Assessment of biomass energy potential for SRC willow woodchips in a pilot scale bubbling fluidized bed gasifier

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Abstract

The current study investigates the short rotation coppice (SRC) gasification in a bubbling fluidized bed gasifier (BFBG) with air as gasifying medium. The thermochemical processes during combustion were studied to get better control over the air gasification and to improve its effectiveness. The combustion process of SRC was studied by different thermo-analytical techniques. The thermogravimetric analysis (TGA), derivative thermogravimetry (DTG), and differential scanning calorimetry (DSC) were performed to examine the thermal degradation and heat flow rates. The product gas composition (CO, CO_2, CH_4 and H_2) produced during gasification was analyzed systematically by using an online gas analyzer and an offline GC analyzer. The influence of different equivalence ratios on product gas composition and temperature profile was investigated during SRC gasification. TG/DTG results showed degradation occur in four stages; drying, devolatilization, char combustion and ash formation. Maximum mass loss ~70% was observed in devolatilization stage and two sharp peaks at 315–500 °C in TG/DSC curves indicate the exothermic reactions. The temperature of gasifier was increased in the range of 650–850 ^{o}C along with the height of the reactor with increasing equivalent ratio (ER) from 0.25 to 0.32. The experimental results showed that with an increment in ER from 0.25 to 0.32, the

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average gas composition of H_2, CO, CH_4 decreased in the range of 9–6%, 16–12%, 4–3% and CO_2 concentration increased from 17–19% respectively. The gasifier performance parameters showed a maximum high heating value (HHV) of 4.70 MJ/m^3, Low heating value (LHV) of 4.37 MJ/$m^3$and cold gas efficiency (CGE) of 49.63% at 0.25 ER. The ER displayed direct effect on carbon conversion efficiency (CCE) of 95.76% at 0.32 ER and tar yield reduced from 16.78 to 7.24 g/m^3 with increasing ER from 0.25 to 0.32. All parametric results confirmed the reliability of the gasification process and showed a positive impact of ER on CCE and tar yield.

Keywords: Renewable energy; Biomass gasification; Bubbling fluidized bed; SRC willow chips; Thermo-analytical techniques; Product gas composition and tar yield.

1 Introduction

The growing energy demand from coal and natural gas leads to a shortage of fossil fuel because of time constraint for its reproducibility and environmental issues regarding fossil's fuel emission: the greenhouse effect and global warming in the near future. Biomass is a preferable energy source due to abundantly available, easily storable, transportable, and independent of location and climate [1]. Biomass is considered as the fourth renewable, potentially sustainable source of alternative energy which meets 14% of the total world's primary energy consumption [2]. It was reported that 4.8 G tons of oil equivalent biomass will be used as a source of fuel in 2050 [3]. Biomass is a carbon-neutral energy source with zero CO_2 emissions [4, 5]. During the combustion of biomass fuels, useful energy and the same amount of CO_2 is released which was absorbed during the plant life cycle and emissions of SO_2 and NOx are extremely low. Therefore, biomass is a good choice as a clean and environment friendly fuel after coal and natural gas [6].

The selection of biomass fuel is dependent on ash/ residue contents, moisture contents, cellulose/lignin ratio, carbon and volatiles, alkali metal contents, calorific value and moisture contents [7, 8]. The process of ignition becomes difficult when the biomass moisture contents are more than 30% [9]. Thermochemical conversion (combustion, pyrolysis and gasification) while biochemical conversion (fermentation and anaerobic digestion) are two main available technologies for biomass conversion into energy [10]. Biomass gasification converts solid carbonaceous biomass into gaseous fuels under controlled conditions with limited oxygen and produces a mixture of hot gases that are cleaned and can be utilized in power generation through gas turbine [11, 12]. The product gas of biomass gasification is considered most important due to direct use for power generation, but it requires suitable operating conditions and product gas cleaning strategies for final applications[13]. The producer gas holds; H_2, CO, CO_2, CH_4, N_2, water vapours and other types of impurities i.e. alkali compounds, chlorine, sulphur, tar, nitrogen, char and particulates [14]. Syngas ($CO+H_2$) produced during biomass gasification is an eco-friendly fuel for electricity generation and considered a versatile technology [15]. Hydrogen gas is an efficient clean energy carrier for the production of electricity that can be produced from biomass gasification. CH_4 and other liquid fuels can also be generated from syngas [1].

Many researchers have reported the studies on the effect of gasifier type, the composition of bed material, gasification temperature, equivalent ratio (ER), biomass feedstock type on the gasification and product gas composition [16]. A variety of designs and technologies were developed in combustion plants, gasifiers, and pyrolysis plants. Fixed bed gasifiers and fluidized bed gasifiers technologies were largely investigated for biomass gasification by a number of scientists in past decades [17]. The disadvantage of the fixed bed gasifiers is the difficulty in

maintenance of the constant operational temperature [16]. Furthermore, the bubbling fluidized bed biomass gasification is largely preferred over other technologies because of high conversion efficiency, uniform temperature profile in the reactor that is suitable for gas-solid interactions.

Karmakar et al. [18] have studied the rice husk gasification in FBG to examine the influence of temperature variation from 650–725°C with air as a gasifying medium at 0.25 ER. Their results suggested that with temperature increment, H_2 and CO were increased in the range of 17.22–18.49% and 24.89–26.59%, while CO_2 and CH_4 were decreased from 14.92–12.61% and 2.62–1.96%. The improvement in CCE from 71.51–75.82% with temperature was due to high conversion of unburned particles at high temperatures. The study of Subbaiah et al. [19] explored the gasification potential of groundnut shell (GNS) in FBG in 650–900 °C at 0.20 to 0.40 ER. They investigate the air-steam gasification that suggested the gas yield of CO and H_2 was increased with a rise in temperature and maximum CCE was 83.4% at 800 °C. The maximum HHV (6.9 MJ/Nm^3) was observed at 0.30 ER and 800 °C temperature.

Singh et al. [19] reported the gasification process of ground Nutshell (GNS) at 0.29–0.33 ER. The gasification temperature was 650–800 °C while air was used as a gasifying agent. They used conventional charcoal in bed heating. The most optimum ER reported for GNS gasification was 0.31 that was showed 5.74% of CH_4, 91% of CCE and 71.8% of CGE. Both the above studies of GNS suggested the optimum ER's were in the range of 0.30–0.31 and Singh et al. [20] study reported highest CCE at 0.31 ER. Sarker et al. [21] reported the alfalfa pellets gasification in FBG that was found attractive fuel for grid power generation. They studied gasification at 0.25 and 0.30 ER and their results demonstrated the increment in bed temperature with an increase in ER. In

addition, the CGE of 39% and the gas yield of 1.6 Nm^3/kg was observed. The LHV of 4.2 MJ/Nm^3 was obtained that indicate the alfalfa is promising biomass in terms of energy conversion. Most of the experimental parameters were enhanced by increasing the input air at a constant feed rate.

Maglinao Jr. et al. [22] analysed the CCE, heating values and gasification efficiencies of three feedstock high tonnage sorghum, beef cattle manure and cotton gin trash in BFG in the temperature range of 730–790 ºC and ER (0.3–0.5). They observed high carbon content and high efficiencies for tonnage sorghum. The optimum H_2 generation was found at 780 °C and 0.40 ER. The steam, as well as air gasification of sawdust, was performed to investigate the thermodynamic effect. Air was proven as an efficient gasifying agent that showed higher energy efficiency than steam gasification. The efficiency was continuously decreased by increasing ER when either steam and air used as a gasifying medium [23]. The product gas composition of rice husk gasification in a BFBG has been investigated previously, the composition of H_2, CH_4, and CO was decreased with an increase in ER, but the composition of CO_2 was increased. The appropriate ER value reported for its gasification was 0.2–0.3. [24]. Mohammed et al. [25] performed gasification in FBG using empty fruit bunch (EFB) as biomass and air as a gasifying agent in the temperature range of 700–1000 °C. The H_2 and CH_4 concentrations were increased from 10.27 to 38.02 and 5.84 to 14.72 % respectively with increasing temperature. The concentration of CO was increased from 21.87–36.36%, while the concentration of CO_2 decreased from 63−12%. The gas yield was reached to ~92% at 1000 °C.

Sciazko et al. [26] reported that air gasification is mostly performed in 726–926 ºC temperature range in FBGs, while during air-steam gasification the increase in hydrogen generation, increased

the produced syngas with a high calorific value that helps to decrease the mixing of hydrocarbon and tar. High molecule weight tar components were observed at high temperature in 100 kW dual fluidized bed gasifier (DFBG). The tar molecules were primarily treated within gasifier and secondary treatment was outside the gasifier by different techniques; baffled filters, rotating particle separators, fabric filters, electrostatic filters, ceramic filters, and scrubbers etc. [27]. The torrefaction effect on syngas quality of SRC chips was investigated in BFBG. Syngas quality was investigated by tar concentration and gas yield. About 47% reduction in tar yield has been reported from BFB gasification of SRC with steam and air as a gasifying medium [28]. Another attempt was made when SRC willow gasification was tried in a down-draft gasifier but results showed that willow chips were not gasified due to bridging within the hopper. Afterwards, a stirring bar was employed to prevent bridging and gasification was done successfully. The product gas collection was unsuccessful, therefore could not be further analyzed [29].

To the best of our knowledge, there is limited information available on short rotation coppice gasification in BFBG. Therefore, this study is designed to fill the gap in knowledge concerning the gasification of SRC willow woodchips and thermochemical assessment. Detailed thermogravimetric analysis and effect of different operating variables such as ER and temperature on product gas compositions of SRC gasification are studied. SRC willow chips were selected due to resprouting capacity after coppice, ease of harvesting, ease of propagation, broad genetic breeding and high yield, which is able to fulfil the energy demands by high power generation [30]. The single planting of SRC can be harvested more than seven times due to resprouting ability [31]. The experiments were performed to investigate SRC gasification in a BFBG using air as a gasifying agent focusing on temperature profiles and product gas composition under different

parameters. The biomass degradation behaviour was examined by TG/DTG to estimate the heat flow and decomposition characteristics of biomass. In addition, the effect of ER on temperature was studied to explore exothermic and endothermic reaction during gasification. The HHV, LHV, CGE and CCE were calculated to examine the performance of gasifier. This is a comprehensive study that discloses the optimum and best operating conditions for SRC willow gasification in a bubbling fluidized bed gasifier. Furthermore, this study also covers the detailed product gas composition analyses to examine the SRC gasification and gasifier performance evaluation.

