



A comparative study on gaseous fuel generation capability of biomass materials by thermo-chemical gasification using stoichiometric quasi-steady-state model

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Abstract The present work investigates the potential of different biomasses for air–steam gasification using thermodynamic equilibrium model. A stoichiometric thermodynamic equilibrium model considering char conversion and tar formation is developed using MATLAB software to compare the gasification performance of different biomasses in terms of product gas yield, heating value and energy efficiency. Regression analysis is performed and correlations for the yield of different syngas constituents are developed in terms of temperature, steam to biomass ratio and equivalence ratio for all the biomasses. For a temperature of 1000 K, steam to biomass ratio of unity and equivalence ratio of 0.25, maximum mole fraction of hydrogen (14.89 %) was obtained from rubber seed shell with a lower heating value and gas yield of 4.71 MJ/Nm³ and 1.18 Nm³/h, respectively.

Keywords Hydrogen · Equilibrium model · Biomass gasification · Char

Introduction

Thermo-chemical gasification is one of the practical methods to extract hydrogen from renewable energy sources such as biomass. Being a clean fuel, use of hydrogen is a potential solution for problems associated with fossil fuel depletion and global warming. Hydrogen yield from biomass gasification depends on many factors

such as type of biomass, moisture content, operating conditions, gasifying agents etc. Effect of different gasifying agents on syngas composition was analysed by Gil et al. [1] and found that compared to steam, tar yield and lower heating value (LHV) of syngas are lower when air is used as the gasifying agent. Another important factor that affects the gasification performance is the type of gasifier used. Warnecke [2] made a comparison of fixed and fluidised bed gasifiers and concluded that in spite of high dust content in the product gas and low ash melting point, fluidised bed gasifiers are attractive for their high heat and mass transfer, favourable solid–gas contact, temperature controllability etc.

Biomass gasification can be investigated using mathematical models when large-scale experimental studies are expensive and difficult to conduct. Extensive reviews on biomass gasification models were presented by Baruah and Baruah [3] and Puig-Arnavat et al. [4]. Among the available gasification models, thermodynamic equilibrium model (TEM) serves as an effective preliminary tool to analyse the effect of different feed stocks and operating parameters on biomass gasification. In general, equilibrium modelling can be achieved through two distinct approaches namely, stoichiometric and non-stoichiometric [5]. Even though both the approaches are conceptually similar, they differ in procedure, as the latter one is comparatively complicated in procedure. Thus, stoichiometric thermodynamic equilibrium models were formulated by many researchers to investigate biomass gasification [6–13]. Zainal et al. [14] successfully demonstrated air gasification of biomass using equilibrium constant expressions of water gas shift reaction and methanation reaction in terms of gasification temperature. TEMs can be modified to improve its prediction accuracy by considering char conversion, tar formation and

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incorporating suitable coefficients to equilibrium constants. Modification of TEM to augment its prediction accuracy was demonstrated by Jarungthammachote and Dutta [15] and Huang and Ramaswamy [16] for air gasification, Loha et al. [17] for steam gasification and Lim and Lee [18] for air–steam gasification. Barman et al. [19] considered tar as a compound containing carbon, hydrogen and oxygen in air gasification. Azzone et al. [20] considered char conversion as a function of equivalence ratio (ER) in air–steam gasification, whereas Lim and Lee [18] expressed it as a function of equivalence ratio and temperature. Abuadula et al. [13] included tar as benzene and unreacted char as 5 % of biomass carbon content in steam gasification model. Ng et al. [9] considered char as solid carbon and tar as a compound containing carbon, hydrogen and oxygen along with correction factors for equilibrium constants as temperature dependent. Nguyen et al. [21] developed a three-stage quasi-equilibrium model for steam gasification of biomass where the deviation from thermodynamic equilibrium was reduced using empirical relations. Application of engineering equation solver (EES) in the modelling of biomass gasification was demonstrated by Puig-Arnavat et al. [22]. Deviation of this model from pure equilibrium is minimised by considering char and tar, pyrolysis, heat loss in pyrolysis, particles leaving the gasifier and setting the amount of CH₄ produced. Present work deals with the stoichiometric modelling of air–steam gasification considering tar and char and its application to compare the effect of different feed stocks in biomass gasification. The present work deals with the comparison of gasification process with different feed stocks in terms of syngas composition, gas yield and first law efficiency.

