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Sustainability Evaluation of Biomass Direct Gasification Using Chemical Looping Technology for Power Generation with and w/o CO_2 Capture

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Abstract: Sawdust direct gasification using chemical looping with hematite as an oxygen carrier was investigated in a 10 kWth interconnected fluidized bed reactor. This was used to develop a biomass chemical looping gasification combined cycle (BCLGCC) model using Aspen Plus software. A technical analysis of a scaled up, simulated and validated 650 MW power plant using BCLG experimental results and industrial data was conducted. The analysis focused on investigating critical parameters that have a significant effect on syngas quality and quantity, hence optimizing the gasification process to obtain higher energy efficiencies for subsequent power generation. An economic and sustainability assessments comparing BCLGCC with 4 different power generation technologies with and w/o CCS was performed. BCLGCC presents promising economic and environmental results, showing that the

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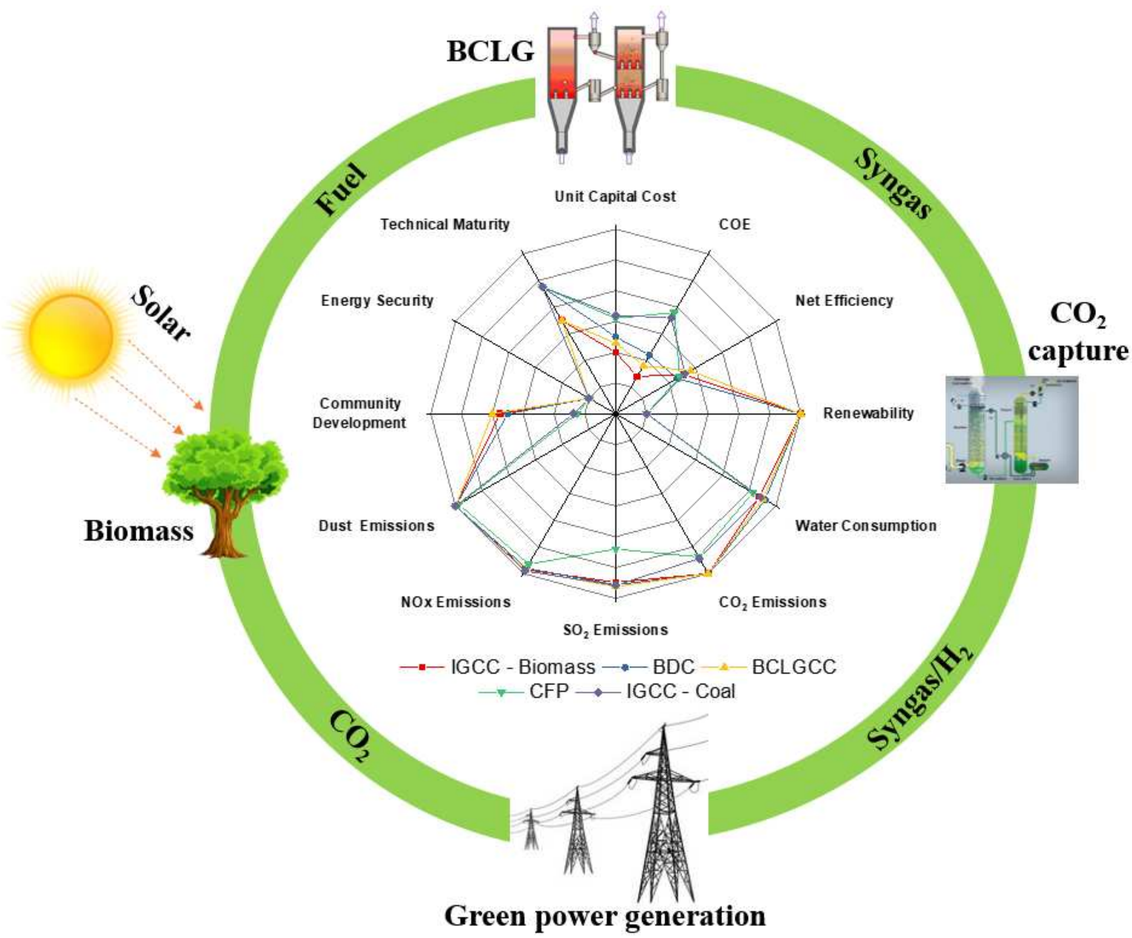
efficiencies of the CCS and Non-CCS plants are equal to 36% and 41%, respectively, with a COE (including government renewable energy subsidies) for both CCS and Non-CCS equal to 15.9 ¢/kWh and 12.8 ¢/kWh, both of which are lower than the average COE in the UK (approximately 17.7 ¢/kWh). This highlights the technical and economic potential and feasibility of BCLGCC compared to conventional power generation processes while promoting Bioenergy with Carbon Capture and Storage (BECCS) technology.

Key words: biomass gasification; chemical looping; power generation; CO_2 capture; sustainability assessment



1. Introduction

The fast-growing developing economies around the world and rapid increase in world population is linked with an everlasting increase in energy demands. The U.S Energy Information Administration estimated that the total world energy consumption is to increase by 28% in 2040 relative to 2015. The energy sector in particular contributes towards 41% greenhouse gas emissions through the use of fossil fuel and is expected to have the fastest growth rate [1]. Approximately 86% of worldwide energy consumption is from fossil fuel due to them being abundant and geographically spread over the world, hence considered as a cheap and reliable source of energy [2]. However, due to fears concerning the imminent and long-term risks associated with their use, it is imperative to push towards sustainable sources of energy. As a result, the Paris Agreement aimed to maintain temperature rise below 2°C by the end of this century relative to per-industrial levels and efforts to further limit the increase to 1.5°C [3], which has encouraged several nations to reduce their CO_2 emissions and move towards renewable energy. The United Kingdom's long-term target was to reduce their greenhouse gas emissions by at least 80% by the year 2050, relative to 1990 levels [4]. However, recently the UK House of Commons passed a net zero carbon emissions bill to be achieved by year 2050 [5], which means radical change in the entire UK's economy and power generation is required to achieve this target. In order to control CO_2 emissions, De Gouw et al., suggested two pathways; either phasing out as much fossil fuel and moving towards renewable energy or developing technologies with higher energy efficiency [6]. Moreover, it has been widely remarked that this target would be impossible to achieve without the employment of Bioenergy [7]. To further support a path towards a net-zero emissions by 2050 is the employment of Bioenergy with Carbon Capture and Storage (BECCS). Furthermore, the low sulphur content in biomass relative to coal results in less SO_X emissions makes it a more desirable fuel [8].

Biomass can be used to generate electricity by either combustion or gasification processes. Combustion takes places under enriched oxygen conditions and has an efficiency of approximately 25%, whereas gasification takes place under partial oxygen conditions to produce syngas which is then combusted to generate electricity, which generally has an efficiency up to 40% [9]. Up to date, biomass gasification processes have been continuously developing via several technologies [10, 11]. Conventional gasification processes are known to use air or enriched O_2 air with steam as the gasification medium [12]. However, using either air or pure oxygen is associated with downsides, which reduces the effectiveness of the process. Air results in a highly N_2 concentrated syngas, as a result reducing its energy density and would not make it suitable for chemical production. Whereas, using pure oxygen would require an additional air separation unit (ASU), which increases the parasitic energy as well as capital and operational costs. Additionally, tar formation in conventional gasification is a disadvantage as it reduces the gasification efficiency and can block downstream equipment [13].

Alternatively, chemical-looping technologies have recently emerged offering higher efficiencies and potentially lower costs. Chemical looping gasification (CLG), essentially involves the oxidation of the fuel using lattice oxygen in the form of a metal oxide (Me_xO_y) as a substitute to molecular oxygen. Instead of using one gasifier and an air separation unit, two reactors are used, a fuel reactor (FR) and an air reactor (AR). The highly oxygen enriched metal oxide is injected into the FR as a solid form where it is reduced to Me_xO_{y-1} as shown in Equation (1). As a result, biomass receives its oxygen in a lattice form instead of a molecular form and syngas is produced [14].

$$C_nH_{2m}O_P + (n-p)Me_xO_y \rightarrow nCO + mH_2 + (n-p)Me_xO_{y-1} \text{ (Reduction)} \quad (1)$$

The reduced oxygen carrier (Me_xO_{y-1}) is then circulated into the AR where it is oxidized back into its initial state (Me_xO_y) by reacting with oxygen from air, Equation (2). The regenerated oxygen carrier is then recycled back into the FR to repeat the process all over again.

$$(2n+m-p)Me_xO_{y-1} + (n+0.5m-0.5p)O_2 \rightarrow (2n+m-p)Me_xO_y \text{ (Oxidation)} \quad (2)$$

Biomass chemical looping gasification is associated with several potential advantages compared to conventional gasification: firstly, the oxygen carrier can provide a source of oxygen while avoiding the cost depleting process of adding an air separation unit. Secondly, the lattice oxygen has a higher chance of partially oxidizing the fuel compared to molecular oxygen (stronger oxidizing strength), resulting in an increase in a higher quality syngas (less CO_2). Moreover, tar cracking is enhanced due to the oxygen carrier's catalytic effect during biomass pyrolysis [15], as a result a thermal cracking process can be eliminated, hence improving the overall gasification process. Furthermore, chemical looping processes result in a reduction in exergy loss due to it undergoing moderate flameless gasification compared to the conventional thermochemical processes which undergoes severe flame gasification [16]. Research into biomass chemical looping gasification processes have been increasing in the past two decades. Experiments have been conducted on pilot scale, testing different types of oxygen carriers, biomass sources, conditions and set up which demonstrated higher efficiencies due to less energy penalties compared to conventional power generation processes [17-22]. However, very little techno-economic evaluation of biomass chemical looping processes (BCLPs) have been conducted, with even less focused on biomass chemical looping gasification to power generation as reviewed by [14]. Typically, Xu et al. only investigated chemical looping partial oxidation process for thermochemical conversion of biomass to syngas without considering the following power generation process [23]. Gopaul et al. compared to two chemical looping gasification processes for unique hydrogen production: one scheme produces H_2 using a CaO sorbent for CO_2 capture and total sorbent recovery; another scheme produces H_2 using Fe-based oxygen carriers for near-total carrier recovery [24]. Although Cormos et al. studied biomass direct chemical looping (BDCL) concept for hydrogen and power co-production from the aspect of process configuration, simulation, thermal integration and techno-economic assessment, nevertheless, in his study chemical looping combustion of biomass (rather than biomass direct chemical looping gasification) coupled with three reactors (fuel reactor, air reactor and steam reactor) are proposed to produce hydrogen and power simultaneously [25]. Besides, Ge et al. [26] recently developed a 30 t/h biomass chemical looping gasification combined cycle (BCLGCC) model on aspen plus which was validated and used to conduct a thermodynamic analysis, testing the effects of temperature and steam/biomass ratio on the HHV, gas composition and power generation. However, the scale of the simulation plant is small (electricity output is about 30-32 MW), which is not suitable for gas-steam turbine combined cycle to produce power due to its bad economic feasibility, as a result no economic analysis or sustainability evaluation of the power plant was conducted in their study. Aghabararnejad et al. [27] compared between biomass conventional gasification using pure oxygen as the gasifying agent and a small scale 7 MW_{th} BCLG system using Co_3O_4 (8%)/Al_2O_3 by developing an Aspen Plus model of each. A basic economic analysis of the Total Capital Investment (TCI) and operational costs were investigated just for the gasification unit, but no techno- economic analysis was conducted for the whole power plant. The author also compared between BCLG and conventional air gasification and concluded that the LHV of the syngas is higher for the BCLG process, which is due to the high concentration of N_2 in the syngas when using air as the gasifying medium.

Unlike the previous studies, this paper will give a comprehensive techno-economic analysis followed by a sustainability evaluation of an industrial scale 650 MW (gross power) direct chemical looping gasification of biomass to power generation, with CCS also considered when investigating the power plant. Allowing us to test the technical and economic feasibility of the process in large scale. An Aspen Plus simulation process of the whole biomass chemical looping gasification using hematite as oxygen carrier based on the experimental data from an interconnected fluidized bed reactor and the industrial or demonstration data was established and validated. A thermodynamic analysis using this model was conducted, discussing the effect of key parameters (oxygen carrier to biomass, gasification temperature, steam to biomass, pressure and WGS degree) that have a significant effect on syngas quality (net power output, syngas LHV and net efficiency) and quantity (gas yield) were investigated to obtain optimal conditions and maximum syngas output for the consecutive power generation process. This was followed by an economic feasibility analysis of the entire biomass direct chemical looping gasification combined cycle power plant. Moreover, a sustainability evaluation for both the BCLGCC-CCS and non-CCS power plants at a scale of 650 MW (gross power) electricity output was also conducted, which was compared to other power generation technologies such as coal-fired power plants, biomass direct combustion, traditional coal gasification and traditional biomass gasification to power generation technologies with and w/o CCS. From the above, the novelty of this paper is that we established a reliable industrial scale plant simulation process with and w/o CCS, on which the key operation parameters and conditions of biomass chemical looping gasification were optimized in order to obtain the maximum power output of the plant. This was followed by conducting a sustainability evaluation of the economic, environmental, social and technical indicators for both the BCLGCC-CCS and non-CCS power plants. The values were then compared to other power plants so as to highlight the advantages of the new BCLGCC plant and give a comprehensive understanding of BCLGCC technology for further development of the technology.

