selling price and fixed capital cost.

Similarly, Figure 10 shows the sensitivity analysis for the CLGCC system. The main product of the CLGCC system is only electricity, so the IRR increases up to 18.98% when the electricity selling price increases by 50%. In contrast, it could be down to 2.29%, while the price of electricity decreases to 50%. Similarly, the IRR can increase up to 19.85%, corresponding to a 50% decrease in fixed capital cost. The hydrogen selling price has no effect at all on the IRR with ±50% variation in hydrogen selling price. The IRR is slightly affected by the variation in biomass price.

From Figures 9 and 10, it can be observed that the yearly running time has a significant effect on the IRR. It could be down to 14.63% and 6.21% for the CLGCC–H and CLGCC, as the yearly running time decreases to 50%. Figure 11 shows sensitivity analysis over the

plant life (15 to 30 years). The IRR decreases to 15.49% and 8.54% for the CLGCC–H and CLGCC corresponding to 15 years decrease in the plant life.

Table 8. Economic comparison of CLGCC and CLGCC–H with biomass.

Items	CLGCC–H	CLGCC
Fuel and material consumption (10^6 t/y)		
Biomass	1.05	1.05
Water	1.24	1.24
Limestone (t/y)	6299	6299
Product		
Sulfur (t/y)	5681.83	5681.83
Hydrogen (99.99 vol% purity) (kg/y)	8.74×10^7	-
Electricity (kWh/y)	1.18×10^9	3.30×10^9
CO ₂	1.55×10^6	1.55×10^6
Capital cost (10^6)		
Fuel preparation	29	29
DFB (including gasifier, combustor, cyclones, fans, other facilities)	413.18	413.18
Air separation unit (ASU)	73.32	73.32
Selexol unit	15.76	15.76
H ₂ PSA unit	21.14	-
Claus/SCOT unit	10.92	10.92
Heat exchangers	26.03	30.38
Power island (GT+ST)	89.97	188.08
CO ₂ compression	10.35	10.35
Fixed capital cost	689.66	770.99
Working capital	137.93	154.20
Total capital investment	827.60	925.18
Raw material	68.46	68.46
Utilities	15.87	15.55
Operating and maintenance	30.86	33.09
Depreciation	23.29	26.07
Interest and surcharges	47.24	54.08
Plant overhead cost	14.81	15.80
Administrative cost	3.26	3.38
Distribution and selling cost	3.26	3.38
Total production cost	207.05	219.80
Hydrogen production cost ^a (USD/kg)	2.37	-
Revenue ^b (10^6 USD)	290.56	231.74
Profit before tax ^c (10^6 USD)	83.51	11.94
Tax rate	25%	25%
Profit after tax (10^6 USD/year)	62.63	8.96
IRR (%)	17.43	11.45
NPV (10^6 USD)	677.67	267.42
Payback Period (y)	7.35	9.99

^a Hydrogen production cost = Total production cost/H₂ yield. ^b Revenue = Hydrogen price × Hydrogen yield + Electricity price × Electricity production + Sulfur price × Sulfur yield. ^c Profit before tax = Revenue – Total production cost.

4.7. Uncertainty Sources of Sensitivity Analysis

According to Cristóbal et al. [65], the following uncertainty sources must be addressed for sensitivity analysis: method of cost calculation, revenue, utility cost, process variables, feedstock price, and transportation. Among the different causes of uncertainty in sensitivity analysis, the method for low TRL technologies cost estimating shows a $\pm 50\%$ deviation, as a result of failures in inflation projection or cost growth. It is due to a highly complex process's unexpected events and technology that have yet to be verified. The proposed project's revenue is primarily determined by the end product's composition, quality, quantity, and selling price, which are determined by its demand in the global market. The cost of utilities varies depending on the process type. Typically, hydrogen, electricity, and feedstock prices are considered basic factors that might fluctuate due to various circumstances, including environmental safety concerns, unpredictable weather conditions, geopolitical situations, and government taxes. In general, the results of laboratory experiments are utilized to scale up processes to obtain the same efficiency. However, in other circumstances, the process

variable costs are dependent on the size of scale-up plants, which has an indirect impact on revenue. The process profitability is also determined by the amount of transportation and feedstock availability.

Figure 9. Influence of product price on IRR for the CLGCC-H.

Figure 10. Influence of product price on IRR for the CLGCC.

Figure 11. Influence of plant life on IRR for CLGCC-H and CLGCC.

5. Conclusions

In summary, we proposed and carried out an in-depth techno-economic analysis of a novel energy conversion system that is based on calcium looping gasification of biomass for hydrogen and electricity production. The CLGCC-H system is investigated by employing process simulation using Aspen Plus simulation and compared to that of the CLGCC. A comprehensive comparison of the thermodynamic and economic performance of the CLGCC-H and the CLGCC systems is conducted.

We explicitly demonstrated, based on a thermodynamic and economic analyses, that CLGCC-H's system energy and exergy efficiency is higher than that of the CLGCC system. Concerning system energy, the CLGCC-H system's peak system energy stood at 64.75%, which is approximately 10.56% higher than the CLGCC system. Moreover, the exergy efficiency of the CLGCC-H system attains its peak at around 60.79%, which is roughly 9.57% higher than the CLGCC system. For economic analysis of the CLGCC-H system, the IRR was reported at 17.43% with a payback period of 7.35 years, while the same for the CLGCC system are 11.45% and 9.99 years, respectively. Based on these thermodynamic and economic analyses outcomes, we conclude that the CLGCC-H could potentially hold a great market value.

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Abbreviations

CLGCC–H	Calcium looping gasification with the combined cycle, for hydrogen
CLGCC	Calcium looping gasification with the combined cycle
IGCC	Integrated gasification with combined cycle
DFB	Dual fluidized bed
IRR	Internal rate of return
CCS	Carbon capture and storage
CLR	Chemical looping reform
ATR	Autothermal reform
SMR	Steam methane reform
CLC	Chemical looping combustion
OC	Oxygen carrier
ASU	Air separation unit
HRSG	Heat recovery steam generator
GT and ST	Gas turbine and steam turbine
PSA	Pressure swing absorption
FCI	Fixed capital investment
CEPCI	Chemical engineering plant cost index
NPV	Net present value.

Nomenclature

η and ε	System efficiency and exergy
E_{net}	Net power generation (MW)
E_{ST}	Power generation of steam turbines (MW)
E_{GT}	Power generation of gas turbines (MW)
E_{AUX}	Power consumption of auxiliary equipment (MW)
$\text{LHV}_{\text{biomass}}$	Lower calorific value of biomass (MW)
FCI	Fixed capital investment (10^6 USD)
Si	Production capacity index
Ct	Cash flows of the year t (10^6 USD)
CRF	Average annual investment ratio
OAM	Annual operation and management cost ratio
Cp	Annual income of products (10^6 USD)
CM	Material cost (10^6 USD)
C_F	Fuel cost (10^6 USD)

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