2 Experimental

2.1 Biomass characteristics

SRC willow woodchips (size: 3–10 mm) from a local SRC willow grower were selected for gasification in bubbling fluidized bed biomass gasifier (BFBBG). The proximate and ultimate analysis of SRC willow woodchips was performed using TGA Q500 and is given in Table 1 [7, 32]. The TGA/DTG analysis was done to determine the thermal behaviour and degradation characteristics of biomass [33]. The TGA/DSC analysis was performed to examine the heat flow per unit mass with temperature under an air atmosphere. The comparison of burning profiles of TG/DTG and TG/DSC was used to determine the stages of thermal degradation of biomass [34].

2.2 Experimental setup of Gasifier

The bubbling fluidized bed biomass gasifier (BFBBG) is schematically represented in Fig. 1 consists of a biomass feeding hopper, screw feeder, fluidized bed gasification reactor, cyclone, gas cooling unit, tar removal unit (a mop fan unit, a biomass/ char bed), electrically heated combustor, an air supply/ preheating system and data acquisition devices. In BFBG, the gasifying reactor can be virtually divided into the bed (gas-solid reaction) zone and the freeboard (gas phase reaction)

zone. The bubble formation within the bed of bubbling fluidized bed biomass gasifier increased the heat transfer rate of bed material and mixing efficiency of fuel particles and gasifying agent. The main gasifier consists of a stainless-steel reactor had dimensions of 108 mm diameter and height of 1800 mm. The fluidization air was entered through a blower. The flow rate of air was controlled by a rotameter. The air was fed into gasifier reactor by an air distribution plate with a pore size of 100 μm and thickness of 10 mm which also allows preheating up to required gasification temperature (700–900 ^{o}C) through electric pre-heater [35].

The SRC willow woodchips were fed into the combustor by a screw feeder just above the air distribution plate. The biomass feeding unit supported the desired feed rate of woodchips by using a screw auger and timed stirrer. The auger is used to transfer woodchips from the hopper into the reactor by an inverter. The inverter was used to control the frequency of the feeder motor. To prevent from backflow of biomass and sand particle into the feeding unit, a small amount of air was also introduced through feeder hopper. The temperature profiles of the gasification reactor were continuously monitored by placing eight K-type thermocouples at different distances from the distribution plate. These thermocouples are labelled as T1, T2, T3, T4 (bottom and middle thermocouples) and T5, T6, T7 and T8 (upper thermocouples) which are located at -5, 9, 17, 25, 45, 74, 105 and 150 cm height above from the distribution plate. T1 was located in the air chamber under the distribution plate, while T2–T5 measures the gasification temperature variation occurring in dense bed zone and T6–T8 are uppermost thermocouples set at freeboard region of reactor, monitoring the temperature variation of the gas exit.

The reactor was equipped with pressure sensors at different heights to examine the fluidization conditions of the bed. The bubble formation, rising and bursting of bed material in the reactor was observed by increasing the fluidization air flow rate that is shown in Fig. 2. The gas particles usually move upward from bed at a minimum fluidization velocity that is less than 5 m/s. The rising of gas from bed create bubbles that maintain bubble emulsion and fluidization state at the bed. After this dense bed, a freeboard region is present that reduces the supercritical velocity and return the particles towards the bed region. In this way, bed material almost remains fixed [36]. The pressure difference is closely checked at dense bed region and freeboard region to detect any sign of defluidization and agglomeration during gasification. If an abrupt change in pressure difference recorded across dense bed region and reactor temperature, it means defluidization.

This research was designed to focus on product gas composition and temperature profiles. All the above mentioned tests were performed to avoid defluidization and agglomeration during gasification [37, 38]. At the exit point of the reactor, a cyclone is fitted for the removal of particles from product gas to achieve high efficiency. The ash particles are collected in an ash pot from the bottom of the cyclone. After cooling, product gas by gas cooler it is introduced into the mop fan cleaning unit. The centrifugal fan casing mop with 70 mm fibre length and 0.4–0.6 mm diameter of each fibre is used for gas circulation, de-dusting of contaminated product gas stream and efficient removal of gaseous contaminants. The efficiency of particles removal is improved by a water spray, fibre number and fibre arrangement. An on-line gas analyzer (ABB Easy Line analyzer) is fitted at the end of a gasifier to continuously determine the product gas (CO, CO_2, CH_4 and H_2) composition. The analyzer calibration was done with standard gas samples. For comparison, the product gas was also analyzed in an off-line offline gas chromatography (GC)

analyser. All of the measured and processed data such as product gas profile and temperature are continuously monitored by computerized systems.

2.3 Gasifier operating conditions

The operational parameters of gasifier and gasification process are characterized by equivalence ratio, temperature profile along with the height of the reactor, feeding rate and gas concentrations at the exit. The constant biomass flow rate was achieved through repeated calibration after regular intervals by checking the amount of biomass flow from the hopper to the gasification reactor in a specific time through the auger. The timed stirrer was adjusted as 5 sec/min to the hopper. Timed stirrer and feeder confirmed the stable feed rate to achieve steady product gas and temperature profile. An auto stirrer was used to stirrer the biomass constantly to prevent bridging and to improving the gas quality [39]. The reliable sampling for reproducible results of product gas analysis was obtained by using different rotation speed of auger i.e., 10, 11 and 12.5 Hz. The auger speed controls the feeding rate of wood chips. The inverter of gear motor controls the auger rotational speed.

The gasification was performed at three different ERs; 0.25, 0.29 and 0.32 in BFBG. The fluidization and gasification process are examined by a set of thermocouples and pressure sensors. Compressed air was used as a fluidization medium at room temperature. The operating conditions for biomass gasification of willow chips are given in Table 2. When desired temperatures have obtained in the gasification reactor, the biomass is introduced into the gasifier and air was fed accordingly to the selected ER values. A small amount of air (3 L/min) at room temperature and 1 atm pressure was introduced into the hopper to prevent any backward diffusion of biomass and sand particles. The ER was modified by adjusting the gasification airflow rats; 45, 65 and 80 L/min

for 0.25, 0.29 and 0.32 respectively. After gasification initiation, a steady state condition was achieved in the reactor after about 30 min of feeding and all variables were continuously monitored. All parameters in the rig (pressure, temperature and product gas composition analysis) were recorded by the data acquisition system.

2.4 Estimation of Gasifier's performance

The performance of BFBG was investigated by calculating gas yield, cold gas efficiency (CGE), Carbon conversion efficiency (CCE), high heating value (HHV) and low heating value (LHV) of product gas. The gasifier performance was monitored by comparing the gasification process at different ERs (0.25, 0.29 and 0.32). These calculations were performed by using following equations 1–5 [6, 21].

a. HHV

HHV= (H_2 conc. in product gas * HHV of H_2) + (CO conc. in product gas + HHV of CO) + (CH_4 conc. in product gas * HHV CH_4) (1)

b. LHV

$$LHV_{gas} = \sum Vi*LHVi \quad (2)$$

where Vi = % composition of gas component in the product gas

LHVi = lower heating value of the individual gas component

c. Gas yield

Total gas yield = Amount of N_2 fed to the gasifier * N_2 concentration in the product gas

$$= N_2\ (m^3/h) * (100- \sum V_{gi}) \quad (3)$$

where $V_{gi} = N_2$ = Air fed into the gasifier (m^3/h) * N_2 concentration in product gas

d. Cold gas efficiency

Cold gas efficiency was calculated by the following equation:

$$CGE(\eta) = \frac{\text{Gas yield} * \text{LHVgas}}{\text{Biomass feeding rate} * \text{LHVbiomass}} * 100 \quad (4)$$

e. Carbon conversion efficiency

$$\mathrm{CCE}(\%) = \frac{12(\mathrm{CO2}+\mathrm{CO}+\mathrm{CH4}+2.5\mathrm{CnHm})*Gas\ yield*100\%}{22.4*298/273} \quad (5)$$

3 Results and discussion

3.1 Thermogravimetric analysis

The Thermogravimetric analysis (TGA) was performed to investigate the chemical reactions and thermal stability of biomass with quantitative measurement of weight loss over a specific temperature range. Fig. 3 shows the sample weight loss and derivative mass loss with temperature by using air as a reacting medium. Blue curve indicated four degradation stages; drying, devolatilization, char combustion and ash formation [40]. Red line shows the DTG curve. The initial mass loss (up to ~9%) was observed in the drying stage at temperatures of 26–125 ^{o}C that is associated with the removal of moisture. SRC willow chips showed 10% moisture content that was determined by ultimate analysis as given in Table 1. This moisture content is most suitable for gasification. The presence of higher moisture content needs more energy for the drying process during gasification [41].

Most of the volatile components, tar, gases and char produced during thermal decomposition of biomass (devolatilization) and almost 70% mass loss of initial weight occurred in devolatilization zone (126–363 ^{o}C) region [33]. The ignition temperature (T_{ign}) represents the onset of devolatilization stage and known as an active pyrolysis region, which was started at 259 ^{o}C. The peak temperature of the devolatilization region was 337 ^{o}C that is known as glass transition temperature (T_g) as derived from TGA/DTG curves [42]. The mass loss of ~27% was observed in

277 the temperature region of 363–513 ºC because of char combustion. Finally, the ash formation was
278 observed in the temperature range of 513–850 ºC containing ~1% biomass sample.
279
280 The DTG curves showed two peaks; first sharp and strong peak was observed in 261–374 ºC
281 temperature range having a maximum rate of mass loss 35 %/min at 338 ºC. This region revealed
282 the degradation of carbon content and devolatilization of biomass. The second peak in the DTG
283 curve indicates the char combustion that was obtained in the secondary pyrolysis zone (275–510
284 ºC) with maximum mass loss rate ~11 %/min at 483 ºC. Beyond 510 ºC, the DTG curve showed
285 a slow rate of mass loss ~1 %/min because degradation is almost completed in this region and only
286 char residue left behind. Thermal degradation behaviour of biomass showed that pyrolysis of
287 hemicellulose usually occurred at a temperature below than 350 ºC. While cellulose pyrolysis
288 occurred in temperature ranges 250–500 ºC. There was no sharp peak beyond 500 ºC because most
289 of the biomass have already degraded and only lignin shows some thermal stability [34, 42, 43].
290 Nyakuma et al. [44] studied thermochemical assessment of the empty fruit bunch (EFB) by heating
291 in the temperature range of 50 °C to 900 °C at 10°C/min heating rate. TGA results suggested four
292 stages of biomass thermal decomposition and 70% weight loss was observed in devolatilization
293 stage at 325 ºC peak temperature.