2. System description & materials

Figure 1 described a complete BCLGCC system for power generation. Biomass is gasified using a metal oxide to produce syngas which is then sent to a water gas shift (WGS) (which is needed for CCS scheme, Figure 1A) and carbon capture section where acid gas is separated from the syngas. The clean syngas is then sent to the combined cycle, consisting of a gas turbine, heat recovery steam generation unit and a steam turbine to generate electricity. The process for the Non-CCS plant is shown in Figure 1B. A BCLG subsystem model was established and validated based on a 10 kW_{th} interconnected dual fluidized bed gasification reactors (Figure 2), which consists of a bubbling bed FR, a cyclone to separate gas from particles escaping from the FR, a fast fluidizing bed AR, another cyclone for the gas exiting the AR, an upper and a lower loop seal. The detail parameters of each part were described in our previous work [19]. Sawdust of pine collected from Guangdong province (China), was used as fuel in the tests. Natural hematite is selected as the oxygen carrier due to its low cost, ability to withstand conditions inside a combustor (good stability at high temperature; high melting point), non-toxic in nature and has no negative environmental impact. The properties of biomass and hematite are shown in Table 1. The detail introduction of each key units such as gasification (FR & AR), WGS and acid gas treatment and combined cycle are presented in the **Supporting Information.**

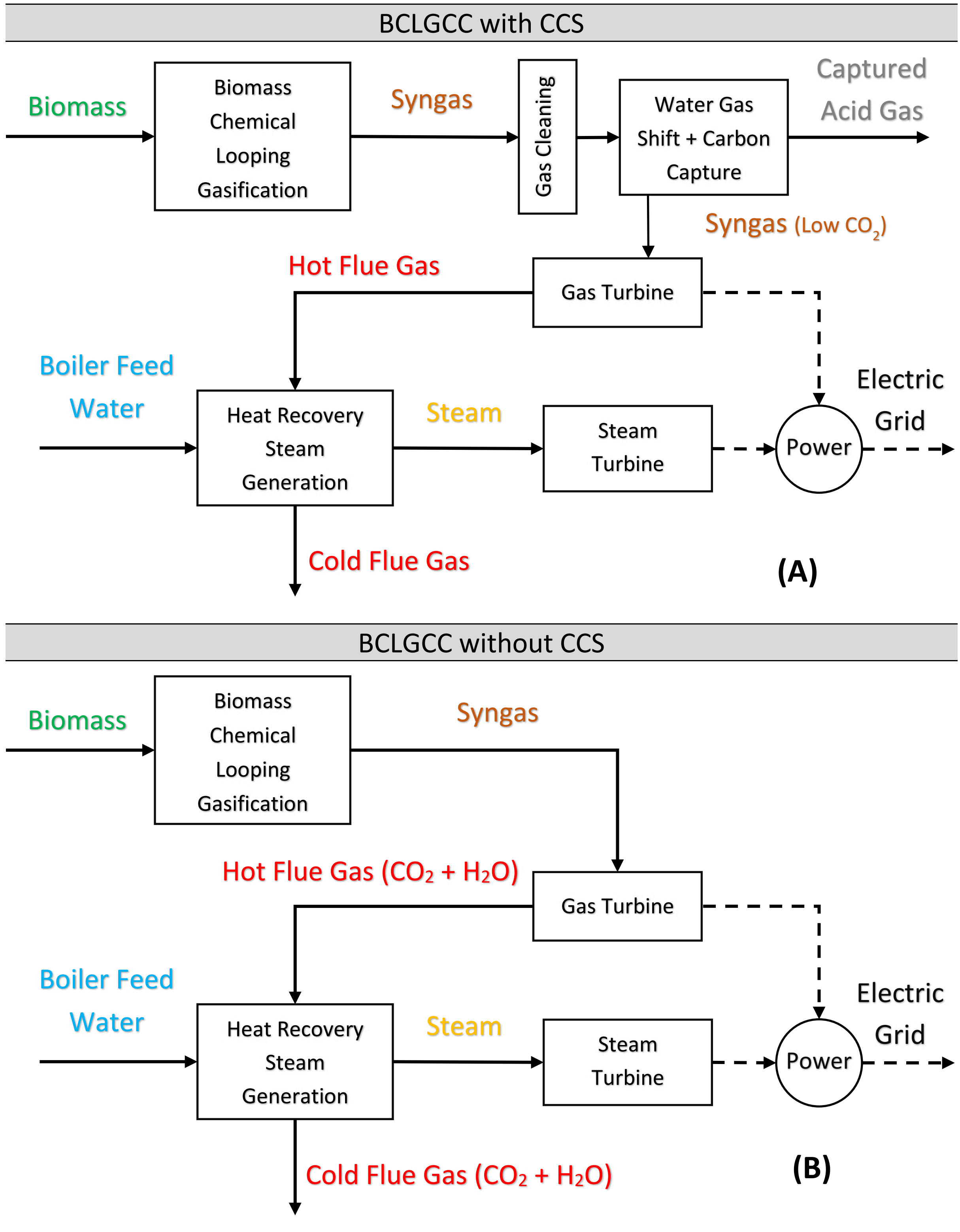


Figure 1. General block flowsheet of a BCLGCC to power generation with CCS (A) and without CCS (B)

Table 1. The properties of biomass sample and hematite oxygen carrier

Biomass										
Proximate analysis (wt.%,db)				Ultimate analysis (wt.%,db)					LHV (MJ/kg,db)	
Moisture	Volatiles	Fixed carbon	Ash	C	H	O	N	S	18.71	
8.39	84.31	6.88	0.42	46.44	6.21	47.29	0.05	0.01		
Hematite, wt.%										
O	Fe	Si	Al	Ca	Zn	P	Mg	K	Ti	Mn
30.1	64.61	2.83	1.4	0.09	0.53	0.10	0.08	0.02	0.03	0.05

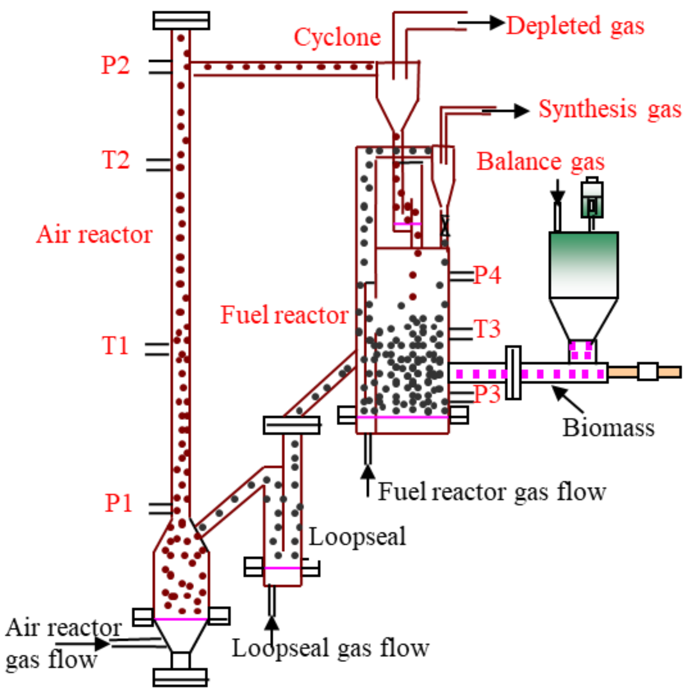


Figure 2. Schematic diagram of the interconnected circulating fluidized beds for BCL

3. Modelling and assumptions

A gasification model for a BCLG to power generation process has been developed based on restricted phase and chemical reaction equilibrium according to experimental study showing a stable and thermodynamic equilibrium results (**Supporting Information** Figure 1S and 2S). Aspen Plus software (V10.0) package was used to simulate the reaction steps occurring in both processes. The property method chosen for the gas – solid modelling was PR-BM (Peng Robinson equation of state with Boston - Mathias modification) [28]. In the simulation, biomass, ash and slag were defined as nonconventional components. The HCOALGEN and DCOALIGT property models were used to determine their enthalpies and densities, respectively. All other components are considered as conventional components, with Fe_2O_3, Fe_3O_4, FeO and carbon being considered as solids. Different unit operation blocks were used to establish the process flowsheets. Table 1S explains the function of each unit block used in the gasification process flowsheet.

Feedstock biomass is an unconventional component in the Aspen Plus software, converting biomass to a conventional component is necessary before its gasification process. RYield reactor has a special function that can convert an unconventional component to a conventional component, which is widely used in the coal or biomass pyrolysis and gasification [29,30]. Therefore, in BCLG process, Biomass (unconventional component) is initially injected into a RYield reactor where it is decomposed into its constituent components (conventional components). In general, this decomposed process is regarded as biomass pyrolysis which is only temporary placeholder. The actual yield distribution of the products out from RYield reactor is calculated using the FORTRAN statement calculation (see Supporting Information), a calculation method within Aspen Plus, for the biomass pyrolysis which is only a temporary placeholder of the biomass based on its given ultimate analysis into its constituent components, including carbon, oxygen, hydrogen, sulphur, nitrogen and ash. Since the ash is an unconventional component in Aspen Plus with an unknown composition, and the unreacted carbon mixing with the ash in biomass gasification, both the ash and the unreacted carbon finally form slag. Therefore, the above process can be simulated by a stoichiometric reactor due to knowable quantities of the ash and the unreacted carbon. The stoichiometric reactor is used when the reaction kinetics are unknown or unimportant but stoichiometry and extent of reaction are known. A stoichiometric reactor is generally used to react all the ash with the unreacted carbon to produce slag in biomass or coal gasification process [31]. The amount of ash would depend on the biomass ultimate analysis. The composition of slag changes depending on the gasification reaction conditions (depending on the amount of carbon conversion efficiency). On the basis of the above, those components out from the RYield reactor are then sent to a stoichiometric reactor where a percentage of carbon (the amount of unreacted carbon in gasification) reacts with the ash to form slag [32], and slag will not react in the FR.

The products from the stoichiometric reactor are sent to the RGibbs FR where they react with the oxygen carrier and steam. The main products from the reactions taking place in the RGibbs reactor are CO, H_2, CO_2, CH_4 and H_2O, as well as some trace components such as NH_3, H_2S and COS. The stream exiting the FR is sent to a cyclone where the syngas is separated from the solids (slag and reduced oxygen carrier) which are sent to the AR. In the AR, air is injected for oxygen to react with and regenerate the oxygen carrier for it to be recycled back into the FR. In general, a typical biomass gasification undergoes the following three main steps including: pyrolysis, gasification and combustion [33]. Biomass decomposes in the absence of oxygen into its heavy and light hydrocarbons as well as char. The energy required to allow this endothermic reaction to take place is supplied by the energy released during the combustion reactions. Since very little tar was collected in the experiments due to catalytic effects of oxygen carrier on tar cracking [15,34], only char and pyrolysis gas products were considered in the biomass pyrolysis process in the simulation. All the reactions in BCLG process is presented in Table 2, especially, combustion reactions of biomass-derived gas with the iron ore in the FR proceeds through reactions (6-11) based on analysing the X-ray diffraction results of OC (Figure 3S). The simulation flowsheet for BCLG is presented in Figure 4S in the **Supporting Information**.

Table 2. Biomass Chemical Looping Gasification Reactions in the Fuel and Air Reactors

	Chemical looping gasification
Reaction Number	**Equations**
	Biomass pyrolysis
1	$C_nH_{2m}O_x \rightarrow char + tar + syngas\ (CO, H_2, CO_2, CH_4, C_nH_{2m})$, (Endo.)
	Biomass gasification
2	$C + H_2O \rightarrow CO + H_2$ (Endo.)
3	$CH_4 + H_2O \rightarrow CO + 3H_2$ (Endo.)
4	$C + CO_2 \rightarrow 2CO$ (Endo.)
5	$C + 2H_2 \rightarrow CH_4$ (Exo.)
	Combustion reactions
6	$CO + Fe_2O_3 \rightarrow 2FeO + CO_2$ (Exo.)
7	$CO + 3Fe_2O_3 \rightarrow 2Fe_3O_4 + CO_2$ (Exo.)
8	$H_2 + 3Fe_2O_3 \rightarrow 2FeO + H_2O$ (Endo.)
9	$H_2 + Fe_2O_3 \rightarrow 2Fe_3O_4 + H_2O$ (Exo.)
10	$CH_4 + 3Fe_2O_3 \rightarrow 2Fe_3O_4 + CO + 2H_2$ (Endo.)
11	$CH_4 + 4Fe_2O_3 \rightarrow 8FeO + CO_2 + 2H_2O$ (Endo)
	Water gas shift (WGS)
12	$CO + H_2O \leftrightarrow CO_2 + H_2$ (Exo.)
	Oxygen carrier regeneration (Air reactor)
13	$4Fe_3O_4 + O_2 \rightarrow 6Fe_2O_3$ (Exo.)
14	$4FeO + O_2 \rightarrow 2Fe_2O_3$ (Exo.)
15	$C + O_2 \rightarrow CO_2$ (Eno.)
	Contaminants
16	$N_2 + 3H_2 \rightarrow 2NH_3$
17	$S + H_2 \rightarrow H_2S$
18	$CO + S \rightarrow COS$

Several assumptions were taken when setting up the simulations including:

1. The process is taking place at steady state;
2. The gasification blocks FR and AR are isothermal;
3. Ash and slag are assumed to be inert therefore do not participate in chemical reactions;

4. Large molecular weight compounds (C_xH_y; were x>1 and y>4) are not considered in the gasification reaction;
5. Gas-solid separation was 99% efficient;
6. Acid gas treatment was assumed to be 90% CO_2 and 98% sulphur.

Models validation of the whole plant. To ensure that the model is accurate and suitable to investigate thermodynamic characteristics of the gasification processes and the combined cycle, they should be validated with previous experimental results. Table 2S in **the Supporting Information** compares between experimental and simulation results for both the BCLG and combined cycle, respectively. It was noted that the difference between the simulation data and the literature or experimental data is within the rational range, implying that the established model is feasible for the simulation of the gasification and combined cycle processes. The other key models such as WGS, gas cleaning, CO_2 capture, heat recovery steam generation subsystem and gas-steam turbine combined cycle unit are detailed modelling and validated in previous work [35].

4. Data evaluation

4.1 Technical analysis

The technical indicators in this assessment are the Net Electric Efficiency (η_{Net}), Cold Gas Efficiency (CGE) and Lower Heating Value (LHV). To evaluate the performance of BCLG power systems, the η_{Net} is calculated using Equation 1, which is in accordance with the first law of thermodynamics.

$$\eta_{Net} = \frac{(W_{Gross} - W_{Syngas} - W_{CCS} - W_{OC})}{LHV} \times 100 \quad [1]$$

where, W_{Gross} is the gross power of the power plant, W_{Syngas} is the power caused by fuel gas compression, W_{CCS} and W_{OC} are the power required for CO_2 capture and OC circulation, respectively.

The Cold Gas Efficiency (CGE) was calculated as the percentage of syngas heating value over the biomass heating value, as shown in Equation 2.

$$\text{Cold gas efficiency, \%} = \frac{\text{Syngas LHV} \times \mathrm{Y_n}}{Biomass\ LHV} \times 100 \quad [2]$$

where $\mathrm{Y_n}$ is the biomass gasified gas yield, which is calculated as the volume of a gaseous component (V_n) per mass of biomass ($M_{Biomass}$) used, as shown in Equation 3.

$$\text{Gas yield } (\mathrm{Y_n}) = \frac{\mathrm{V_n}}{\mathrm{M_{Biomass}}} \quad [3]$$

The LHV of the syngas mixture was determined by its gaseous composition and can be calculated using Equation 4.

$$\mathrm{LHV} = 10110.71\,\frac{\mathrm{kJ}}{\mathrm{kg}}(\%\mathrm{wt.\,CO}) + 120850\,\frac{\mathrm{kJ}}{\mathrm{kg}}(\%\mathrm{wt.\,H_2}) + 50231.25\,\frac{\mathrm{kJ}}{\mathrm{kg}}(\%\mathrm{wt.\,CH_4}) \quad [4]$$

Since solid carbonaceous fuels negatively impact the environment, it is important that the amount of CO_2 generated during the process of power generation is measured and estimated. Even though biomass is used and is assumed to be carbon neutral, capturing the CO_2 after the power generation process can result in a negative emission, as a result aiding in tackling climate change. The reason for using gasification to generate electricity is due to it converting the carbonaceous fuel into syngas (H_2 + CO). This mixture of gas (syngas) then flows into a water-gas shift reactor, reacting with steam to increase H_2 concentration and convert the CO into CO_2. This allows us to capture the CO_2 pre-combustion, therefore reducing CO_2 emissions into the environment. Equation 5 was used to determine the degree of water gas shift achieved by varying the flowrate of steam, and Equation 6 was used to calculate the ratio of carbon captured from the syngas.

$$WGS\ Degree = \frac{CO\ in\ syngas - CO\ in\ shifted\ gas}{CO\ in\ syngas} \quad [5]$$

$$Carbon\ Capture\ Ratio = \frac{CO_2\ in\ shifted\ gas}{CO_2 + CO + CH_4\ in\ syngas} \quad [6]$$

4.2 Economic analysis

The CLGCC power plant in this paper is estimated using the method developed by NETL, which is described in [36]. It is a reliable tool when comparing between power plants. In this economic analysis, the Total Plant Cost (TPC) and the Cost of Electricity (COE) are calculated due to their wide application when comparing between power plants. To calculate these values, the cost of investment for an equipment is calculated based on market investigation to obtain reliable results. The market costs are used to estimate the cost of the equipment depending on the size using the Equation 7.

$$I_2 = I_1 \times (\frac{Q_2}{Q_1})^n \quad [7]$$

where I_1 & Q_1 are the equipment cost and production capacity of the reference equipment, respectively, I_2 & Q_2 represent the equipment cost and production capacity of the estimated equipment, respectively. Symbols n represents the scale exponent. All cost values for each unit are given in Table 3S, the detailed calculation of the Total Overnight Cost (TOC) is introduced in the **Supporting Information** including calculations of the variable and fixed costs in Tables 4S, 5S and 6S.