Table 1 Proximate analysis result of biomass samples

Feed stock	FC (wt.%)	VM (wt.%)	M (wt.%)	A (wt.%)
Rice husk	12	58	12	18
Coconut shell	17	71	8	4
Saw dust	16	76	7	1
Coir pith	20	57	10	13
Rubber seed	24	51	11	14

Table 2 Ultimate analysis result of biomass samples

Feed stock	N (wt.%)	C (wt.%)	S (wt.%)	H (wt.%)	O (wt.%)
Rice husk	2.43	34.35	0.31	5.22	57.66
Coconut shell	0.26	45.61	0.34	5.61	48.16
Saw dust	0.19	46.46	0	5.82	47.51
Coir pith	0.60	44.08	0	4.09	51.21
Rubber seed	2.13	41.11	0.27	6.60	49.88

Characterisation of biomass sample

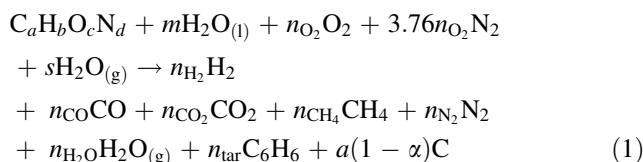
Knowledge of chemical and physical properties of biomass, which serves as an essential pre-requisite of any biomass gasification study, can be obtained through characterisation tests. Characterisation tests were conducted on locally available biomasses in Calicut, Kerala, India such as rice husk, coconut shell, saw dust, coir pith and rubber seed shell [23]. The proximate and ultimate analyses results of the biomasses are given in Tables 1 and 2, respectively.

Model development

The objective of present work is to develop a thermodynamic equilibrium model to analyse the influence of different locally available feed stocks on gasification process. A stoichiometric thermodynamic equilibrium model for air–steam biomass gasification, incorporating char and tar, is developed based on the following assumptions,

- Gasifier is considered as a steady-state system with uniform temperature and pressure throughout.
- All the gases behave ideally.
- Gases except H₂, CO, CO₂, CH₄ and N₂ are considered dilute.
- N₂ is considered as inert in the entire process.
- Biomass is considered to be made up of Carbon, Hydrogen, Oxygen and Nitrogen.
- Steam is supplied at superheated condition of 1 bar and 300 °C.
- Tar and char are assumed as benzene and graphitic carbon, respectively.

By considering chemical formula of feedstock as C_aH_bO_cN_d, global gasification reaction can be written as:



where, *a*, *b*, *c* and *d* are the number of atoms of carbon, hydrogen, oxygen, and nitrogen per mole of dry and ash free biomass. *m* is the number of moles of moisture per mole of

biomass. n_{O_2} , n_{H_2} , n_{CO} , n_{CO_2} , n_{CH_4} , n_{N_2} , n_{H_2O} and n_{tar} are the stoichiometric coefficients of oxygen, hydrogen, carbon monoxide, carbon dioxide, methane, nitrogen, steam and tar, respectively. s is the number of moles of steam supplied and α is the carbon conversion factor, expressed as a function of temperature and equivalence ratio [18].

$$\alpha = 0.901 + 0.439 \times \left(1 - e^{(-ER + 0.0003T)}\right) \quad (2)$$

Tar yield as a weight percentage of total gasification products is given by [13],

$$\text{Tar}_{wt.\%} = 35.98e^{(-0.00298T)} \quad (3)$$

Total weight of the gasification product is obtained by applying mass balance to the global reaction between the reactants and the products. So mass of tar yield (m_{tar}) is given by,

$$m_{tar} = \frac{\text{Tar}_{wt.\%}}{100} (\text{biomass feed} + \text{SBR} \times \text{biomass feed} + \text{moisture in biomass} + \text{air supplied}) \quad (4)$$

where SBR is steam to biomass ratio, defined as the ratio of mass flow rate of steam to mass flow rate of biomass.