294 3.2 Differential scanning calorimetry

295 Fig. 4 shows TG/DSC curve heat depicted the heat flow per unit mass during exothermic and
296 endothermic reactions with temperature variation. The main reactions that take place during
297 biomass gasification are given in Table 3. In accordance with Le Chaetlier's principle, endothermic
298 products and exothermic reactants are favoured at high temperature. At initial, drying and heating

of biomass require heat and endothermic reactions are favoured. Two sharp peaks were observed in the temperature range of 315–500 °C which showed the exothermic reaction of gasification.

Most of partial char combustion and devolatilization occurs in this region, which releases a large amount of heat due to the combustion of unburned particles and release of volatiles. At this stage, TGA also confirmed the breakdown of larger chains of hydrocarbon into smaller chains, and thermal decomposition of fuel into the gaseous product [34]. The heat flow curve suggested that as the gasification proceeds with the rise of temperature, endothermic reactions such as Boudourad reaction, water gas shift reaction and methane reforming reaction were favoured. The gas products are then reformed through these reactions [45]. Zhao et al., [46] reported the detail pyrolysis of corn straw and soybean straw with TG/DTG and TG/DSC analysis in a fixed bed reactor. Their results found four pyrolysis stages and temperature increased with increasing heat rate that produces high char yield.

3.3 Effect of ER on gasifier temperature

The temperature of gasification is considered important in determining the composition and yield of the product gas. Temperature is not an independent factor in gasification. The temperature profile of gasifier is linked to the amount of air available (ER), therefore, both are considered important parameters for gasification. The temperature profiles of gasifier are controlled by ER; with an increase in ER the amount of air introduced into the reactor is also increased which enhances the oxidation rate of biomass. This enhanced oxidation rate increases the heat release and carbon conversion content which results in an increment in the temperature of gasifier reactor. Fig. 5 presents the influence of ER on the temperature profiles of the gasifier reactor (T2–T8). By increasing ER from 0.25 to 0.32, the temperature distribution in all regions such as; dense board

and freeboard showed an increase to their maximum values of 817, 812, 785, 777, 778, 782, 548 ^{o}C for T2 to T8 respectively. Therefore, higher ER value showed that most of the partial combustion occurs in the gasifier reactor due to increased temperature.

This increasing temperature with ER also continued distinctive differences between all sensors at a different height. This increment in temperature was due to more air entered into the reactor in oxidation zone and promote combustion that has increased the temperature of gasifier [21]. The results indicated that when ER is increased from 0.25 to 0.29 then 0.32, the temperatures at all thermocouple in the reactor were smoothly increased. As the high temperature is achieved in the gasifier, a series of endothermic reactions including drying, char combustion with exothermic reactions (devolatilization) started which describe the temperature behaviour along with the height of the reactor. At the exit of the gasifier reactor, the temperature is decreased because of heat losses and endothermic char combustion and tar cracking [47]. This trend is also confirmed by the heat flow curve shown in Fig. 4. Nevertheless, there is a decrease in temperature at -5 cm distance (T1) from the distribution plate because of introducing an additional air without preheating into the reactor. [48]. The highest temperature observed in the oxidation zone while the lowest temperature was recorded in the upper portion of gasifier in the pyrolysis region [49]. The high temperature favoured water gas shift reactions which further promote steam methane reforming reaction [50]. By comparing the effect of different ER value, the higher temperature was observed with ER=0.32 which indicate the increase in temperature by increasing ER value.

Temperature is considered the most important factor that affects the gasification process. González-Vázquez et al., [51] estimated the effect of temperature on pine kernel shell gasification,

product gas concentrations, gas yield and CGE. The results concluded that the rise in temperature from 700 to 900 °C favoured high H_2 production and best gasifier performance. From literature, it is observed that higher temperature is responsible for higher hydrogen concentration and low tar content because of thermal tar cracking reactions. Due to this more volatiles were released at high temperature and increased the overall gas yield. The BFBG is considered a promising option for hydrogen generation. Therefore, hydrogen generation through thermochemical route such as biomass gasification in BFBG is remarkable technology. The desired gasification temperature is achieved by partial combustion. The yield of producer gas, CGE and tar contents in the syngas are dependent on the temperature of gasification [52]. Perez et al. [53] investigated the thermodynamic and fluid-dynamic analysis of sugarcane bagasse in BFBG. Geldart's types of particles were used to examine the fluidization parameter in gasification. The results showed the 4.56 MJ/Nm^3 of LHV and 0.8–1.21 mm of ideal particle size was suggested for large scale gasification.

3.4 Effect of equivalence ratio on product gas compositions

The equivalence ratio (ER) has a greater effect on concentrations of product gas and calorific value of syngas that directly affects the performance parameters of gasification [15]. The ER was varied from 0.25 to 0.32 to examine the effect of ER on product gas composition. The gasification airflow rate was changed from 45 to 65 and then 80 L/min to obtain a selected range of ER in the gasifier. These experiments were performed with feeding rates 1920.9, 2126.8 and 2469.6 g/h for 0.32, 0.29 and 0.25 ER respectively. The stable and constant biomass feeding is used for biomass gasification because it is considered important for reliable product gas sampling and their analysis. The feeding process and product gases are affected by moisture content, auger rotational speed, dimensions of

biomass particle. The size of SRC willow was 3–10 mm that provides excellent heat transfer rate [54, 55].

3.4.1 Online analysis of product gas compositions

ER values distinguish the combustion and gasification process. ER value less than 1 is considered for gasification and optimum range from 0.2–0.4 has been reported for biomass gasification. If ER value is taken below 0.2 then it produces unnecessary char, syngas with low heating value and incomplete gasification occurred. While ER values above 0.4 also cause a problem, such as extreme production of combustion products (CO_2, H_2O) [50]. The product gas found from air gasification of biomass is usually composed of some combustible and incombustible gases. The analysis of these products (CO, CO_2, CH_4, and H_2) was done by online gas analyzers. Four runs of gasification were performed at each ER value to check the repeatability and reliability of the experiments and product gas values. Fig. 6 gives the distribution of product gas composition at various ERs monitored by the on-line gas analyzer as well as GC analyzer. Only slight differences in product gas composition were observed for each run, which confirmed the reliability and reproducibility of the experiments.

The product gas concentrations with online gas analyzer are presented in Table 4. At 0.25 ER, the average concentrations of CO, CO_2, CH_4 and H_2 were 16.98, 17.49, 4.43 and 9.95% respectively. The concentrations observed at 0.29 ER were 14.20, 18.19, 3.94 and 8.26 % for CO, CO_2, CH_4 and H_2 respectively. While the compositions of product gas at ER 0.32 were 12.72, 19.21, 3.88 and 6.30% for CO, CO_2, CH_4 and H_2 respectively [56]. These values show that with increasing ER value from 0.25 to 0.32, the concentration of CO, CH_4, H_2 decreased while CO_2 increased. The product gas composition obtained from Fig. 6(a-c) indicates that the concentration of CO

decreased from 16.98 to 12.72% with an increment in ER from 0.25 to 0.32. The concentrations of CH_4 and H_2 were also decreased from 4.43–3.88 % and 9.95–6.30% respectively by increasing ER in the selected range. The decrease in H_2 concentration was observed because of the water gas shift reaction in which H_2 consumption rate was greater as compared to H_2 formation rate [56]. While the decreased concentration of CH_4 from 3–4% with increasing ER was due to methanation.

However, the CO_2 composition was found to increase from 17.49 to 19.21% by increasing ER from 0.25 to 0.32. By increasing ER, more air entered into the reactor, CO and CH_4 were burned with O_2 and formed CO_2 due to excessive availability of air. The results of CO, H_2 and CH_4 compositions displayed opposite trend to CO_2 composition in product gas profile of gasification. These trends were due to different ER values because ER indicates the quantity of actual air available for volatile formation and gasification of fuel. During gasification, gas products react with oxygen and produce CO, CH_4, H_2 and CO_2 gases. These gaseous products are formed by series of reaction including carbon reaction, Boudouard reaction, water gas shift reaction or methanation reactions as already depicted in Table 3 [52].

Therefore, low concentration of combustible gases (CO, CH_4 and H_2) was found at high ER value and diminished the product gas quality. The low ER favoured endothermic reactions that are char + CO_2 and water gas shift reactions. Therefore, more char and CO_2 used in these reactions which lower the CO_2 composition [57]. Considering the trends of gas concentrations with decreasing ER, the maximum increment is obtained for CO concentration while the small increment is found in CH_4 concentration. The increment in H_2 concentration is seen smaller than the increment in CO concentration. Likewise, the increment in CO concentration is greater than the decrease in CO_2

concentration with decreasing ER. Similar results were reported by Sarker et al. [21] which showed the increase in CO_2 concentration when the ER value was increased from 0.20–0.35.