The COE of a power plant, which is the revenue received when generating electricity during the first year is calculated using Equation 8.

$$\mathrm{COE} = \frac{\mathrm{TOC}\times\mathrm{CRF}+(OC_{VAR})(CF)+OC_{FIX}}{(\mathrm{CF})(\mathrm{kWh})} \quad [8]$$

where CRF is the Capital Recovery Factor, calculated using Equation 9, OC_{VAR} is the sum of all variable operational costs, OC_{FIX} represent the fixed operational cost (including fuel at 100% capacity) CF is the Capacity Factor and kWh is the net kilowatt-hours of power generated at 100% capacity.

$$\mathrm{CRF} = \frac{i}{1-(1+i)^{-N}} \quad [9]$$

where, i is discount rate and N is the plant life.

The total annual cost for the OC ($COST_{Annual}$) is calculated using Equation 10.

$$COST_{Annual} = COST_{OC} \times \frac{M_{OC}}{LT_{OC}} \quad [10]$$

where $COST_{OC}$ is the cost of the OC per mass, M_{OC} is the total solid inventory and LT_{OC} is the OC lifetime (1315 h) [37].

CCS technology can be used to reduce carbon emissions despite the fact that it reduces the efficiency and increasing the cost of the whole process, the Cost of CO_2 Capture can be another parameter to evaluate their economic feasibility of the process. It can be calculated using Equation 11;

$$Cost\ of\ CO_2\ Capture = \frac{(COE_{CCS}-COE_{Non\ CCS})\$/MWh}{(tCO_2/MWh)_{captured}} \quad [11]$$

To test the effect of plant size of the capital cost, variable cost, fixed cost and COE, Equations 12, 13, 14 and 15 are used, respectively.

$$TOC_s = TOC_R \times (\frac{MW_s}{MW_R})^{0.7} \quad [12]$$

$$VC_S = VC_R \times (\frac{MW_s}{MW_R}) \quad [13]$$

$$FC_S = FC_F \times (\frac{MW_s}{MW_R})^{0.7} \quad [14]$$

$$\text{COE} = \frac{\text{TOC}_S \times \text{CRF} + (VC_S)(CF) + FC_S}{(\text{CF})(\text{MW}_S)} \quad [15]$$

where, TOC_s and MW_s are the total overnight cost and net power of the scaled plant, whereas, TOC_R and MW_R are the capital cost and net power of the reference plant, respectively. $VC_S\ and\ VC_R$ are the variable costs of the scaled and reference plant, respectively. $FC_S\ and\ FC_R$ are the fixed costs of the scaled and reference plants, respectively.

4.3 Sustainable assessment

Sustainable indicators assist in assessing and providing a holistic and integrated evaluation of a process performance. This will allow us to understand the advantages and disadvantages of the analysed process. Four indicators are used to assess the sustainability of a process including, economic, environmental, social and technical sustainability. The detailed introduction and composition of the above four indicators are presented in the **Supporting Information.**

To understand, evaluate and compare the overall sustainability of different power plants more clearly, the aforementioned indicators are normalised according to Equation 16 [38].

$$X_{ij} = \frac{x_{ij} - worst\{x_j\}}{best\{x_j\} - worst\{x_j\}} \quad [16]$$

where x_{ij} and X_{ij} represents the indicator and normalised indicator j for the process *i*, respectively; $\text{worst}\{x_j\}$ and $\text{best}\{x_j\}$ are assumed to be the worst and best cases of indicator

j. The normalised indicator (X_{ij}) varies from 0 - 1. The higher the value the better its sustainability is.

5. Results and discussion

5.1 Technical assessment

Table 3 lists the performance results of the CCS and Non-CCS power plants. Both CCS and Non-CCS power plants were set to have the same gross power output of 650 MW, with an inlet biomass flowrate of 6480 and 6600 ton/day, respectively. CCS power plant has a 90% CO_2 capture and resulted in a 5% decrease in efficiency (36%) compared to Non-CCS (41%). The BCLG reactions cannot occur at temperatures below 650 °C, and the sintering of oxygen carrier is prone to occur when the temperature exceeded 1000 °C [19]. A thermodynamic analysis was conducted to measure the effect of temperature of the FR (750 - 950°C) on gas composition, LHV, net power, cold gas and net efficiencies, while keeping the AR temperature 100°C higher than the FR. Similar tests were also conducted measuring the effect of pressure ranging from 1 to 13 atm for both FR and AR, OC/B ratio ranging from 0 - 10, and S/B ratio from 0.2 - 1.8.

Table 3. Performance results of both CCS and Non-CCS plants

Parameter	Non-CCS Plant	CCS Plant
Feed Rate, ton/day	6600	6480
Gas Turbine Output, MW	417	377
Steam Turbine Output, MW	233	273
Gross Plant Size, MW	650	650
Syngas Compression, MW	95	92.2
OC Circulation, MW	8.8	8.8
CO_2 Capture Power	-	45.5
Net Power, MW	546	504
CO_2 Captured, %	-	90%
Amount of CO_2 captured, kmol/s	-	2.37
WGS Steam Flowrate, kmol/s	-	2.25
Biomass LHV, MW	1429	1403
Efficiency, %	41	36

5.1.1 Effects of Temperature

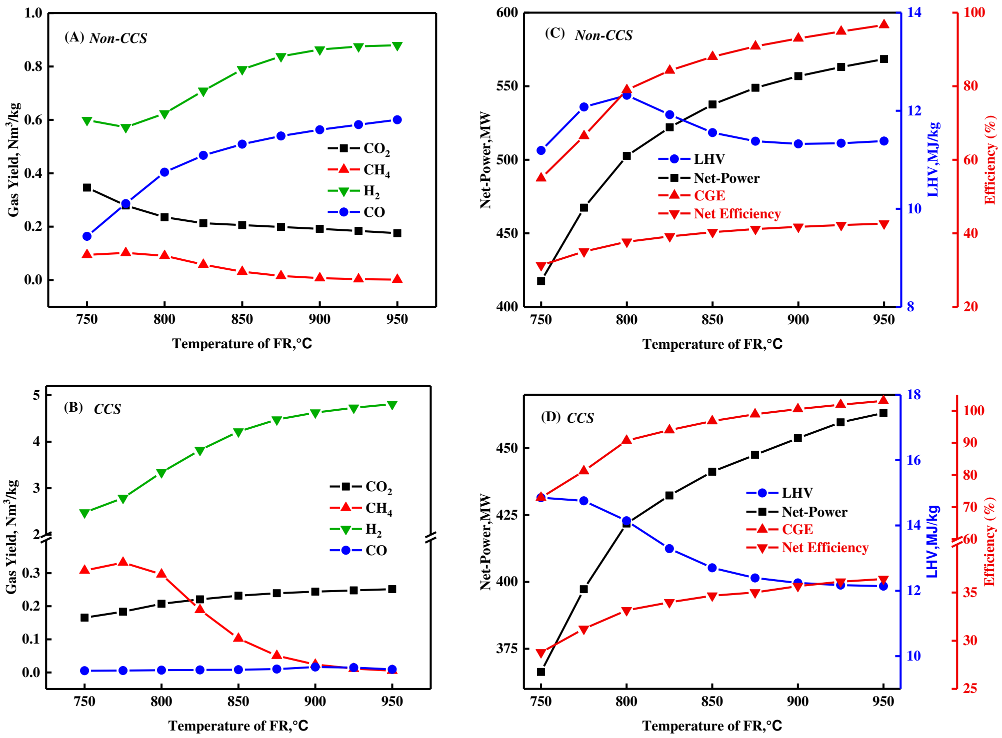


Figure 3. Effect of temperature on both CCS and Non-CCS power plants

Temperature has a significant effect on the syngas composition. As shown in Figure 3A, as temperature increased from 750 °C, H_2 and CO experienced an increase in yield, whereas, CH_4 and CO_2 yields decreased. The change in compositions reached a plateau at around 875°C, therefore, was taken as the optimum temperature, ensuing a balance between further increase in temperature and the percentage increase in efficiency. These effects can be explained by the thermodynamics of reactions (1 -12) in the Table 2. The LHV of the syngas sent to the combined cycle, increased from 11.2 MJ/Nm^3 at 750°C to 12.3 MJ/Nm^3 at 800°C, which is due pyrolysis gas (having high LHV due to more CH_4 formation). However, as temperature further increases, the LHV decreases and plateaus at around 875°C. This decrease in LHV is compensated by an increase in gas yield due to increased carbon conversion, as a result increasing the CGE of the gasification process. This increase in CGE resulted in an increase in net power and consequently the net efficiency of the power plant.

This can be observed in Figure 3B. Comparing BCLGCC w/o CO_2 capture with BCLGCC with CO_2 capture, as temperature increased from 750 °C H_2 yield increased rapidly while CO_2 slightly increased. On the other hand, CH_4 composition decreased while CO remained negligible. The reason for that is due to the presence of a WGS unit, converting all the CO into CO_2, where 90% is captured. Since the ratio between C and H is 1 to 4 in CH_4, more H_2 is produced as it reacts with the OC as temperature increases. From Figure 3C and 3D, an increase in temperature resulted in an increase in CGE due to increase gas yield, net power and net efficiency. The net efficiency of the capture power plant was approximately 6% lower than the non-capture power plant.

5.1.2 Effect of OC/B ratio

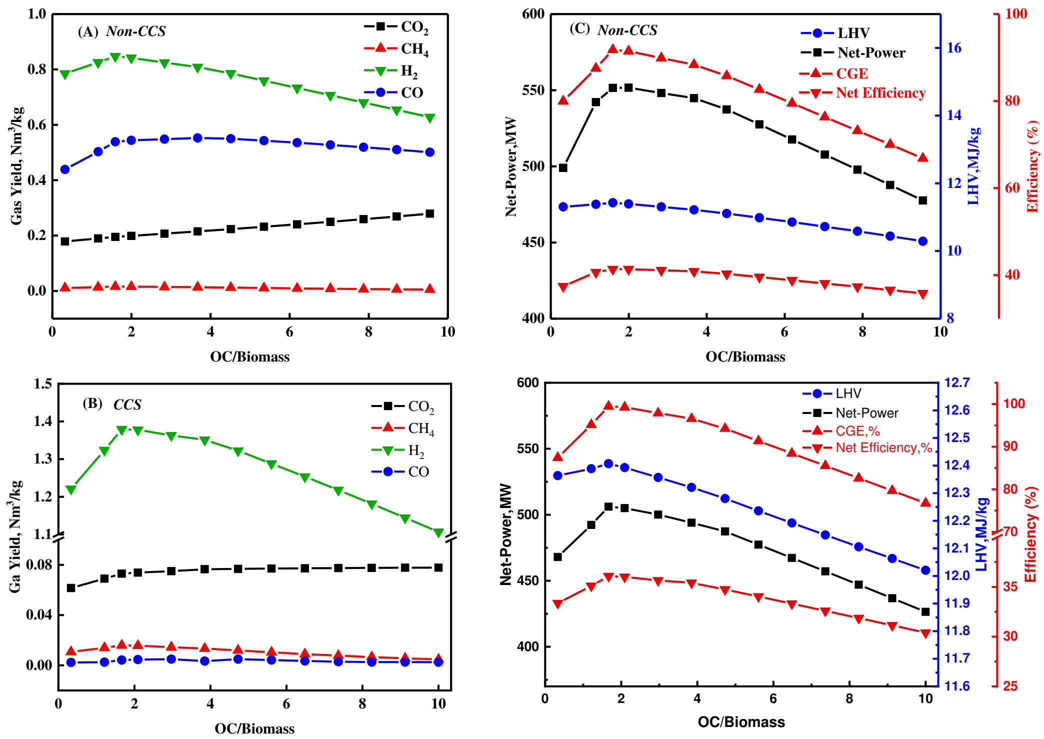


Figure 4. Effect of OC/Biomass ratio on both CCS and Non-CCS power plants

The OC/biomass (OC/B) ratio is an important parameter in BCLG to obtain high-quality syngas. High amount of oxygen carrier will result in complete combustion while little amount of oxygen carrier will result in insufficient amount of oxygen and gradual decrease in gasification temperature of the reactor. Figure 4A demonstrated that as the OC/B ratio increased from 0.30 to 2 H_2 yield increased, and similarly CO, CO_2 and CH_4 yield increased. This is because as we increase the OC/B ratio, the amount of lattice oxygen increases hence converting more carbon into CO/CO_2. Additionally, more OC results in an increased catalytic cracking of the tar, therefore resulting in higher amount of carbon being converted into syngas. Further increase in OC/B ratio beyond 2, stagnates CO yield while gradually increasing CO_2, while CH_4 and H_2 yields experience a gradual decrease, due to the increased oxidation, resulting in CO, CH_4 and H_2 being converted into CO_2 and H_2O. Compared to BCLGCC with capture, the trends in Figure 4B are similar, however, the CO and CH_4 yields can be assumed to be negligible while H_2 flowrate is approximately double, mainly due to the WGS and carbon capture processes. The slight increase in CO and H_2 compositions between 0.3 and 2 OC/B ratio resulted in an insignificant effect on the LHV compared to the effect it had on the gas yield which resulted in an increase in CGE. The increase in OC/B ratio from 0.3 to 2 OC/B also resulted in a 50 MW net power increase for the non-capture plant, but approximately 36 MW increase for the capture plant. As a result, increasing the net efficiency for both capture and non-capture plants by 2.6% and 4%, respectively. However, as OC/B increases more than 2, all factors gradually decrease due to complete combustion taking place, as demonstrated in Figures 4C &D.