Five separate equations are required to determine the five unknown constituents of product gas. These equations can be developed from the elemental balance of C, H, O and N and from the equilibrium constant expressions [Eqs. (10) and (12)] of water gas shift reaction [Eq. (9)] and hydrogasification reaction [Eq. (11)], respectively.

Elemental balance

Carbon balance,

$$n_{CO} + n_{CO_2} + n_{CH_4} + 6n_{tar} + a(1 - \alpha) - a = 0 \quad (5)$$

Hydrogen balance,

$$2n_{H_2} + 4n_{CH_4} + 2n_{H_2O} + 6n_{tar} - b - 2m - 2s = 0 \quad (6)$$

Oxygen balance,

$$n_{CO} + 2n_{CO_2} + n_{H_2O} - c - 2n_{O_2} - m - s = 0 \quad (7)$$

Nitrogen balance,

$$2n_{N_2} - d - 2 \times 3.76n_{O_2} = 0 \quad (8)$$

Water gas shift reaction



Considering equilibrium constant K_1 for water gas shift reaction,

$$K_1 = \frac{n_{CO_2}n_{H_2}}{n_{CO}n_{H_2O}} \quad (10)$$

Hydrogasification reaction,



Considering equilibrium constant K_2 for hydrogasification reaction,

$$K_2 = \frac{n_{total}n_{CH_4}}{(n_{H_2})^2} \quad (12)$$

For ideal gas, K_1 and K_2 can be expressed as a function of temperature [14], given by Eqs. (13) and (14).

$$K_1 = \exp\left(\frac{5878}{T} + 1.86 \ln T - 0.27 \times 10^{-3}T - \frac{58200}{T^2} - 18\right) \quad (13)$$

$$K_2 = \exp\left(\frac{7082.842}{T} - 6.567 \ln T + \frac{7.467 \times 10^{-3}}{2}T - \frac{2.167 \times 10^{-6}}{6}T^2 + \frac{0.0702 \times 10^{-5}}{2T^2} + 32.541\right) \quad (14)$$

Thus, equilibrium composition of the product gas is obtained by simultaneously solving three linear equations [Eqs. (5)–(7)] and two non-linear equations [Eqs. (10) and (12)] in MATLAB platform using Newton–Raphson method.

Lower heating value of the dry product gas is estimated from the gas composition and is expressed in volume basis as [24],

$$\text{LHV} = 10.79Y_{H_2} + 12.26Y_{CO} + 35.81Y_{CH_4} \quad (15)$$

Gasification efficiency of the process is given by,

$$\eta_{\text{gas}} = \frac{\text{Energy content in the product gas}}{\text{Energy content in biomass} + \text{energy content in steam}} \quad (16)$$

Model validation

Prediction capability of the developed model is checked by comparing the predicted gas composition with experimental results [25]. The deviation of model result from experimental values is quantified using the statistical parameter of root mean square (RMS) error,

$$\text{RMS} = \sqrt{\frac{\sum (X_e - X_p)^2}{N}} \quad (17)$$

where X_e , X_p and N are experimental data, predicted value and number of observations, respectively. An average RMS = 4.7205 is obtained when eight sets of experimental results are compared with their corresponding theoretical predictions, for a gasifier pressure of 1 bar, as given in Fig. 1.