Though, the current study showed enhanced CH_4 concentration nearly 4–3%, which is greater than CH_4 concentration (2.6–2.0%) of wheat straw pellet gasification as reported Sarker et al. [21]. Kim, Yang et al. [48] reported that a decrease in the concentration of H_2 and CO was due to water gas shift reaction. By increasing ER from 0.25 to 0.32, the H_2 product gas was decreased by approximately 9–6% due to the dominant oxidation reaction at higher ER value because oxidation in gasification reactor dominates and produce less H_2 gas [58]. The higher concentration of CO in the product gas was also reported by Makwana, Joshi et al. [59] which was due to the lower value of ER. They gasified rice husk and char of rice husk by varying the ER from 0.30 to 0.38. The higher CO (18%) and H_2 (5.6%) composition were found at low ER due to endothermic steam reforming reaction. In the present study, the gasifying medium was air which increased oxidation reaction by producing CO_2 from the utilization of CO and O_2. The air also promotes complete oxidation of fixed carbon component and oxygen resulting CO concentration drops. More residual carbon is produced by increasing gasifying medium and combustion due to pyrolysis in steam gasification which increases the carbon conversion efficiency.

3.4.2 Gas chromatography analysis of product gas composition

The product gas composition analysis was also carried out through Gas chromatography (GC), to check the reliability and reproducibility of the online gas analyzer. When all gasification parameters become stable, the four sampling bags of 0.5 litre were filled with product gas for offline GC analysis. This analysis was performed with the same ER's as used for online analysis. The composition of product gas measured at the inlet of combustor was comprised of CO_2, CO,

H_2, CH_4 and a small amount of O_2. This gas composition was important for partial oxidation. Table 4 shows GC analysis of product gas compositions at different ERs.

The GC analysis showed 16.64, 17.99, 4.67, 8.96% concentrations of CO, CO_2, CH_4 and H_2 respectively at 0.25 ER. Karatas, Akgun et al. [52] reported pilot-scale gasification of natural wood at the same ER 0.25 as reported by the present designed study. They reported the 5% concentration of CH_4 that is nearly similar to the current study of SRC at this ER. The concentrations observed at 0.29 ER were 14.50, 18.16, 4.06 and 8.31% for CO, CO_2, CH_4 and H_2 respectively. While the compositions of product gas at ER 0.32 were 12.29, 18.24, 4.22 and 6.53% for CO, CO_2, CH_4 and H_2 respectively [56]. The results of product gas concentration by GC analysis are in clear agreement with the online analyser. The GC analysis results showed that CO_2 increased from 17.99 to 18.24% with increasing ER (0.25–0.32). This is because the oxygen supply was dominant as of increasing air feeding that burns volatile component of biomass by producing more CO_2. This gas also dilutes the other combustible gases therefore, at higher ER less concentration of H_2, CH_4, and CO is produced. At 0.32 ER, the concentration of CH_4 was low (4.22%) but it can significantly change the gas heating values [60].

The product gas composition results of online analyses and off-line GC analysis are already given in Table 4. The results of product gas obtained from both analyzers are then compared to check the efficiency of analyzers. The comparison of product gas profile at 0.25 ER is shown in Fig. 6(a). Their results showed the one-factor difference in H_2 concentration of GC and online analysis. The H_2 concentration recorded by GC analyser was 8.96%, but online analyser recorded 9.95%, that is marginally different. However, all other remaining product gas composition showed stability from

both analyses [50, 57]. The comparison of product gas at 0.29 ER is represented in Fig. 6(b). The results indicated a slight difference in CH_4 composition. The CH_4 concentration recorded by GC analyser was 4.06%, however, online analyser recorded 3.94%. While the other gas compositions were almost the same from both analysers. At ER 0.32, a comparison of product gas profiles from GC and online analyzers is given in Fig. 6(c).

The comparison confirmed the quantitative composition of product gases that shows only one factor difference in CO_2 concentration while other gas compositions almost same and showed uniformity from both analysers. This comparison was effective and confirmed the efficiency of both analysers. Therefore, the qualitative and quantitative results of product gas at different ER from both GC and online analysers are in excellent agreement with each other and confirmed the efficiency of analysers and the energy profile of SRC willow.

3.5 Gasifier performance evaluation

The performance of bubbling fluidized bed gasifier was examined in terms of product gas concentrations and carbon conversion. A set of desirable efficiency parameters were investigated to check the efficiency of converting input mass into synthesis gas product with reduction of tar.

3.5.1 Bed temperature and tar yield

The effect of ER on average bed temperature and tar yield was studied. Fig. 7 confirms that ER values have a positive linear correlation with average bed temperature and negative linear correlation with tar yield. When the bed temperature increased from 776 to 817 ^{o}C, a decrease in tar yield from 16.78 to 7.24 g/m^3 by was noticed by increasing ER from 0.25 to 0.32 respectively. Therefore, the increment in the ER in the gasifier enhanced the average bed temperature about ~40

481 °C, which is considered effective for gasification. This increment in bed temperature with ER was
482 due to more air availability for oxidation reaction that promotes heat release.
483
484 The high bed temperature also favoured H_2 and CO concentration in accordance with Le
485 Chatelier's principle, which depicted that reactant substance in exothermic reactions and product
486 substance in endothermic reactions favoured at a higher temperature. Therefore, endothermic
487 steam gasification reaction is strengthened at a higher temperature with increasing ER [18]. The
488 increase in bed temperature with ER was also due to the uniform size of bed particles, which was
489 easily deposited on the bed surface. These particles were burnt easily when coming in contact with
490 bed surface and release heat. Air can easily circulate throughout the reactor due to pressure
491 fluctuation in the reactor and bed surface that improved fluidization and combustion behaviour
492 and increased the bed temperature [20]. Improper fluidization may cause agglomeration and failure
493 of the gasification process [38]
494
495 The increase in carbon conversion efficiency and bed temperature with the increase of ER from
496 0.25 to 0.32 improves the tar decomposition. This is because of thermal cracking and reforming
497 reactions [50]. ER played an important role in tar content and their properties. By increasing ER
498 from 0.25 to 0.32, air availability is increased which decreased the tar yield from 16.78 to 7.24
499 g/m^3 as given in Fig. 7. This decreasing trend was due to more combustion of hydrocarbons and
500 tar cracking reaction at high ER. The tar cracking reactions including steam gasification reaction
501 as well as reforming reaction decrease the tar yield. The partial combustion reaction increased the
502 temperature of the reactor, which favoured char gasification with high conversion rate and reduces

503 the tar yield [37]. However, higher ER improves the bed temperature that causes tar decomposition
504 and decreased the tar yield and this reduction in tar yield is very important for gasification.

505 **3.5.2 Evaluation of LHV and gas yield**

506 The gasification parameters such as LHV, Cold gas efficiency (CGE) and gas yield were
507 investigated to examine the gasifier performance efficiency. The CGE is the percentage of LHV
508 of biomass converted into LHV of product gas. CGE is also called gasification efficiency. As the
509 ER value is varied from 0.25 to 0.32, all gasification parameters (gas yield, CGE, LHV, CCE)
510 were also changed. Fig. 8 shows a decrease in LHV and an increase in the gas yield of product gas
511 with increasing ER values. The LHV was decreased from 4.37 to 3.67 MJ/m^3 by increasing ER
512 from 0.25 to 0.32. This was due to less production of combustible gases (CO and H_2) which
513 favoured a decrease in LHV. The LHV was decreased due to exothermic water gas shift reaction
514 as well as dilution of syngas with air nitrogen [12]. The HHV was decreased from 4.70 to 3.95
515 MJ/m^3 while the LHV was also decreased with increasing ER, this trend was also observed in the
516 previous studies. Karatas, Akgun et al. [52] reported that the LHV of walnut shell and pistachio
517 shell was decreased by increasing ER from 0.19 to 0.37. It can be seen in Fig. 8 that the gas yield
518 increased from 3.93 to 4.55 m^3/h by increasing ER from 0.25 to 0.32 respectively.
519
520 The increment in gas yield is because of increment in the gasifying medium that is caused by
521 increasing ER. That is the reason the gas yield is varied with a gasifying agent. The temperature
522 becomes higher in oxidation zone at high ER, which favoured high volatilization of biomass and
523 char gasification. Moreover, further increment in temperature, increase the gas yield due to the
524 steam reforming reaction. Meng et al. [61] reported the sawdust gasification with different
525 gasifying agents. The gas yield was changed from 2.11 to 2.41 m^3/h with increment in the ER from

0.20–0.30. The amount of gas yield obtained in the present study is more than gas yield reported for sawdust. This increment in gas yield could be due to biomass type and operating conditions of gasifier but the overall trend of gas yield with ER is in accordance with the literature.

3.5.3 Determination of CGE and CCE

At the selected ERs 0.25, 0.29 and 0.32, the CGE varied from 49.63, 47.89 to 46.43% respectively also shown in Fig. 9. Cold gas efficiency was influenced by ER. CGE examines how much heating content of biomass is used to convert the feedstock into product gas. By increasing ER, the low heating value (LHV) of producer gas decreased because of excessive oxidation of feed and more inert nitrogen is also introduced with air which diminished the quality of product gas [62]. Therefore, this rise in air availability diminished the product gas quality due to the large oxidation reaction of biomass.

The decreasing LHV of product gas showed the total energy conversion into product gas is decreased, which decreases the CGE [39]. Similar findings and trends of CGE and LHV with ER have been reported by Hamad, Radwan [63], Ahmed et al. [64] and Guo et al. [49]. The CCE is the percentage of gasified carbon content to the total carbon content in the added feed [18]. In an ideal system only, most of the biomass is to be transformed into desirable product gas mixture and other secondary particulates. However, in case of woodchips biomass gasification, carbon, oxygen and hydrogen in the feedstock are transformed into a mixture of synthesis gas, secondary products including carbon dioxide, methane and higher gaseous hydrocarbons and other unwanted particulates such as sulfur species, particulate matter and tars. The carbon conversion calculations at different biomass feeding rate were performed by using constant gasification airflow rate (44.72 L/m) and during 60 min of feeding. The carbon conversion efficiency (CCE) was estimated at

549 0.32, 0.29, and 0.25 ER with 1920.9, 2126.8 and 2469.6 g/h feeding rates respectively, that is also
550 displayed in Fig. 9.
551
552 The results indicate that the highest carbon conversion (95.76%) was achieved with 1920.9 g/h
553 feeding rate of biomass at 0.32 ER value. The CCE was 90.68 and 95.48% at 0.29 and 0.25 ER
554 respectively. By increasing ER from 0.25 to 0.32, the CCE efficiency is varied from 95.48 to 95.76
555 %. When ER is increased, more air is introduced into gasifier that favoured exothermic oxidation
556 reaction. This exothermic reaction increased the temperature of the gasifier and also promotes the
557 steam reformation that in turn increased the carbon conversion rate [50]. By increasing ER from
558 0.25 to 0.32 more carbon content of the biomass was converted into product gas (CO, CO_2 and
559 CH_4) which leads to gradually increase in carbon conversion rate. Therefore, CCE reached its
560 maximum value (95.76%) at 0.32 ER. Diyoke et al. [57] also reported that CCE depends on the
561 rate of oxidation of carbon particulates Therefore, CCE is increased and CGE is reduced with
562 increasing ER from 0.25 to 0.32.