5.1.3 Effect of Pressure

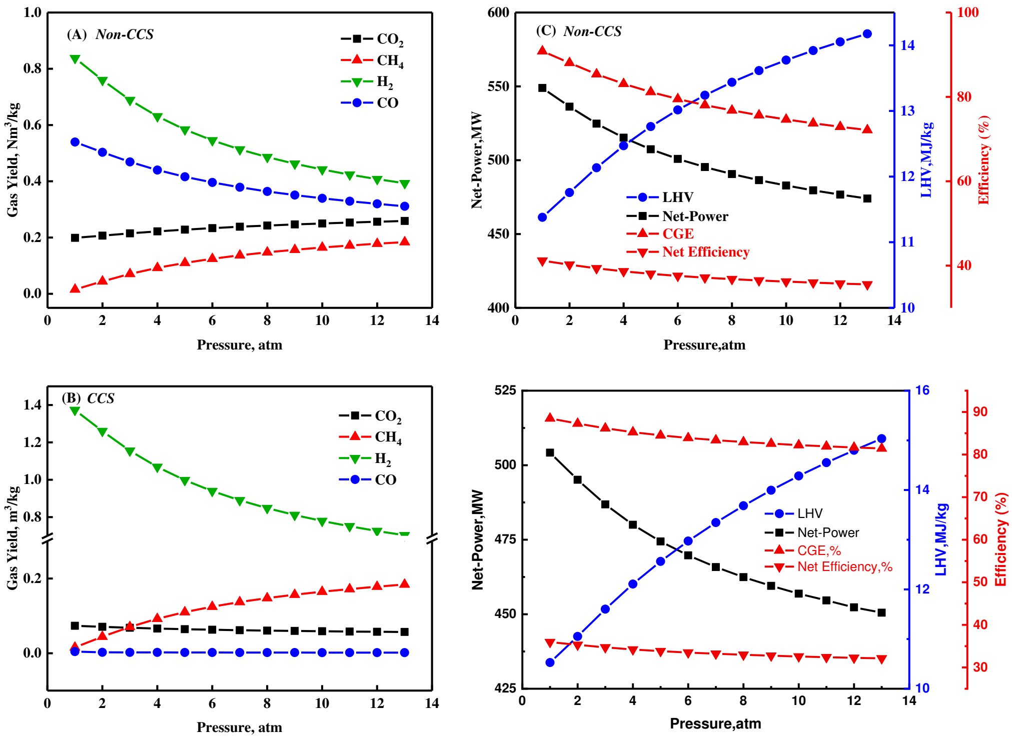


Figure 5. Effect of pressure on both CCS and Non-CCS power plants

Figure 5A illustrates that an increase in pressure shifts equilibrium towards the side with less gaseous moles. For the non capture plant, H_2 and CO flowrates decreased while CO_2 and CH_4 increased with an increase in pressure. The main reason is that the gasification reactions (reactions 2, 3 and 4) in Table 2 are restrained under high pressure, thus resulting in a decrease in H_2 and CO decrease as pressrue increases. However, a high pressure is conductive to generating CH_4 according to reaction 5 while suppressing CH_4 conversion (reaction 10). This consequently increased LHV from 11.4 MJ/Nm^3 to approximately 14 MJ/Nm^3, but reduced the gas yield therefore reducing CGE, net power and consequently net efficincy (from 41.2% to 35.6%), as demonstarted in Figure 5C. The capture plant, as shown if Figures 5B & D showed similar trends however experiencing a higher H_2 flowrate and a relatively constant CO and

CO_2 flowrate. The net efficiency of non capture plant decreases by approximately 6% compared to capture plant which decreases by 4% as pressure increases from 1 to 13 atm.

5.1.4 Effect of Steam

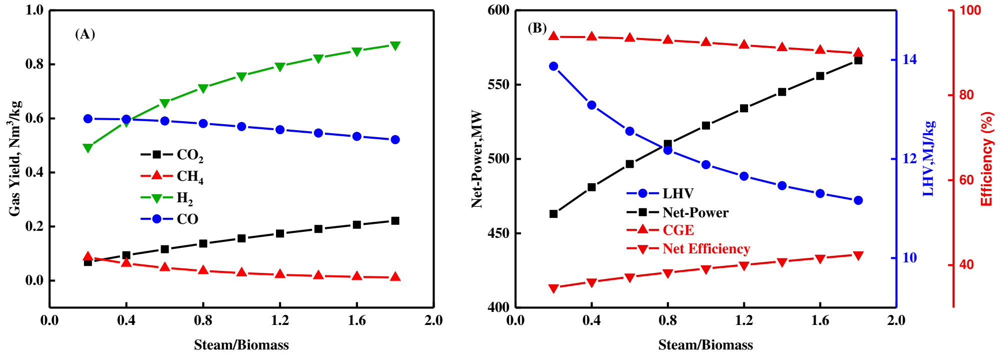


Figure 6. Effect of steam/biomss on both CCS and Non-CCS power plants

Figure 6A showed that an increase in S/B ratio resulted in an increase in H_2 and CO_2 but a decrease in CO and CH_4 yields. This can be explained according to the equilibrium shift of reactions 1, 2 and 12 (Table 2) shifting towards the right-hand side, therefore increasing the CO_2 and H_2 concentrations. As shown in Figure 6B, the LHV decreases, since methane (has a high LHV) is consumed, to produce CO and H_2 which have a low LHV. This decrease in LHV results in the CGE also decreasing. However, increasing S/B ratio increases the combustible gas yield which increases the net-power generated, consequently increasing the net efficiency. The optimum S/B ratio was taken as 1.5 since net efficiency started to plateau. This has only been tested for the Non-CCS power plant; however, the following section discusses the effect of WGS on the CCS power plant.

In order to investigate the synergistic effects of the above key factors on the system performance, we divided the factors into two categories, operation parameters (gasification temperature and pressure) and technological conditions (OC/B and S/B). The effects of them on energy efficiency of plants were presented in Fig. 7S in the **Supporting Information.** Temperature seems to have a higher impact on energy efficiency compared to pressure within the range that has been tested. Temperature results in a steeper increase in energy efficiency up to 875°C, whereas pressure has a gradual decrease in energy efficiency. Therefore, changing temperature would be more effective than changing pressure, high temperature and low pressure are beneficial to improving plant energy efficiency. Steam inputted into FR can obtain more H_2 and CO, increasing the effective composition of fuel gas thus enhancing system energy efficiency, however, OC provides enough heat for promoting steam reacting with other reactants, but exceeded OC will consume H_2 and CO via combustion reactions. Change in OC/B ratio resulted in a steep increase then a sharp decrease after reaching a peak at an OC/B ratio equal to 2. SO as to achieve the maximum energy efficiency of the plant , the OC/B ratio should firstly satisfy the reactor self-heating requirements, followed by a suitable ratio of OC/B to S/B to obtain the maximum energy efficiency of the plant.

5.1.5 Carbon Capture

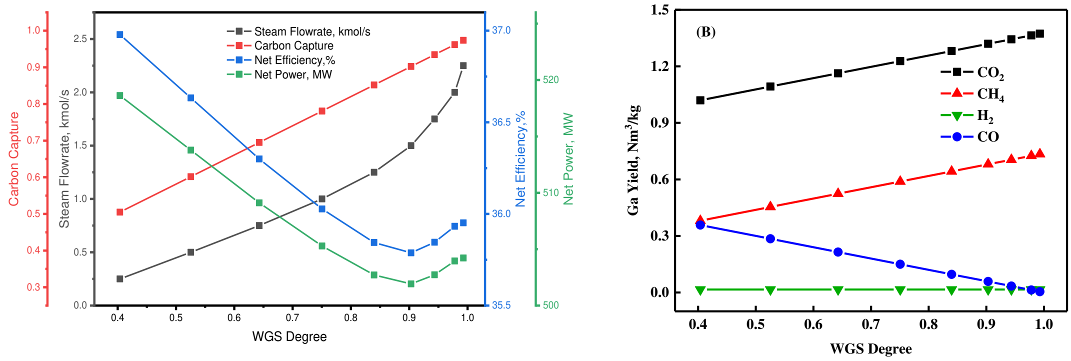


Figure 7. Effect of WGS degree on the CCS power plant

Figures 7A and B outlines the results of testing the effect of water gas shift. This is done by injecting steam in a Water Gas Shift reactor, where it reacts with the syngas, converting CO into CO_2 and H_2O into H_2. This allows for the CO_2 to be captured before the syngas combustion process. As steam flowrate increases, the degree of water gas shift increases (more CO converting into CO_2), increasing amount of carbon captured, consequently increasing the power consumption during the carbon capture process. As a result, the net efficiency of the power plant decreases from 37% to approximately 36%. This can be observed when steam flowrate is equal to 2.25 kmol/s steam flowrate (WGS Degree = 0.993 and Carbon Capture Ratio = 0.974). Further increase in steam does not have any effect on the WGS degree or carbon capture, hence was taken as the value for CCS tests. Even though an increase in carbon capture would reduce efficiency of the power plant it will support negative emissions technology especially with biomass being the source of fuel.

5.2 Economic assessment

5.2.1 Cost distribution

Based on the techno-economic analysis, FR temperate = 875°C, FR pressure = 1 atm, OC to Biomass ratio = 2, steam to biomass ratio = 1.5 and 0.993 WGS degree were taken as the optimum conditions to be used when conducting the economic analysis. It is noted that the above optimized conditions obtained in this paper are based on the thermodynamics analysis, hence, if we just consider the thermodynamics performance of the plant, these values can be used as general conditions and are suitable for all BCLGCC (w/o CCS) plant with different sizes. However, the economic impact values may change for a different plant size. The change in operation parameters and conditions will lead to variation in heat flow, mass flow and gas composition, thus affecting the device size as well as its investment. Since the economic performance are significantly influenced by the plant scale, those values are taken as optimum for a 650 MW (gross power) power plant (biomass input = 6600 ton/day (non-ccs) and 6480 ton/day (ccs)) in this study and not for all sizes. The economic analysis was based on the assumption that biomass price is equal to 12.4 $/GJ (£10), negative emissions incentive is equal to zero and the power plant is available 80% of the time. The amount of OC required in the process was increased by 10% to account for OC escaping the reactors with the syngas/flue gas. Table 4S presents the breakdown cost in calculating the TOC, variable and fixed costs. Based on the data, the power plant with capture increased the capital cost by 30%, fixed cost by 20% and variable cost 1% compared to non-capture.

5.2.2 Effect of plant size

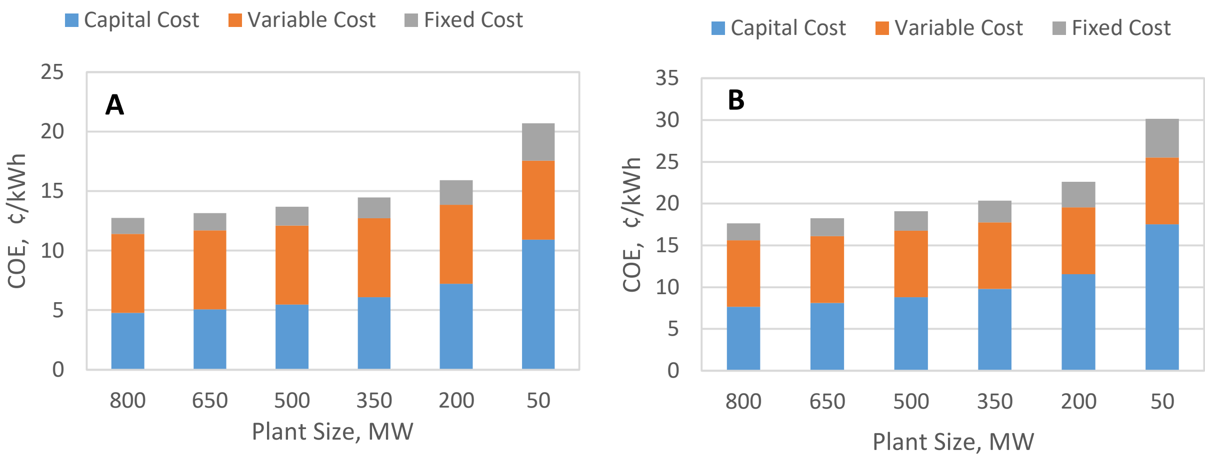


Figure 8. Effect of Non-CCS (A) and CCS (B) plant size on the COE

Figures 8A and B show the effect of COE including the breakdown contribution of capital, variable and fixed costs for both CCS and Non-CCS plants, respectively. The COE of the 650MW power plant was calculated using Aspen Plus data and using Equation 15, however the costs for the different sizes were scaled using Equations 12, 13 and 14. The scaling of the capital and fixed costs was exponential; however, the scaling of the variable cost was proportional. The graphs followed an expected trend of decrease in COE with increase in plant size. However, the graphs reached a plateau at around 500 MW, further increase in plant size did not have significant impact on the COE.

5.2.3 Sensitivity analysis to COE

A sensitivity analysis was conducted to test the effect of different variables on the COE of both, CCS and Non-CCS plants.

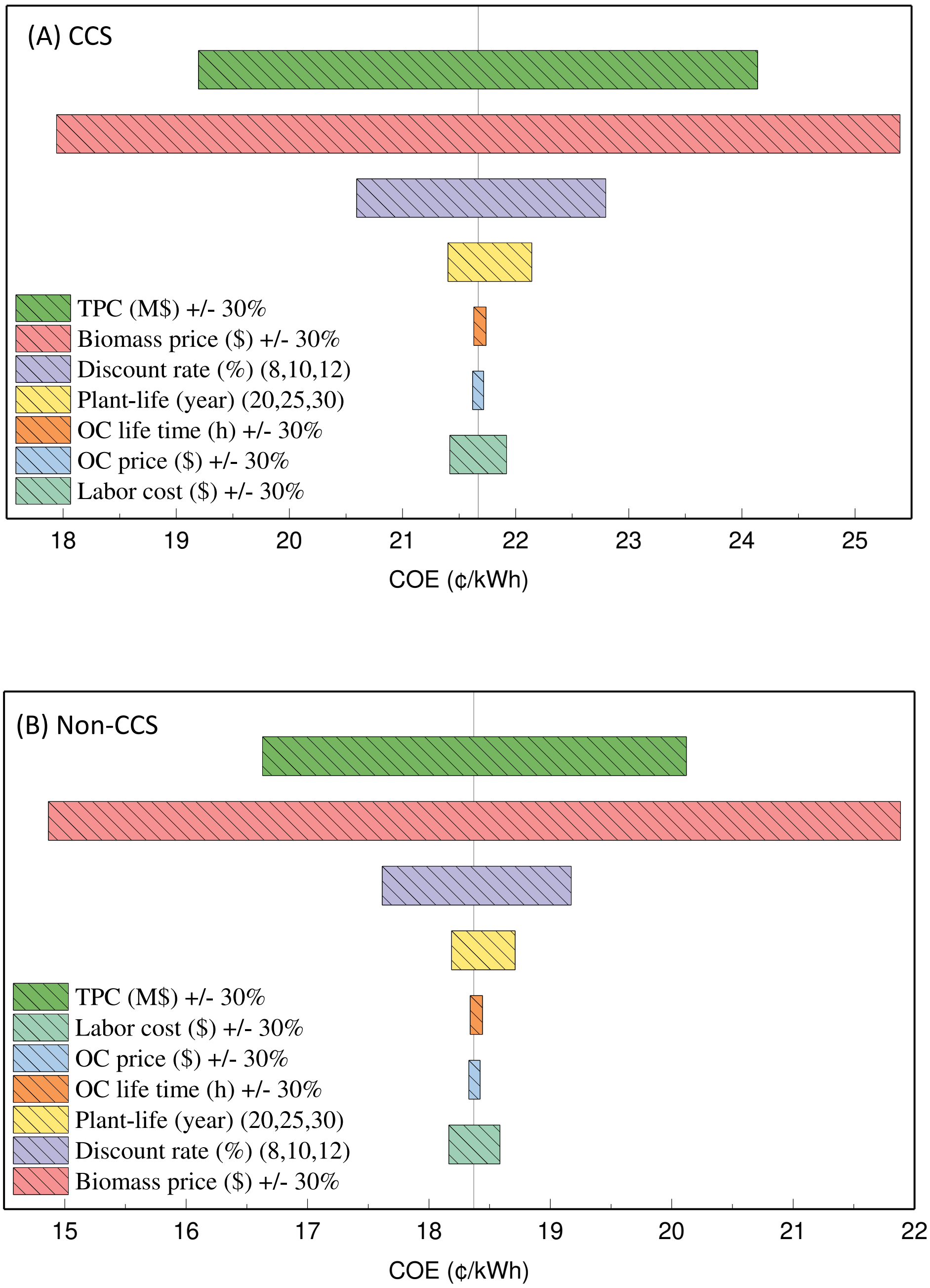


Figure 9. Sensitvity analysis of different variable for both CCS (A) & Non-CCS (B)

Figures 9A and B represent the effect of TPC, Biomass price, discount rate, plant life, labour cost, OC price and lifetime on both CCS and Non-CCS power plants, respectively. For CCS, it can be observed that if TPC is varied by +/- 30% the COE (21.7 ¢/kWh) will vary between 24.1 – 19.2 ¢/kWh (11.3% difference in COE), whereas for Non-CCS the COE (18.4 ¢/kWh) will vary between 20.1 – 16.6 ¢/kWh (9.5% change in COE). The bigger gap between both values shows that varying the TPC has a greater effect on the COE in a CCS compared to a Non-CCS power plant, which is due to the additional cost of the carbon capture unit. Moreover, since this type of power plant has not been constructed on large scale before, its project and process contingencies are quite higher, hence increasing its capital cost and COE. The price and lifetime of the OC seemed to have a very negligible effect on the COE for both CCS and Non-CCS due to the low price of hematite (95 $/ton). Labour cost had a slight effect on the COE, but a greater effect was observed when the plant life increases from 20 to 25 years (increased by 0.27 ¢/kWh and 0.19 ¢/kWh for both CCS and Non-CCS). Discount rate seems to be the third most effective variable on the COE after TPC and biomass price. Regarding the effect of biomass price (12.4 $/GJ), if we vary its price by +/- 30%, COE will range from 17.9 – 25.4 ¢/kWh and 14.9 – 21.9 ¢/kWh for CCS and Non-CCS, respectively.

It can also be observed that the price of biomass had the highest effect on the COE for both CCS and Non-CCS plants followed by the TPC. This shows the key role biomass price plays in influencing the economic feasibility of the entire process. In the UK, biomass is generally imported from the US and Canada [39], therefore increasing its price due to shipping, however this process could be more feasible in geographic locations where biomass is abundant and cheap. Nevertheless, the main current financial support scheme presented by the UK government for renewable electricity is the Renewable Obligation Certificate (ROC) [40]. The ROC's current value for generating electricity using 100% biomass is 4.84 p/kWh (5.8 ¢/kWh). This would therefore reduce the COE for both CCS and Non-CCS to 15.9 ¢/kWh and 12.8 ¢/kWh, respectively, which reduces the costs below the average cost of electricity in the UK (17.7 ¢/kWh (14.37 p/kWh) [42]). From a policies perspective, further incentives can be introduced for negative emissions which could drive the commercialization of BECCS technology. The graphs showing the effect of varying each of the aforementioned variables on the COE can be found in the **Supporting Information** (Figure 5S).

Figure 10A estimated the effect of increasing negative emissions incentive and its effect on the COE for both CCS and Non-CCS. It had no effect on the Non-CCS plant as expected, however it would have an effect on the CCS plant since capture of CO_2 will result in negative emissions. The COE including the ROC government subsidies for CCS (15.9 ¢/kWh) is already lower than the average COE in the UK (approximately 17.7 ¢/kWh). Introducing negative emissions incentive of no more than 39 \$/t-CO_2 in addition to renewable government subsidies can further reduce the COE to 12.8 ¢/kWh (which is the same as the Non-CCS plant). Figure 10B shows the effect of the capacity factor on the COE. It can be observed that the CF has more effect on the CCS plant compared to the Non-CCS plant, with a difference of 7.16 ¢/kWh and 5.12 ¢/kWh between 0.5 and 1 capacity factor, respectively. Figure 10C illustrates that the COE increases as the WGS degree increases, converting more CO into CO_2. This is due to more energy being consumed for CO_2 capture, as a result reducing the plant's net power. The COE increases from 20.9 ¢/kWh to 21.7 ¢/kWh as the WGS degree increases from 0.4 to 0.99, however, the cost of CO_2 capture decreases from 64.6 \$/t-CO_2 to 342.7 \$/t-CO_2. Table 5S compares the cost and performance of the 3 main biomass-based technologies, including; biomass combustion, biomass integrated gasification combined cycle (BIGCC) and BCLGCC. Coal direct combustion and coal integrated gasification combined cycle technologies are also introduced and compared. From previous literature data regarding the capital cost, COE and efficiency of both capture and non-capture plants were collected and compared for an estimated gross 650 MW plant size. The values in the Table 5S do not include any government subsidies or renewable energy incentive. The results showed that BCLGCC shows promising net electric efficiency and COE compared conventional power generation technology (biomass combustion and BIGCC).

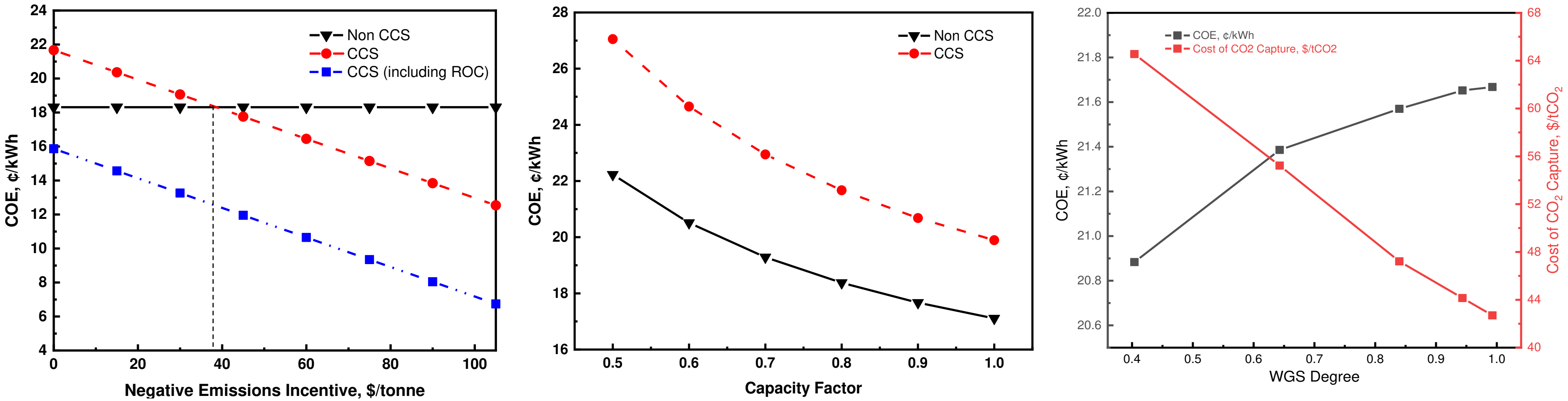


Figure 10. Effect of negative emissions incentive (A), capacity factor (B), WGS degree and cost of CO_2 capture (C) on COE

5.2.4 Sustainability analysis

The previously mentioned indicators (economic, social, environmental and technical indicators) in section 4.2 and sub-indicators for the sustainability assessment is presented in Table 4. The values for each indicator, including the reference point (worst and best case for each indicator) were either taken from literature [36, 38, 41, 43] or are calculated for the worst possible value based on the fuel feedstock properties. The results are presented in a graphical form in Figures 11, for the CCS and non-CCS plants, respectively.

Table 4. Sustainability performance assessment between 5 power generation technologies

Indicator (subindicator)	IGCC -Biomass		BDC		BCLGCC		CFP		IGCC – Coal		Reference Value	
CCS/Non-CCS	CCS	Non-CCS	CCS	Non-CCS	CCS	Non-CCS	CCS	Non-CCS	CCS	Non-CCS	Best	Worst
Economic												
Unit capital cost, M$/MW	4.0	2.8	3.5	1.6	3.7	2.6	2.9	1.4	2.8	2.0	0	5
COE, ¢/kWh	23	16	19	14	21	18	13	7	12	8	0	25
Environmental												
Net Electric Efficiency, %	31	36	26	35	36	41	28	36	32	39	100	0
Renewability, %	100	100	100	100	100	100	0	0	0	0	100	0
Water consumption, kg/kWh	3.50	3.02	2.30	1.71	2.80	2.40	4.9	3.83	3.25	2.4	0	28.4
CO_2 emissions, g/kWh	0	0	0	0	0	0	114	905	103	821	0	908
SO_2 emissions, g/kWh	2.6	1.9	2.2	1.6	1.9	1.7	8.0	6.2	2.1	1.7	0	25
NO_X emissions, g/kWh	1.7	1.5	2.4	1.8	1.5	1.4	4.2	3.3	1.2	0.95	0	59
Dust emissions, g/kWh	0.034	0.030	0.081	0.060	0.030	0.026	2.2	1.7	0.10	0.08	0	93.6*
Social												
Community Development, staff/$MW_{(e)}$	11	9	10	8	12	10	1	0.5	1.5	1	20	0
Energy Security, %	0	0.05	0**	5	0	0	0	3	0	0	100	0
Technical												
Technical Maturity	0.5	0.75	0.75	1	0.5	0.5	0.75	1	0.75	1	1	0

*All ash in the fuel is assumed to be dust

** Will be operating in early 2020's[44]

Economic sustainability

Comparing between the economic performance of the 5 different power generation technologies (with and without capture), it can be implied that gasification (BIGCC, BCLGCC and CIGCC) require a higher capital cost per unit power as well as higher COE due to the complexity of the system, except for BCLGCC which demonstrates a lower COE which is due to its higher efficiency compared to the other processes. Additionally, we can see that coal power plants have a slightly lower capital investment per unit energy compared to biomass power plants, however a significantly lower COE due to the lower cost and higher heating value of coal compared to biomass. Moreover, CCS technology increased the capital cost of the power plants on average by approximately 1.1 M$/MW, which then resulted in an increase in COE by approximately 4 - 5 ¢/kWh.

Environmental sustainability

Biomass fuelled power plants showed lower efficiencies compared to coal fired power plants, except for BCLGCC which demonstrated similar efficiencies for CCS and non-CCS plants (36% and 41%, respectively) in comparison to coal fuelled IGCC. Biomass IGCC requires higher water consumption compared to BDC for tar scrubbing. BCLGCC requires less water due to the catalytic cracking of the tar by the oxygen carrier. In terms of renewability, the biomass-based power plants are 100% renewable since biomass is a renewable source of energy, whereas for coal-based power plants the renewability is zero. The CO_2 emissions from the biomass-based power plants is assumed to be equal to zero due to biomass being carbon neutral, whereas CFP and coal IGCC released 905 and 821 g/kWh, respectively, which is due to the higher efficiency of IGCC. The CCS process is assumed to capture approximately 90% of the CCS, hence resulting in 114 and 103 g/kWh being released, respectively. Biomass-based power plants technically should have negative emissions as they are removing CO_2 from the atmosphere, but in this analysis, it is assumed to be zero.

Social sustainability

Developing biomass-based power plants can increase the number of jobs by 8 - 12/ MWe, since more agricultural residue will be required as fuel. Therefore, requiring the agricultural industry to restructure, hence creating more jobs. Whereas, coal-based power plants provide much less jobs (0.5 - 1.5 jobs/MWe). However, sourcing the biomass from abroad would improve the job market for the location where the biomass is sourced. Take the UK as an example, however, most of the biomass in the UK is imported from the US and Canada, hence will not provide as many jobs within the UK. The shift towards biomass is to move towards renewable sources of energy while ensuring energy security. Currently the energy security from coal power plants is low (approx. 3%), while biomass power plants is relatively higher (6%), and zero energy security from CCS power plants. The UK parliament recently passing a bill to reach net-zero emissions by 2050, therefore biomass energy sources will see a greater push and an increase in its use in the following decades. Drax power plant will be the first biomass power plant in the UK to be integrated with CCS technology [44]. With BCLGCC technology utilizing biomass efficiently in power generation while providing a lower COE, even with CCS technology, we could see an increase in its energy security in the following decades.

Technical Sustainability

This indicator measures the maturity level of the technology for it to achieve a specified function and to demonstrate whether the technology is commercially feasible. The specified power generation technologies in Table 4 shows that combustion technologies are the most mature due to their simple process compared to gasification, especially with coal power generation technology being more mature compared to biomass fuelled technology. Biomass gasification process to power is slightly complicated compared to that of coal. BCLGCC has only been demonstrated as pilot plants, hence is still under research stage, therefore reducing into maturity and commercial reliability. CCS technology have been demonstrated; however, the technologies maturity is not as mature relative to Non-CCS processes.