563 # 4 Conclusions

564 In this present study, gasification characteristics of SRC willow chips were investigated using
565 bubbling fluidized bed gasifier (BFBG) at 600–850 ^{o}C and at different equivalence ratios of 0.25,
566 0.29 and 0.32. The thermochemical investigation was done by TG/DTG and TG/DSC analysis to
567 explore the thermal stability and degradation characteristics of biomass. Furthermore, the influence
568 of ER on concentrations of product gas was examined by online and offline analysis. The main
569 findings of the study are summarized as follows:

570 • The TGA/DTG analysis was performed to examine the thermal degradation characteristics
571 of biomass. The highest weight loss observed in the devolatilization stage was ~70% in the

temperature range between 126 and 363 ^{o}C. While two sharp peaks observed within the range of 315 to 500°C in TG/DSC curves indicate the exothermic reactions. Heat release can be utilized in power generation.

- By increasing ER, the temperature profiles of reactor increase and the highest temperature were observed in dense board region in the range of 650–850 ^{o}C. The increased bed temperature with increasing ER is considered important for tar reduction and to improve the carbon conversion rate.
- An increment in ER from 0.25 to 0.32, the GC and online analysis showed the average concentration of CO, CH_4 and H_2 decreased in the range of 16–12%, 4–3% and 9–6% respectively. In addition, the CO_2 concentration increased from 17–19 % in the product gas composition. This is because of more air availability for oxidation at high ER, which diminishes the product gas quality and lower the combustible gas concentrations. Both GC and online analysis of product gas compositions showed clear agreement with each other.
- Both the gas yield and CCE increased while LHV, CGE and tar yield gradually decreased with increasing ER from 0.25 to 0.32. The maximum carbon conversion efficiency of 95.76% was observed at 0.32 ER. These parameters results confirmed the reliability of the gasification process, gasifier performance and product gas composition.
- TGA and gasification results showed the high thermal stability and high carbon conversion efficiency of selected SRC willow chips. Therefore, SRC willow biomass is recommended as renewable energy fuel for the future power generation industry and for the other applications.

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List of Tables

Table 1. Proximate and ultimate analysis of SRC willow chips.

Biomass fuel	**Ultimate analysis (wt %)[a]**					**Proximate analysis (wt %)[c]**				
	C	H	N	O[b]	S	M	VM	FC	Ash	LHV(MJ/m^3)
SRC willow chips	45.4	5.7	0.8	48	0.1	2.9	82.5	12.9	1.7	4.4

M - Moisture; VM - Volatile matter; FC - Fixed carbon.
[a] On dry-ash-free basis.
[b] Calculated by the difference.
[c] On dry basis except for moisture which is on an as received basis.
[d] Low heating value (dry)

Table 2. Operating conditions of bubbling fluidized bed biomass gasifier (BFBBG).

Equivalent Ratio (ER)	0.25, 0.29 and 0.32
Gasification air flow rate (L/m)	45, 65, 80
Hopper air flow rate (L/m)	3
Fluidization velocity (m/s)	<5
Feeding rate (g/h)	2469.6, 2126.8 and 1920.9
Heater temperature setup (^{o}C)	650–850
Screw feeder motor frequency (Hz)	10, 11, 12.5
Silica sand particle size Dimension (μm) Density (kg/m$^{3)}$	 212–300 1520

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776 **Table 3.** Major recations of gasification.

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Reaction name	Reactions	ΔH^0 (KJ/mol)
Oxidation	$C(s)+O_2 \leftrightarrow CO_2$	-394.0[a]
	$C(s)+1/2O_2 \leftrightarrow CO$	-123.0[a]
Boudouard	$C(s)+CO_2 \leftrightarrow 2CO$	+172.0[b]
Water gas	$C(s)+H_2O \leftrightarrow CO+H_2$	+131.0[b]
	$C(s)+2H_2O \leftrightarrow CO2+2H_2$	+77l.0[b]
Methanation	$C(s)+2H_2 \leftrightarrow CH_4$	−87.0[a]
Water gas shift	$CO+H_2O \leftrightarrow CO_2+H_2$	−41.0[a]
Steam reforming	$CH_4+H_2O \leftrightarrow CO+3H_2$	+206.0[b]

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779 [a] Negative sign indicates the exothermic reactions.

780 [b] Positive sign indicates the endothermic reactions.

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783

784 **Table 4.** Product gas analysis and gasifier performance of SRC willow chips

Feedstock	**SRC willow chips**		
ER	0.25	0.29	0.32
Feeding rate (g/h)	2469.6	2126.8	1920.9
Product gas composition (Vol %)			
H_2	9.95[a] 8.96[b]	8.26[a] 8.31[b]	6.30[a] 6.53[b]
CO	16.98[a] 16.64[b]	14.20[a] 14.50[b]	12.72[a] 12.29[b]
CH_4	4.43[a] 4.67[b]	3.94[a] 4.06[b]	3.88[a] 4.22[b]
CO_2	17.49[a] 17.99[b]	18.16[a] 18.16[b]	19.21[a] 18.24[b]
LHV (MJ/m^3)	4.37	3.89	3.67
CGE (%)	49.63	47.89	46.43
CCE (%)	90.68	95.48	95.76
Gas yield (m^3/h)	3.93	4.17	4.55
Tar yield (g/m^3)	16.78	12.45	7.24

785 Online analysis[a]

786 GC analysis[b]

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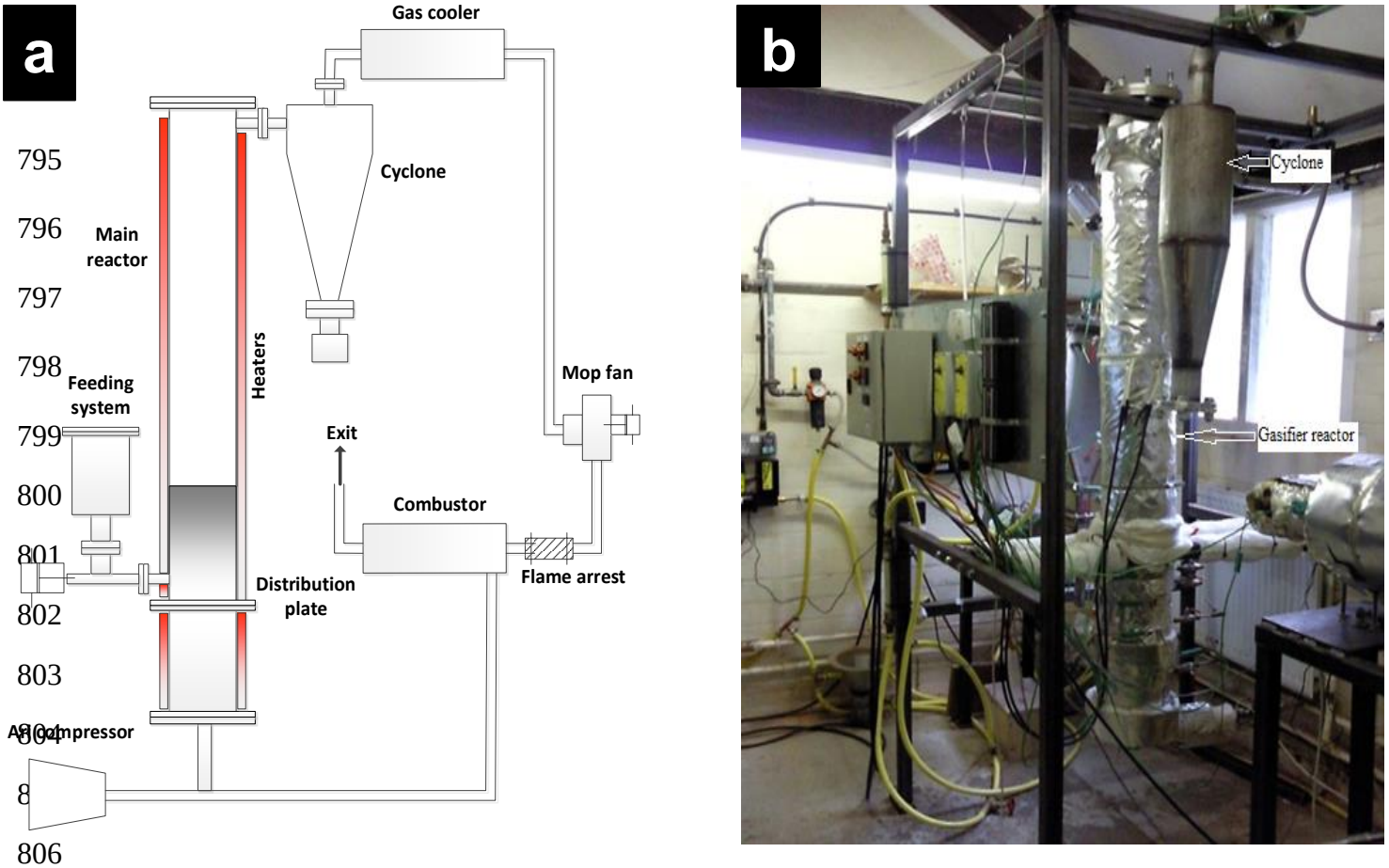
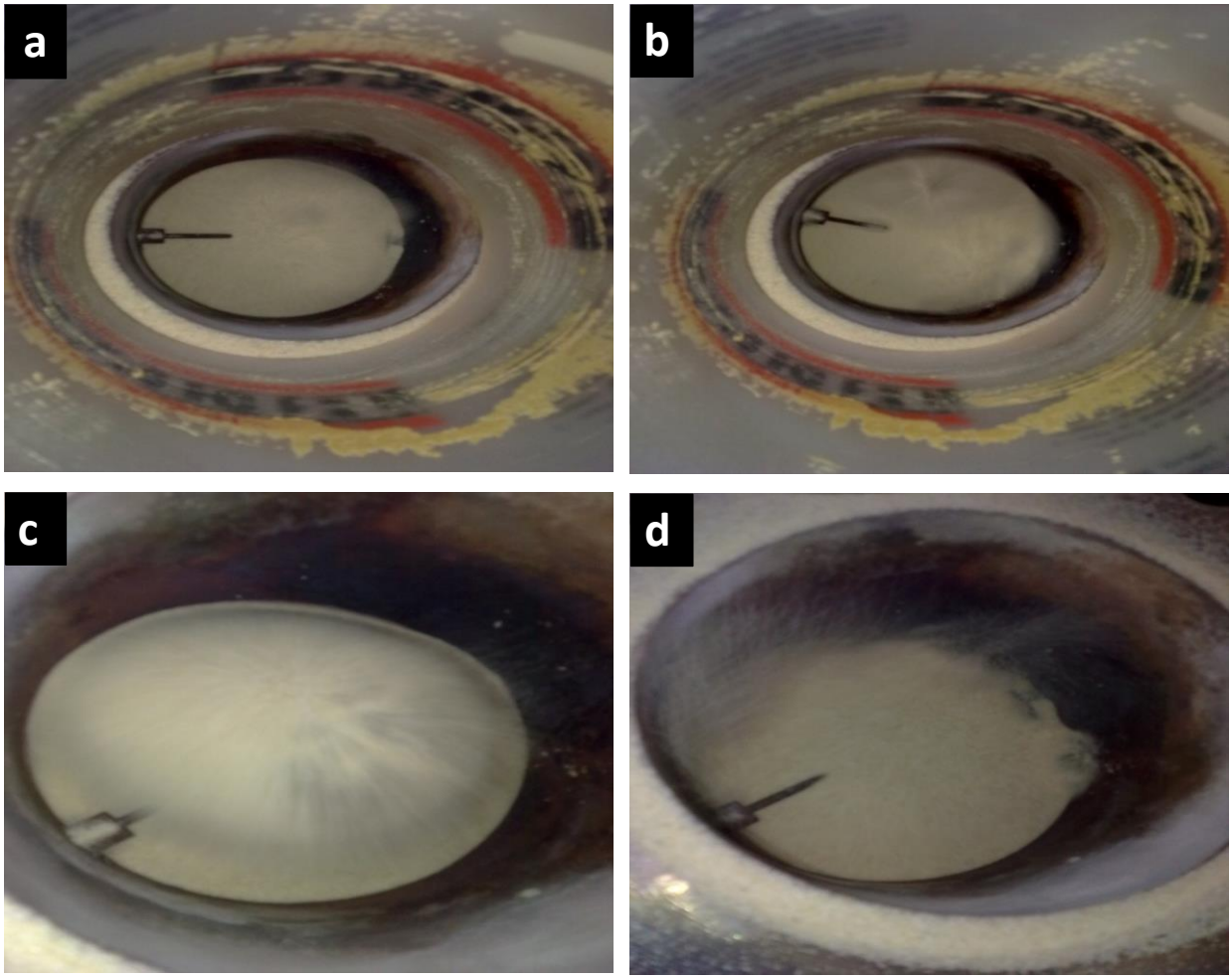