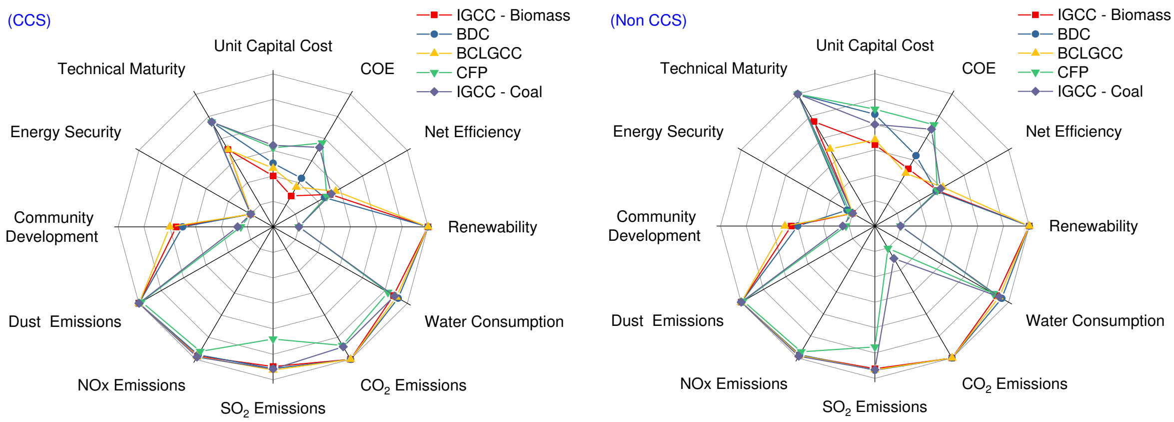


Figure 11. Sustainability evaluation of biomass and coal power generation processes

6. Conclusion

BCLGCC proves to be a novel and effective power generation process. This paper establishes a reliable simulation process of biomass chemical looping gasification combined cycle to power plant, and the key technical parameters are optimized to obtain the maximum electricity output. Subsequently, a comprehensive study of the technical and economic feasibility of an industrial scale (650 MW gross power) BCLGCC power plant while taking into consideration the sustainability impact of the power plant relative to current power generation technologies was conducted. Both CCS and Non-CCS processes were modelled and evaluated, giving a net efficiency of approximately 36% and 41%, respectively. These values are higher compared to conventional coal/biomass combustion and IGCC processes. The high efficiencies of these processes are associated with the costs, hence a lower COE of 21.7 ¢/kWh and 18.4 ¢/kWh for CCS and Non-CCS processes, respectively. Taking into consideration the

UK's government renewable energy subsides, further decrease of the COE to 15.9 ¢/kWh and 12.8 ¢/kWh for CCS and Non-CCS power plants, respectively, could be achieved. Additionally, if negative emissions incentives are also introduced, the economic feasibility of the power plant with CCS will be more obvious. When comparing the techno - economic performance of BCLGCC with other power generation technology, it can be seen that BCLGCC demonstrates higher net efficiency with a lower COE. A sustainability assessment conducted comparing between 5 different power generation technologies (with and w/o CCS) demonstrates that BCLGCC presents promising economic and environmental results, with an increase in community development, but a low energy security due to the process not being commercially established, as a result it is still not as technically mature as the other power generation technologies. However, its promising results, especially with the UK heading towards a net-zero emissions by 2050, BECCS technologies will become a vital option and will play a big role to achieve this target.

Acknowledgements

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Highlights

- A reliable BCLGCC simulation process at 650 MW is established and validated
- The entire BCLGCC process is optimized to obtain the optimal power output
- BCLGCC with and w/o CCS present a net efficiency of approximately 36% and 41%
- BCLGCC with and w/o CCS show a lower COE of 18.4 ¢/kWh and 21.7 ¢/kWh
- Sustainability of BCLGCC is conducted and compared to other power plants

Supporting Information

Sustainability Evaluation of Biomass Direct Gasification Using Chemical Looping Technology for Power Generation with and w/o CO_2 Capture

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Description of the key units

- **Gasification (FR & AR)**

Biomass is initially injected into the FR where it undergoes devolatilization, pyrolysis, combustion and gasification reactions in the presence of an iron-based oxygen carrier, providing lattice oxygen for the combustion/gasification reactions to take place. The depleted oxygen carrier passes through the lower loop seal into the AR where it is regenerated by reacting with molecular oxygen from air. The regenerated metal oxide is separated from the flue gas via a cyclone and sent back to the FR after passing through the upper loop seal. The two reactors are electrically heated in an oven which supplies heat for start-up and compensates heat loss during the operation. Thermocouples and differential pressure transducers were located at different points of the prototype to display the operating conditions and monitor the cycling stability of the fluidized bed in real time. The outlet gases from the air reactor and fuel reactor were induced with suction pump to an ice−water cooler where the steam was condensed and removed. The produced gases were collected with gas bags and analyzed by an offline gas chromatograph.

- **Water gas shift and acid gas treatment**

The syngas produced contains high amount of carbon and sulphur components. However, for this process to be carbon neutral and prevent sulphur emissions, they should be captured pre-combustion. Therefore, a Water Gas Shift (WGS) unit is added where syngas reacts with high temperature steam in a reactor, shifting the reacting towards producing more CO_2 and H_2. This is also a H_2S hydrolysis unit where the H_2S is converted into COS. Since the reaction taking places in the reactor is exothermic. The shifted syngas is cooled followed by a selexol acid gas treatment section, capturing 90% CO_2 and approximately 95 - 98% COS.

- **Combined cycle**

Gas turbines have been commonly used in power generation processes. A conventional natural gas fired gas turbine with a simple cycle generally has an efficiency of 35%. However, new power plants enhance the process with an additional heat recovery steam generator (HRSG) block followed by a steam turbine, which is known as a combined cycle. The reasoning behind that is for the heat in the exhaust gas to be captured via the HRSG process, converting feed water into steam which is sent to a steam turbine for power generation. A gas turbine (simple cycle) coupled with a HRSG and steam turbine. The heat recovered from the exhaust gas is used to generate high pressure steam which then passes through the steam turbine, dropping the pressure and temperature of steam, converting heat into shaft work to generate electricity. The steam turbine is divided into 3 stages; high-pressure steam, intermediate-pressure steam and low-pressure steam. This is due to the high pressure of steam which will result in large expansion if pressure is reduced all at once.

Experimental results

- **Stability of reaction system**. From the Figure 2, P1 and P2 are the points were pressure was measured in the AR while points P3 and P4 are measured in the FR. The typical bed pressures throughout the reaction time are displayed on Figure 1S. The pressures at points P1, P2, P3 and P4 remain relatively consistent and fluctuate around 0.56, 0.45, 1.05 and 0.18, respectively. This indicates that the system is in a stable operating condition and that the OC have been circulating from the AR to FR throughout the process.

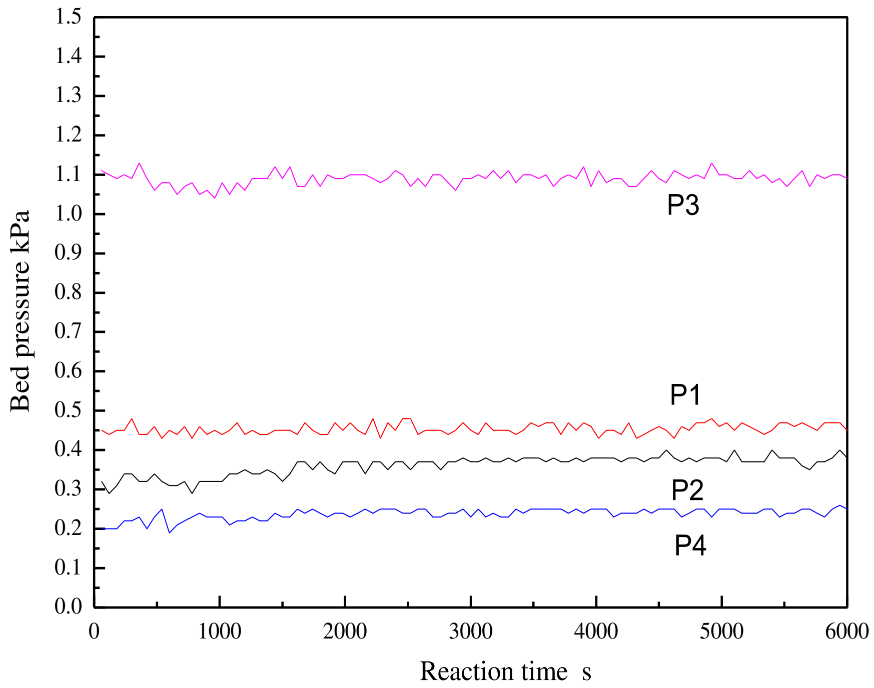


Figure 1S. Bed pressure change with reaction time

- **XRD patterns of oxygen carriers.** Experiments have been conducted on fresh, reduced and regenerated oxygen carrier samples, in which they were examined using X-ray diffraction to determine the crystalline phases formed on each sample. It was detected that the original crystal form of fresh oxygen carrier primarily constitutes of Fe_2O_3, SiO_2 and other metallic oxidizes (Al_2O_3, MgO, CaO etc.). Since the Fe_2O_3 plays a key role in the reaction process as lattice oxygen transfer medium and the SiO_2 in hematite oxygen carrier also has positive effects in preventing the aggregation or sintering for oxygen carrier particles. Therefore, in order to further highlight evolution of oxygen carrier crystal form in the reaction process, the diffraction peaks of iron oxides and SiO_2 are marked in the XRD spectra and the other impurities oxides in hematite are too little to be ignored. The crystal form of fresh hematite oxygen carrier characterized by XRD is shown in Figure 2S. Powder X-ray diffraction (XRD, X'Pert PRO MPD) using Cu Kα (40 kV, 40 mA), was used to analyse the crystal structure of fresh and reacted samples. The samples was scanned at a rate of 2° min^{-1} between 2θ = 10°−90° with a step of 0.0167°. The samples were degassed under vacuum at 493 K for 6h before measurement. The reduced samples and oxidized samples were collected from the fuel reactor and air reactor separately after 60 h of operation. There were three crystalline phases of Fe_2O_3, Fe_3O_4, and FeO in the reduced samples. Fe_3O_4 and FeO phase were not found in the fresh or regenerated samples of the fuel reactor, which manifested that Fe_2O_3 was mostly reduced to Fe_3O_4 and FeO. The results suggested that the oxygen carrier could return into its original form in the AR after it has been reduced in the FR.

- **Equilibrium composition of gasified gas.** Figure 3S showed the equilibrium composition of gasified gas and carbon conversion rate changes over reaction time and number of cycles. It was clear that the reaction system (fuel reactor) reached thermodynamic equilibrium after 20 hrs, and the gas composition and carbon conversion rate maintained a stable status.

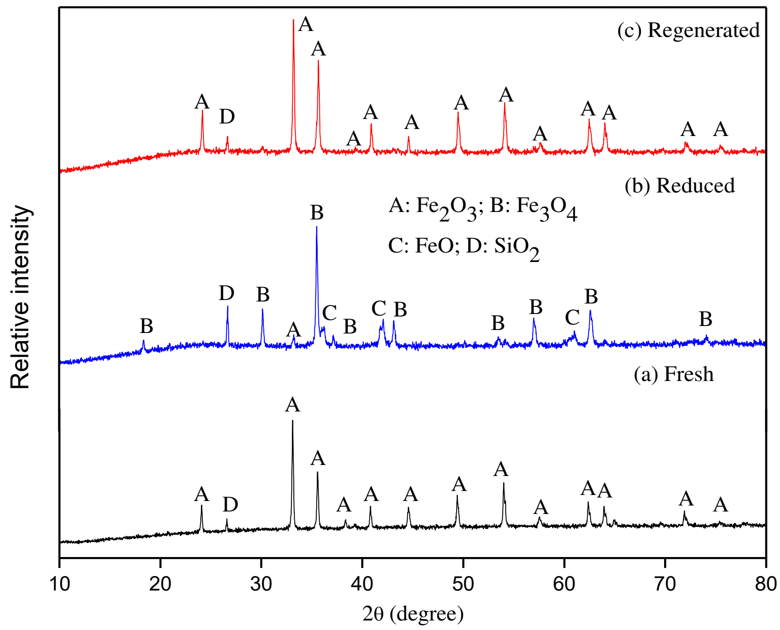


Figure 2S. XRD patterns of fresh and used oxygen carrier

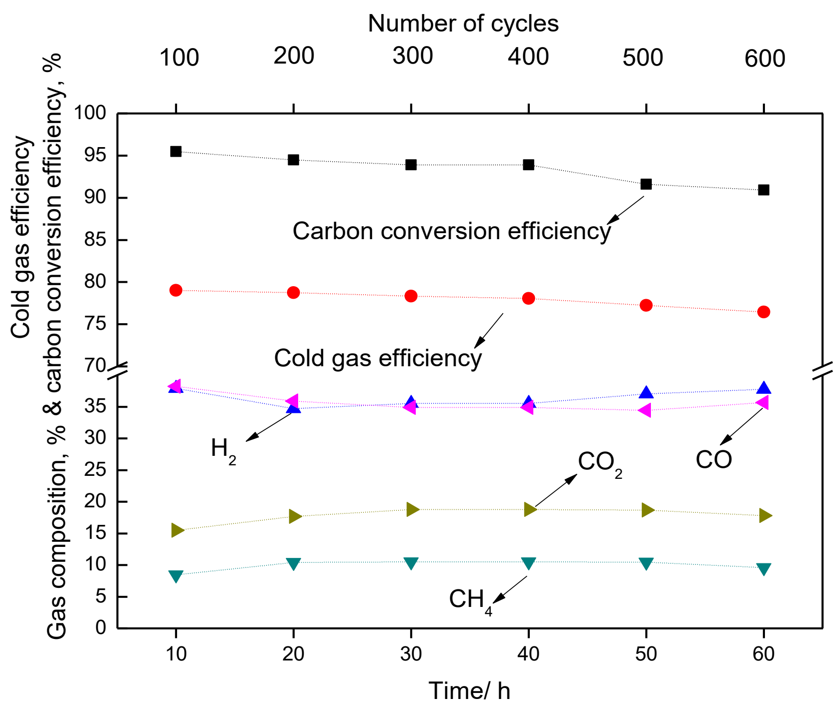
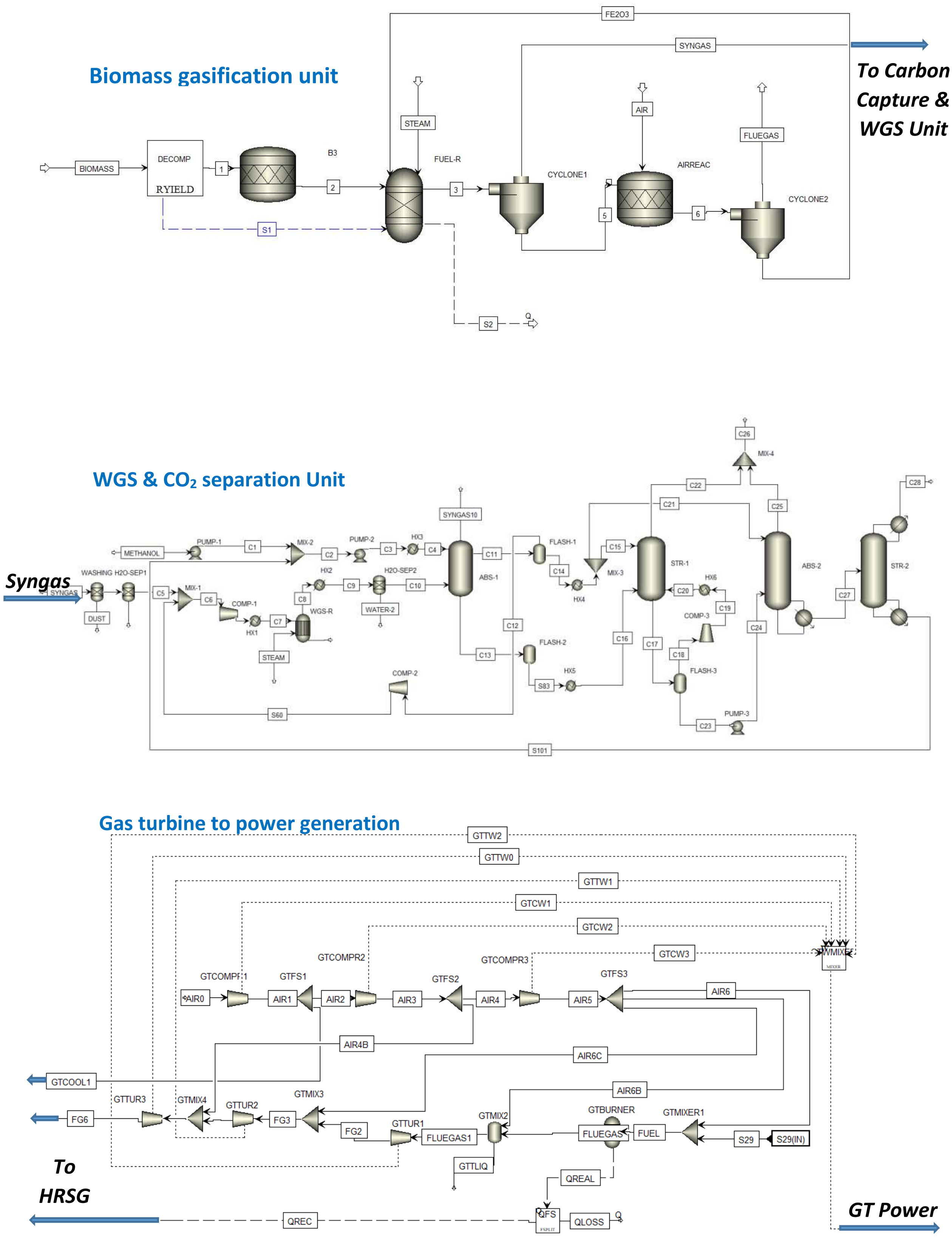


Figure 3S. Gas composition, carbon conversion and cold gas efficiency as a function of reaction time and number of cycles



Biomass gasification unit
FE2O3
SYNGAS
To Carbon Capture & WGS Unit
STEAM
AIR
FLUEGAS
BIOMASS
DECOMP
RYIELD
B3
FUEL-R
CYCLONE1
AIRREAC
CYCLONE2
S1
S2
WGS & CO₂ separation Unit
Syngas
SYNGAS10
METHANOL
PUMP-1
MIX-2
PUMP-2
HX3
ABS-1
FLASH-1
MIX-3
STR-1
ABS-2
STR-2
MIX-4
WASHING
H2O-SEP1
DUST
MIX-1
COMP-1
HX1
HX2
WGS-R
H2O-SEP2
WATER-2
COMP-2
FLASH-2
HX4
HX5
HX6
COMP-3
FLASH-3
PUMP-3
S60
S83
S101
Gas turbine to power generation
GTTW2
GTTW0
GTTW1
GTCW1
GTCW2
GTCW3
GTCOMPR2
GTCOMPR3
GTFS1
GTFS2
GTFS3
AIR0
AIR1
AIR2
AIR3
AIR4
AIR5
AIR6
AIR4B
AIR6C
AIR6B
GTCOOL1
FG6
GTTUR3
GTMIX4
GTTUR2
FG3
GTMIX3
FG2
GTTUR1
FLUEGAS1
GTMIX2
GTBURNER
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GTMIXER1
S29
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QREAL
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QFS
QLOSS
To HRSG
GT Power

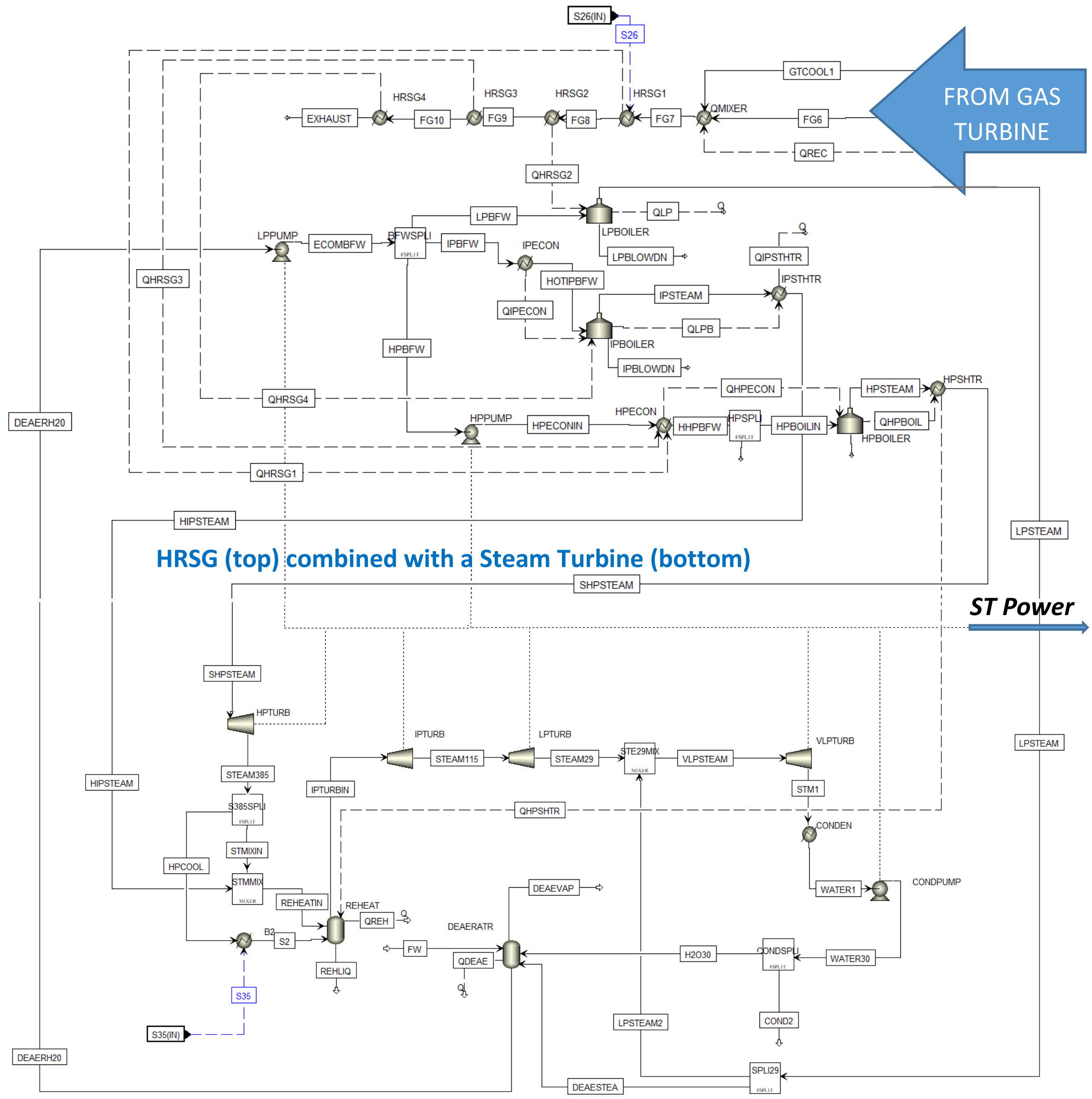

Figure 4S. The simulation flowsheet of biomass chemical looping gasification to power generation

Process simulation

The simulation process of biomass chemical looping gasification to power generation is shown in Figure 4S. An additional water gas shift reactor followed by combined with a CO_2 capture unit process has also been added for tests to be compared between BCLG with and without CO_2 capture. Both processes (CCS and Non-CCS) were then coupled with a combined cycle for electricity generation.

Table 1S Description of the operation blocks

Aspen plus block	Block ID	Description
RYield	DECOMP	Yield reactor – Based on mass balance this reactor converts the nonconventional biomass into conventional compounds, by decomposing biomass into constituent elements
RStoic	SLAGFORM	Stoichiometric Reactor - Reacts a percentage of unburnt carbon with the ash to form slag
RGibbs	FUEL-R	Gibbs free-energy reactor – Simulates the gasification reactions by using the direct minimization of Gibbs free energy to determine the equilibrium composition
Cyclone	B1	Cyclone – Separates ash from syngas by specifying split fraction
RStoic	AIRREAC	Stoichiometric Reactor - Air regenerates the oxygen depleted oxygen carrier. Moreover, air reacts with the unburnt carbon.
Cyclone	B2	Cyclone – Separates the oxygen carrier from the flue gas (Mainly N_2 and CO_2)

Table 2S Comparison of experimental and simulation results for BCLGCC

Variables	Conditions		Variables	Results	
	Literature	Simulation		Experimental results (vol. %)	Simulation results (vol. %)
Fuel reactor temperature, °C	800	800	H_2	34.36	39.22
Fuel reactor pressure, atm	1	1	CH_4	9.98	10.92
Steam/biomass ratio	0.85	0.85	CO	38.02	34.25
Oxygen carrier/biomass	5/3	5/3	CO_2	17.64	15.61
Carbon conversion efficiency, %	93	93	LHV (MJ/Nm^3, dry basis)	12.08	12.48
Flow rate of syngas, ton/hr	237.3 [2]	237.3	Gas Yield (Nm^3/kg, dry basis)	1.41	1.32
Flowrate of air, ton/hr	1605.3 [2]	1605.3			
Exhaust Temperature, °C	601.1 [2]	603.9			
Power Output, MW	319.6 [2]	310.1			

Table 3S. Parameters for scaling plant cost

Unit	Scaling Parameter	Reference cost (M$)	Reference scale	Scaling parameter	Reference
Gasification Units	Biomass flowrate, tonnes/hr	68.4	74.13	0.8*	[3]*/ [4]
WGS Reactor	H_2 + CO flowrate, kmol/s	21.33	2.45	0.65	[5]
CO_2 Absorption	Syngas flowrate, kmol/s	46.47	3.99	0.7	[5]
Gas Turbine	Power output, MW	28.57	266	0.75	[6]
HRSG	Heat Exchange, MW	23.13	355	1	[6]
Steam Turbine	ST gross power, MW	25.85	275	0.67	[6]
CO_2 Drying and Compression	CO_2 flowrate, kmol/s	55.39	4.18	1	[5]

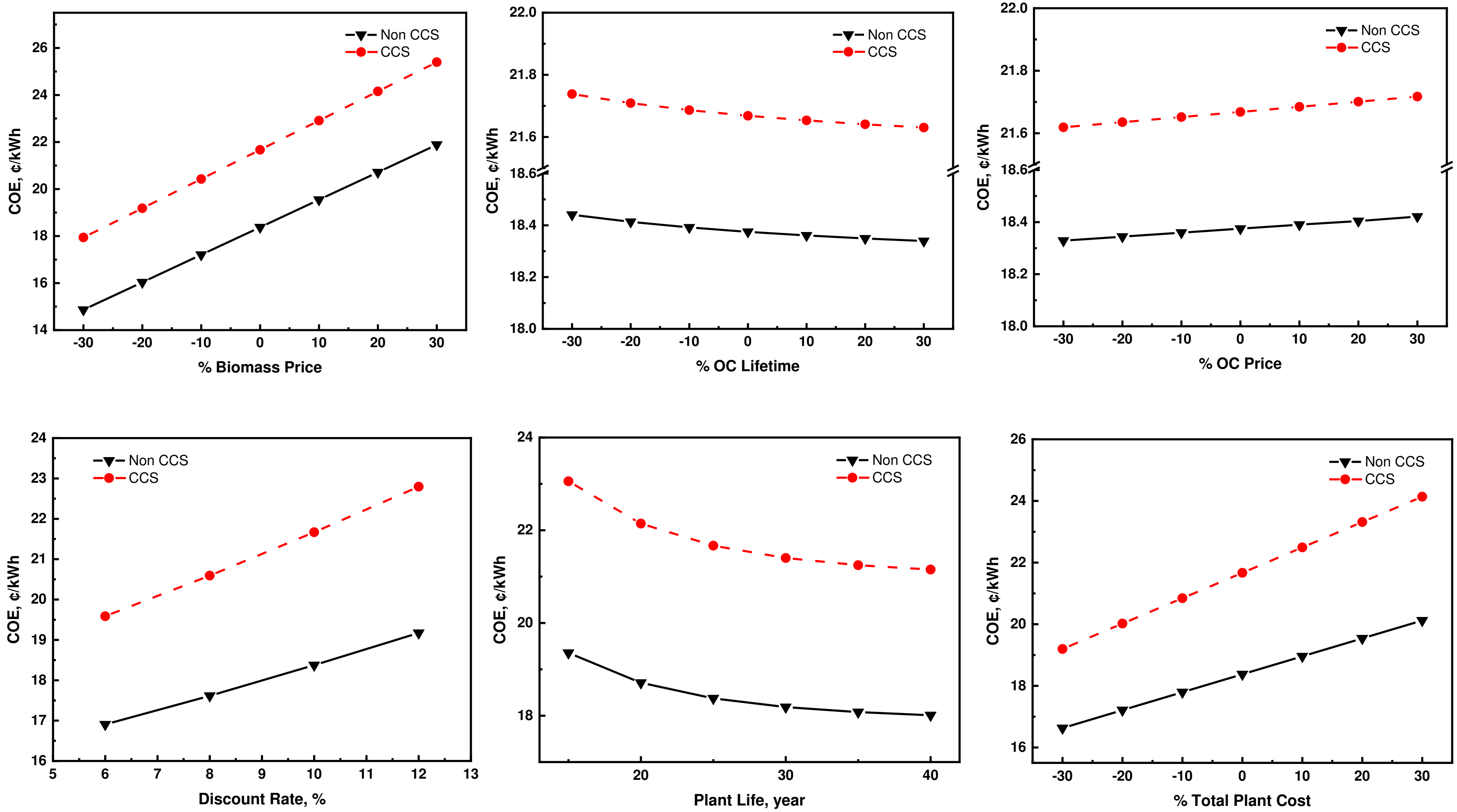

Figure 5S Effects of some key economic variables on COE

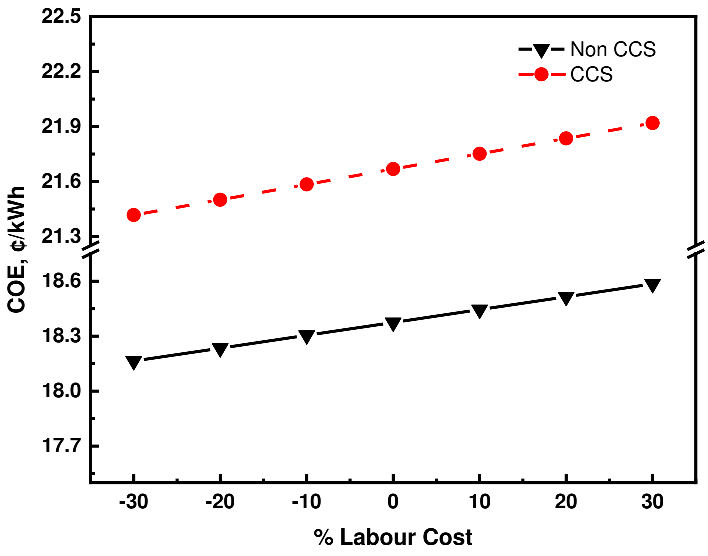

Figure 5S. *(continued)*

Indicators for sustainable assessment

- Economic indicator consists of the investment and production cost.

Investment cost: It is the capital investment cost of the power plant per unit capacity used to compare between different power plants.

Production cost (COE): The production cost of converting fuel (biomass or coal) into electricity, hence represented as the cost of electricity (¢/kWh).