Fig. 1. (a) Schematic diagram of bubbling fluidized bed biomass gasifier; (b) Experimental set up of bubbling fluidized bed reactor and cyclone.



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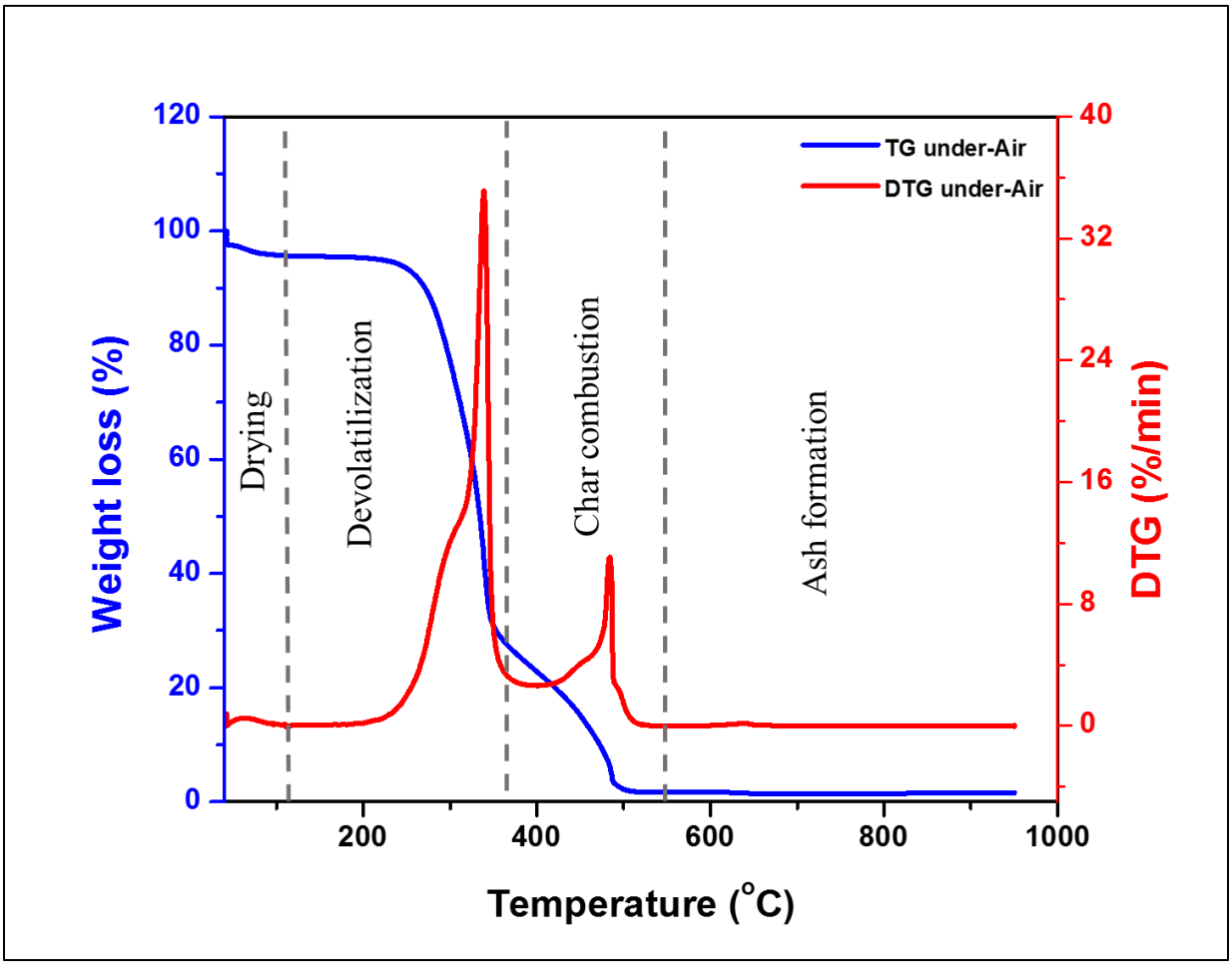

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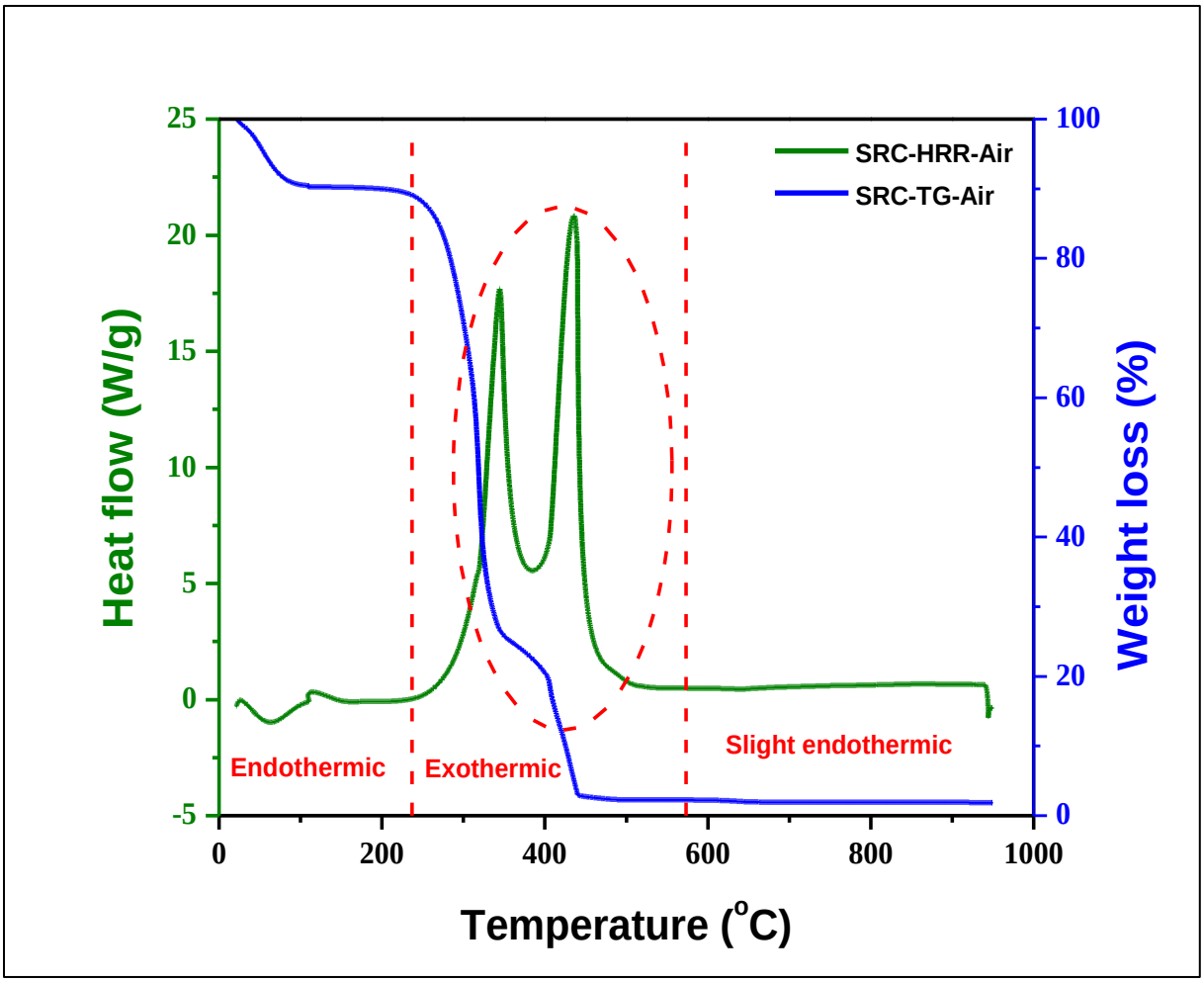

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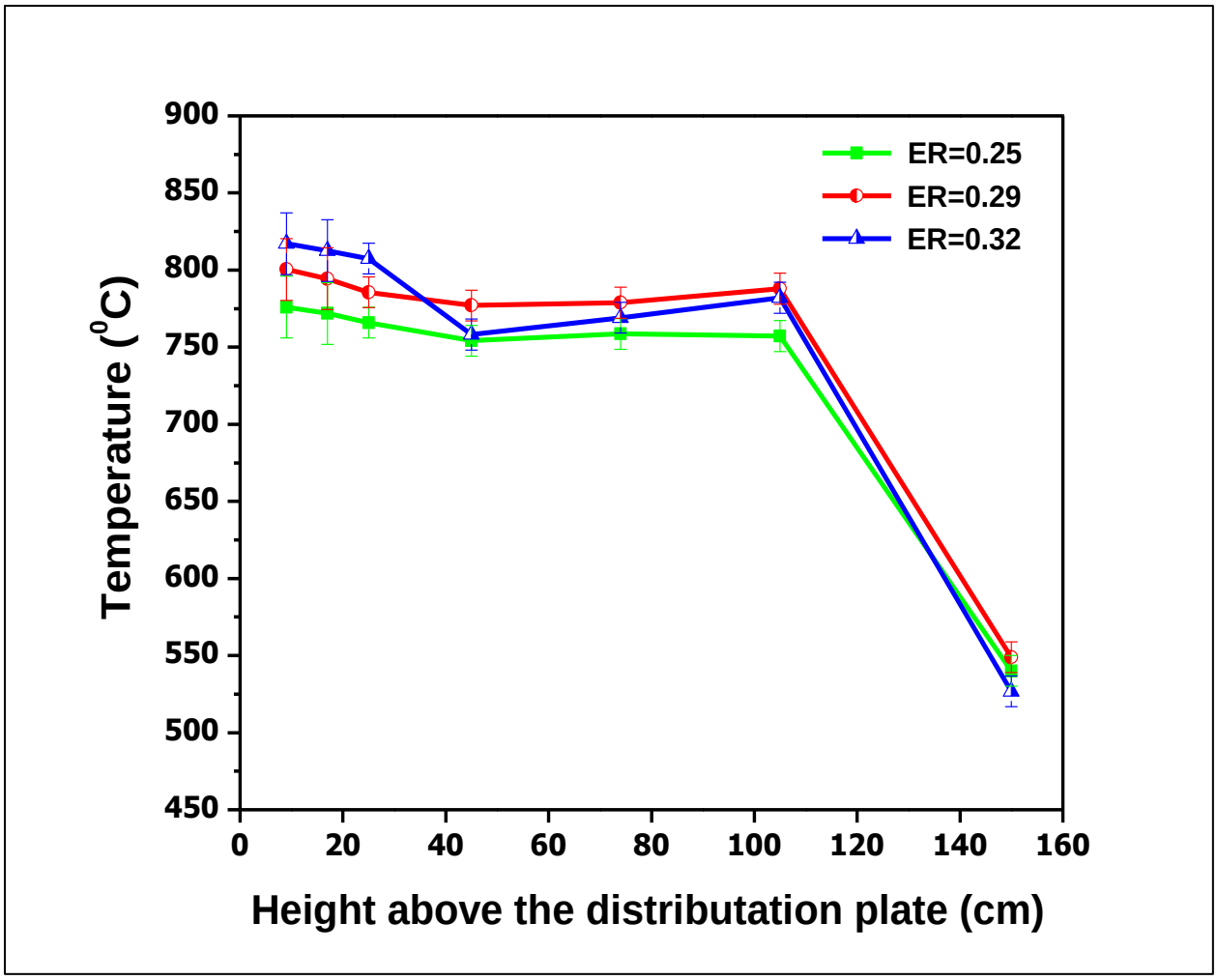

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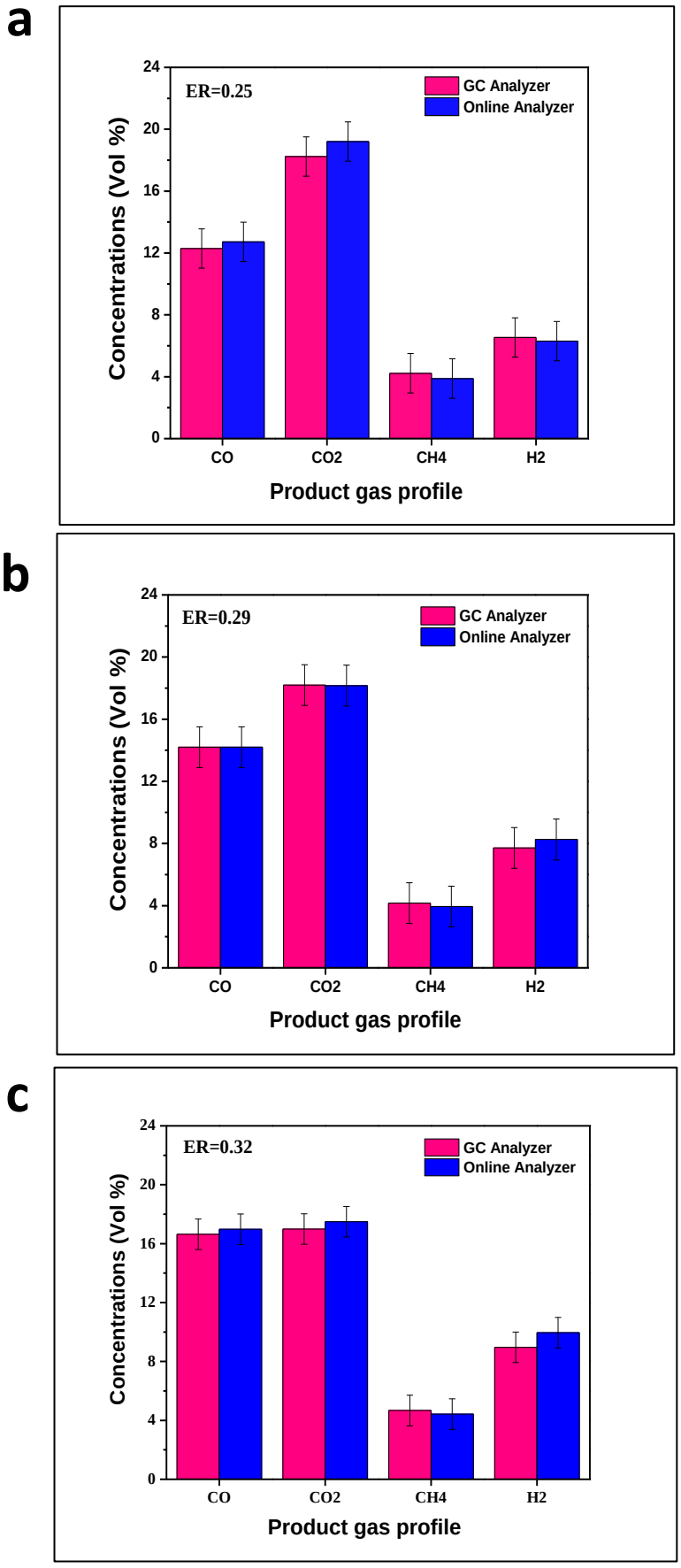
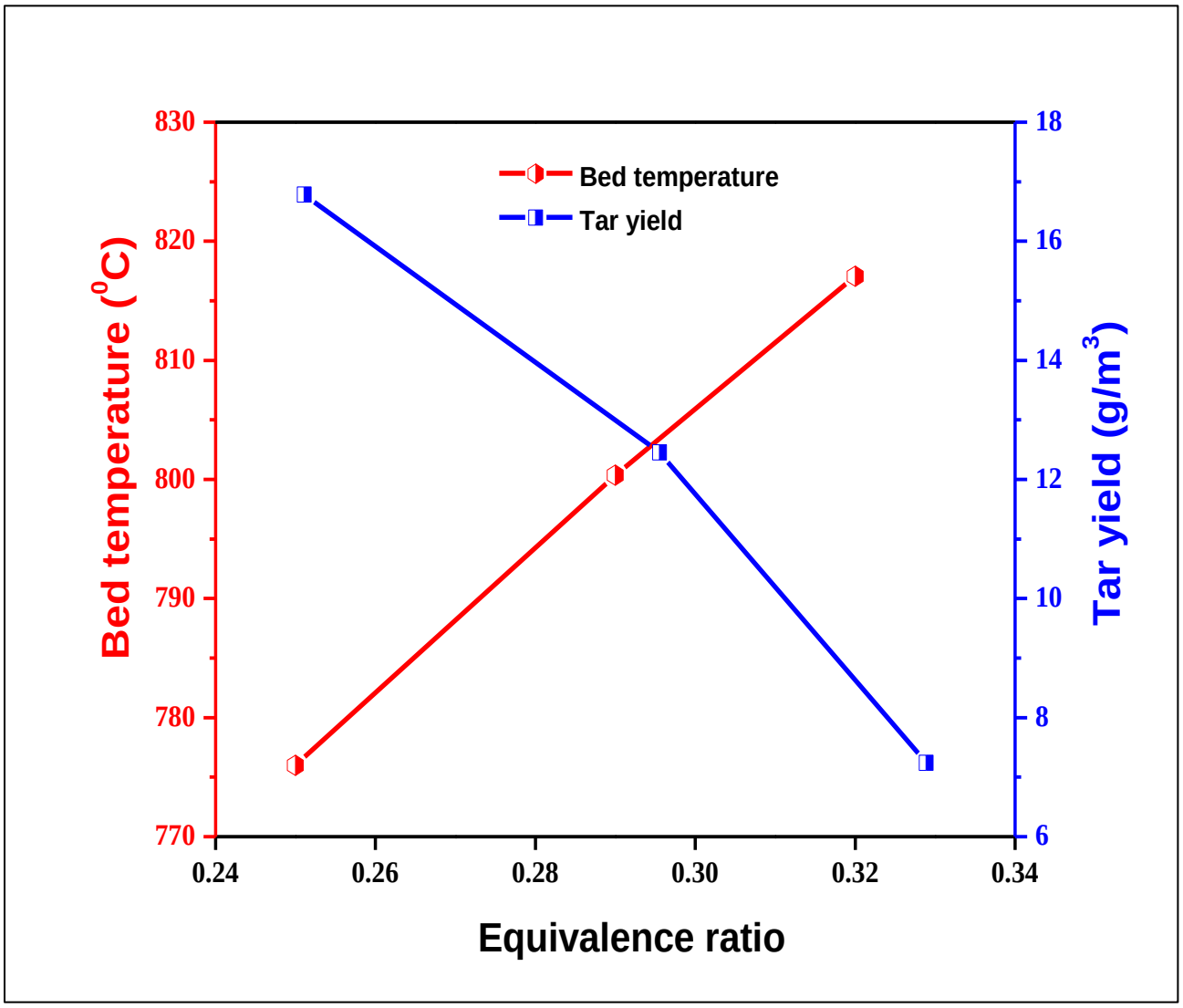