The values for the economic comparison was conducted between 10 different power generation processes (Table 7S) based on previous literature, and equations 12 – 15 were used to scale up plant sizes to 650 MW (gross power) in estimating COE [4,5,7–13, 15].

- Environmental indicators. Power generation processes requires the consumption of raw materials and energy, as well as releasing waste into the environment. This results in resource depletion and environmental degradation. The environmental indicators covered in this paper are the following: net energy efficiency (electricity efficiency), water consumption, renewability and pollution emissions.

Net energy efficiency: Power generation from coal/biomass is essentially converting them to a source of energy that can be easily utilized. A higher net energy efficiency of a power plant, indicates better fuel utilization, hence providing more power while lower pollutants emission into the atmosphere. The net energy efficiency can be calculated using Equation 1. The values for the net energy efficiency of other power plants (Table 7S) are based on previous literature.

Renewability: A factor to measure sustainable development. It is a way to diminish the use of fossil fuels and promote renewable fuels. It is expressed as the mass ratio of the renewable feedstock to the total feedstock. Power generation processes that only use fossil fuel as energy source are given zero whereas ones that use biomass are given 100%.

Water Consumption: Due to environmental protection and water scarcity, one of the main goals for optimization of power plants is to reduce water consumption and its efficient use. The indicator for water consumption is calculated as tonnes of water per unit power output (ton/kWh).

Pollution Emission: Refers to SO_2, NO_X, CO_2 and dust emissions released from power plants which can cause detrimental effects to the environment. They are measured in grams per unit power output (g/kWh).

- Social indicators can be the fundamental elements in sustainability which includes community development and energy security.

Community Development: This indicator is qualitative which consists of more than one variable, however employment opportunities at the power plant is adopted as the main factor. It is calculated employee number per unit power output (employee number/MWe).

Energy Security: To ensure national security, having a reliable source of energy is essential. Biomass and coal can provide this security due to them being a reliable source. The energy security indicator is expressed as the ratio between the expected power capacity from the technology to the total electricity demand. It can also be affected depending on the government policies depending on the region. Currently in the UK there is no large-scale CCS power plants, hence all are given zero energy security. The rest are calculated based on the average percentage of energy contribution to the UK power supply.

- Technical indicators. Such indicator can be categorized into several variables including system reliability, system operability, technical maturity, etc. However, this study will only focus on technical maturity. Quantitatively technical maturity of each technology will be assessed using a categorical scaling method from 0 - 1, where 1 indicates a large scale industrial fully developed technology; 0.75 demonstrates pilot scale testing; 0.5 represents small test phase; 0.25 demonstrates laboratory test phase; and 0 indicates that basic research hasn't even started. Since coal fired power plant and biomass direct combustion have been tested widely in large scale before they both get 1, however with CCS (tested on pilot scale) it is given a 0.75. Similarly, coal IGCC was tested and developed in the UK during the 1970's, hence given a technical maturity value of 1 but 0.75 with CCS. Whereas with Biomass IGCC was not developed in large scale as much as coal, hence given 0.75 technical maturity and a 0.5 with CCS. BCLGCC has been tested in pilot scale and is still being developed hence was given technical maturity value of 0.5 with and without CCS.

Economic Analysis: Calculation of the Total Overnight Cost (TOC)

Figure 6S presents a flowchart summarizing the breakdown to calculate the TOC. The sum of all the unit's capital costs calculated using Equation 7 will give you the Total Plant Equipment Costs (TPEC). However, calculating the Bare Erected Cost (BEC) would need to take into consideration the direct and indirect costs which can be estimated by considering the assumptions in Table 4S [1], followed by adding the process and project contingencies (25% of BEC and 20% of BEC + Process Contingency, respectively) to calculate the Total Plant Cost (TPC). Finally, to calculate the Total Overnight Cost (TOC), the owner's cost is added to the TPC. Table 7S summarizes the values of all the parameters used to calculate costs.

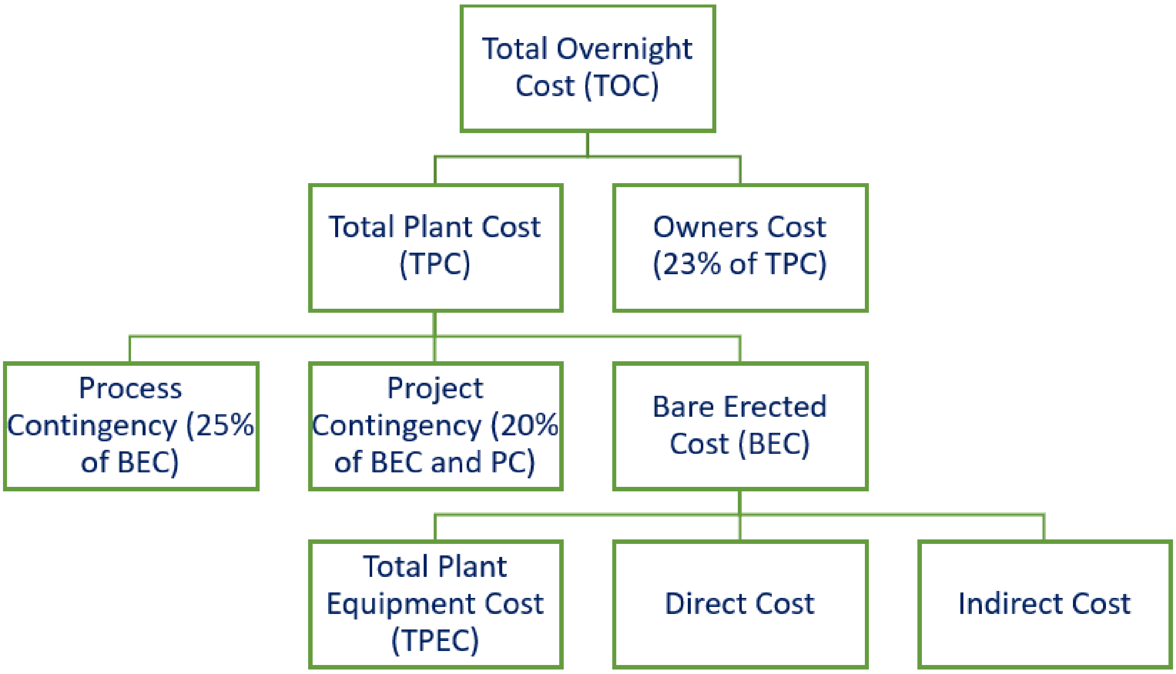

Figure 6S. Flowsheet breakdown to calculate the TOC

Table 4S. Direct and Indirect costs for calculation of BEC and Estimations for the owner's cost

Direct Cost	TEC	Owner's Cost	Estimate Basis
Purchased Equipment Installation	30%	Pre-production cost	6 months operating labour 1 month maintenance materials at full capacity 1 month non-fuel consumables at full capacity 1 month waste disposal 25% of one month's fuel cost 2% of TPC
Instrumentation and Control	20%	Inventory capital	0.5% of TPC 2 months supply of fuel at full capacity 2 months non-fuel consumables at full capacity
Piping	55%	Land	1.8% of TPC
Electric System	20%	Financing cost	2.7% of TPC
Buildings	12%	Other owner's costs	15% of TPC
Yard Improvements	15%	Total Owners Cost	23% of TPC
Services Facilities	40%		
Indirect Cost	**TEC**		
Engineering and Supervision	30%		
Legal Expenses	4%		

Table 5S. Values of different parameters used when calculating costs

Parameters	Values	Reference
Capacity Factor	0.8	
Plant Life, years	25	
Discount Rate, %	10	
Capital Recovery Factor	0.11	
Energy for CO_2 capture, GJ/tCO_2	0.44	[14]
Total Fixed Operating Cost		
Labor Cost, $/hr	34.65	[15]
Working Hours Per Week, hr	40	
Operating Labour, 650 MW Plant	109	[16]
Operating Labour CCS plant	110% of Non-CCS	[17]
Operating Labor Burden	30% of base	[15]
Labor Overhead Charge Rate	25% of labor	[15]
Maintenance Labor	1.25% of labor	[15]
Property Tax & Insurance	2% TPC	[15]
Total Variable Operating Cost		
Price of OC, $/ton	95	[3]
OC Lifetime, hr	1315	[18]
Boiler Feed Water, $/ton	0.11	[19]
Cost of Biomass, $/GJ	12.4 (£10.0)	[7]

Table 6S. Summary of TOC, variable and fixed cost breakdown

	Non-CCS (M$)	CCS (M$)
Gasification Units	173	170
Gas Turbine	40	37
HRSG	56	55
Steam Turbine	23	26
WGS Reactor	-	32
CO_2 Absorption	-	33
CO_2 Drying and Compression	-	29
Total Equipment Cost (TEC)	292	382
Direct Cost	561	734
Indirect Cost	100	130
Bare Erected Cost (BEC)	953	1246
Process Contingency	238	312
Project Contingency	238	312
Total Plant Cost (TPC)	1429	1870
Owners Cost	329	430

Total Overnight Cost (TOC)	1759	2300
Annual Operating Labour (OL)	9.5	10.5
Maintenance Labour Cost	11.9	13.1
Administrative and Support Labour	5.4	5.9
Property Taxes and Insurance	28.6	37.4
Total Fixed O&M Cost	55.4	67.0
Biomass	558.8	548.1
Oxygen Carrier	7.35	7.20
Boiler Feed Water	0.8	0.7
Total Variable O&M Cost	567	556

Table 7S. Techno-economic comparison between 5 power generation technologies [4,5,7– 13, 15]

Items	Direct Combustion		Integrated Gasification Combined Cycle		Chemical Looping Gasification Combined Cycle		Direct Combustion		Integrated Gasification Combined Cycle	
Gross Plant Power	~650		~650		~650		~650		~650	
Fuel	Biomass		Biomass		Biomass		Coal		Coal	
Carbon Capture	No CCS	CCS	No CCS	CCS	No CCS	CCS	No CCS	CCS	No CCS	CCS
Percentage Capture, %	0	~90	0	~90	0	~90	0	~90	0	~90
CO_2 Purity, %	-	~99	-	~99	-	~99	-	~99	-	~99
Biomass LHV, MJ/kg	16.5 - 18.5						25 - 32			
Cost of Fuel, $/GJ	9 - 12						2 - 3			
Plant Capital Cost, M$/MW$_{(Net)}$	1.4 - 1.8	3.3 – 3.8	2.6 – 3.0	3.7 – 4.2	2.4 – 2.8	3.4 – 3.9	1.2 - 1.6	2.6 – 3.0	1.8 – 2.2	2.7 – 3.1
Cost of Electricity, ¢/kWh	12 - 16	18 - 22	15 - 19	19 - 25	16 - 20	19 - 23	6 - 8	11 - 14	7 - 9	10 - 13

Net Electric Efficiency, %	34 - 37	24 - 28	35 - 40	26 - 33	40 - 43	34 - 37	35 - 38	25 - 28	37 - 41	30 - 34

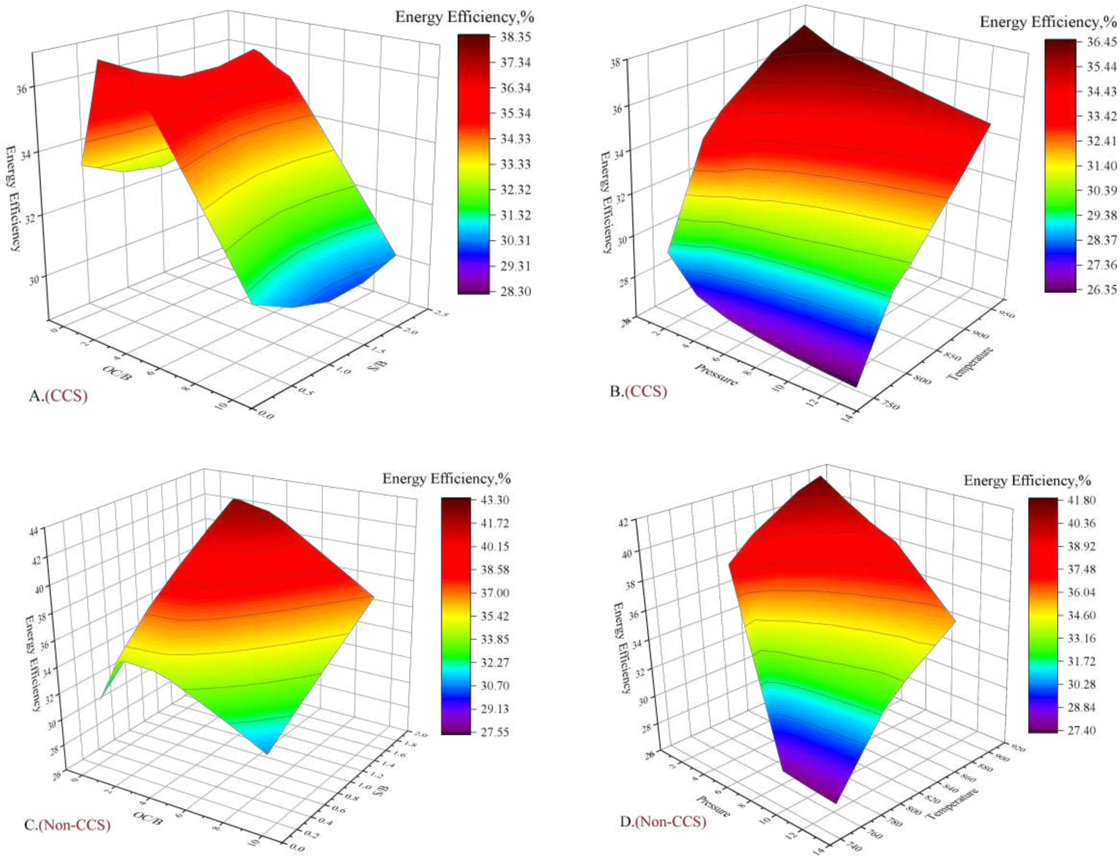

Figure 7S. Interaction of operation parameters and technological conditions on energy efficiency of the plant

FORTRAN statement calculation for biomass pyrolysis to decomposed into its constituent components.

The RYield is used to simulate the biomass pyrolysis which is only temporary placeholder. Fortran expression is defined to calculate the actual yield distribution of pyrolysis. The following Fortran code shows the process in detail:

```
FACT IS THE FACTOR TO CONVERT THE ULTIMATE ANALYSIS TO A WET BASIS.
        FACT = (100 - WATER) / 100
        H2O = WATER / 100
        ASH = ULT(1) / 100 * FACT
        CARB = ULT(2) / 100 * FACT
        H2 = ULT(3) / 100 * FACT
        N2 = ULT(4) / 100 * FACT
        CL2 = ULT(5) / 100 * FACT
        SULF = ULT(6) / 100 * FACT
        O2 = ULT(7) / 100 * FACT
```

These calculations assume that the inlet stream consists entirely of biomass. ULT is defined as the biomass ultimate analysis on a dry basis. The variable WATER, defined as the percent H_2O in the PROXANAL of biomass, is used to convert the ultimate analysis to a wet basis. The remaining eight variables (H_2O through O_2) are defined as the individual component yields in the RYield block.

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