Fig. 6. Effect of different equivalence ratios on the product gas concentrations along with the comparison of GC and online analysis; (a) 0.25 ER, (b) 0.29 ER and (c) 0.32 ER.



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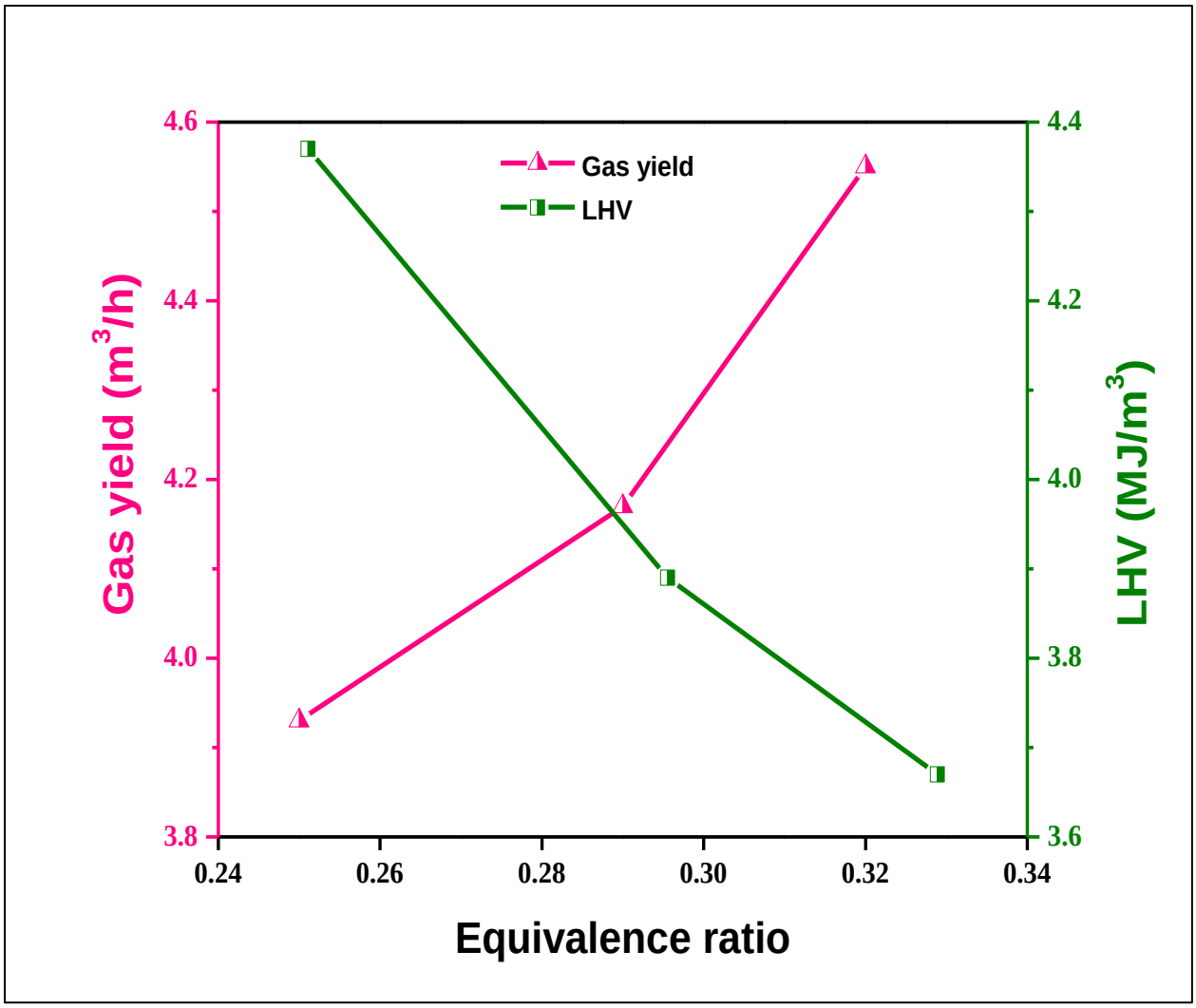

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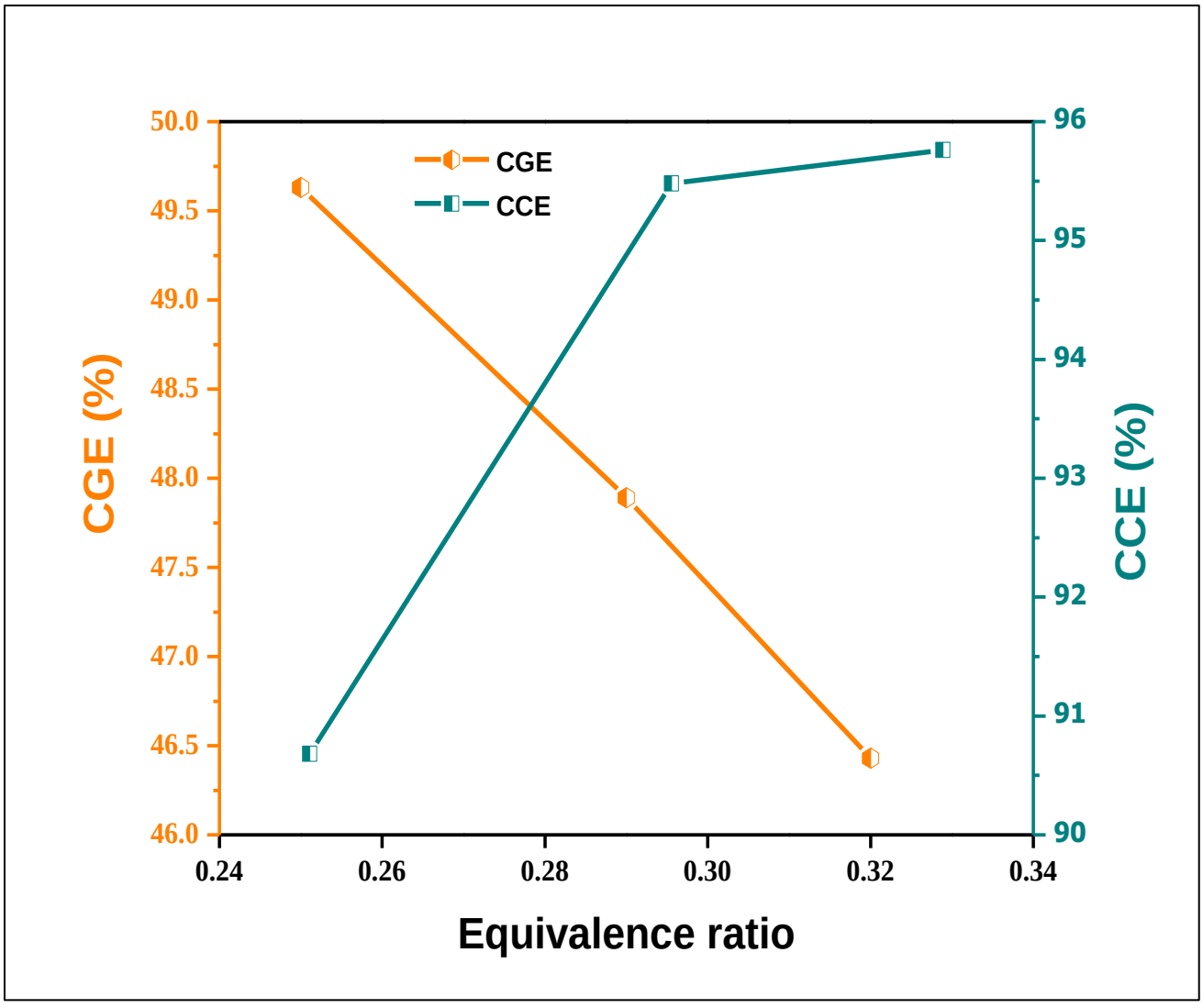

Fig. 9. Effect of equivalence ratio (ER) on CGE and CCE.