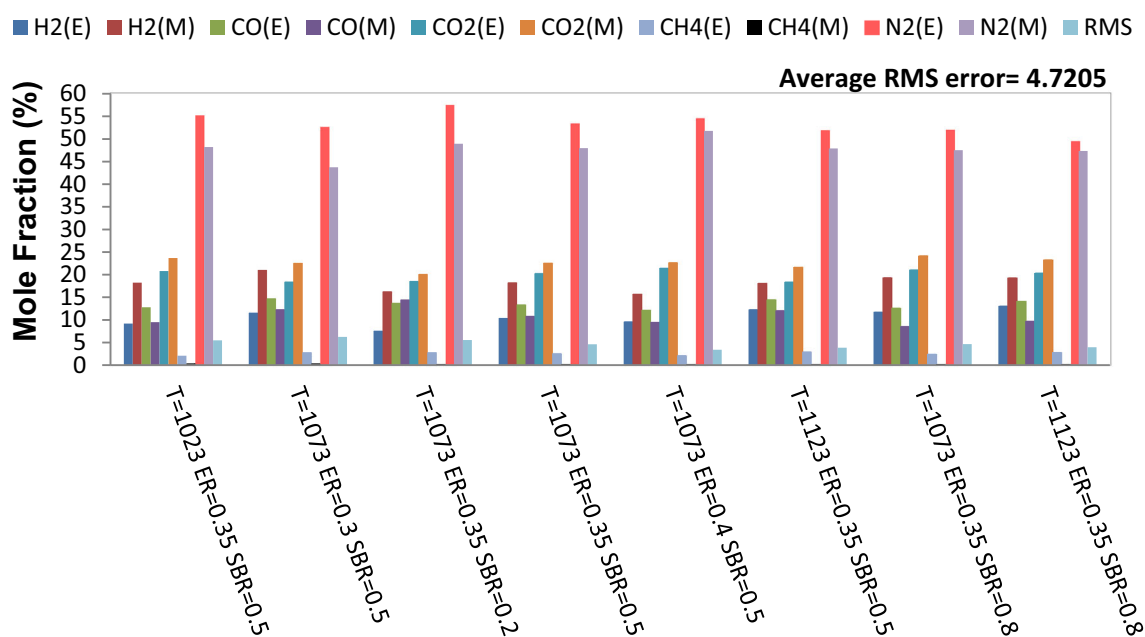


Fig. 1 Comparison between experimental and model results. *E* experimental result, *M* model result

Model modification and non-equilibrium factors

It is observed that CO_2 and CH_4 concentrations were under-predicted and H_2 and CO concentrations were over-predicted by the present model from the experimental values. Melgar et al. [26] made similar observation when the syngas composition predicted by their model is compared with the experimental results of Jayah et al. [27]. Same results were obtained when the model predicted gas composition [28–31] is compared with the experimental results of Rapagna et al. [32] and Hofbauer et al. [33]. The prediction accuracy of the model is improved by multiplying K_1 and K_2 with suitable coefficients C_1 and C_2 , expressed as functions of ER [18].

$$C_1 = p_1 e^{p_2 ER} \quad (18)$$

$$C_2 = p_3 - p_4 ER \quad (19)$$

Average RMS error is checked by varying the values of p_1 , p_2 , p_3 , p_4 , C_1 and C_2 , and the values corresponding to minimum average RMS error are selected for the model. Average RMS error is reduced from 4.7205 to a minimum of 1.3713 by introducing suitable values of C_1 and C_2 in the model ($C_1 = 0.12e^{3.2 ER}$ and $C_2 = 41 - 50ER$). Figure 2 shows a fair agreement between experimental results and that obtained from modified model.

Model application

The stoichiometric quasi-equilibrium model developed is used to predict the influence of key process parameters such as gasification temperature, steam to biomass ratio

and equivalence ratio (ER) on syngas composition, heating value and energy efficiency. Gasification performance of different biomasses, at a gasifier pressure of 1 bar, is compared by keeping mass flow rate as 1.0 kg/h and varying temperature, SBR and ER in the ranges of 900–1500 K, 0–3.5, 0.15–0.45, respectively.

Results and discussion

Effect of gasification temperature on product gas composition

Effect of gasification temperature on product gas composition is depicted through Fig. 3. Among all the feed stocks, rubber seed has the maximum potential for hydrogen generation under same operating conditions. For rubber seed shell, coconut shell and saw dust, H_2 mole fraction is found to be increasing with temperature up to 1300 K, and remains unchanged thereafter. However, for rice husk and coir pith H_2 concentration decreases with increase in temperature beyond 1300 K. For all feed stocks, the increase in H_2 concentration is more pronounced at lower temperature ranges (900–1300 K). Similar variation on H_2 concentration with temperature was observed by Lv et al. [34]. This trend of H_2 is mainly due to the reversal of exothermic water gas shift reaction at higher temperature ranges. For unit SBR and $ER = 0.25$, maximum hydrogen concentration values for rice husk and coir pith are found to be 17.67 and 16.92 %, respectively, at 1300 K. Figure 3b shows that coir pith, rice husk and saw dust have almost

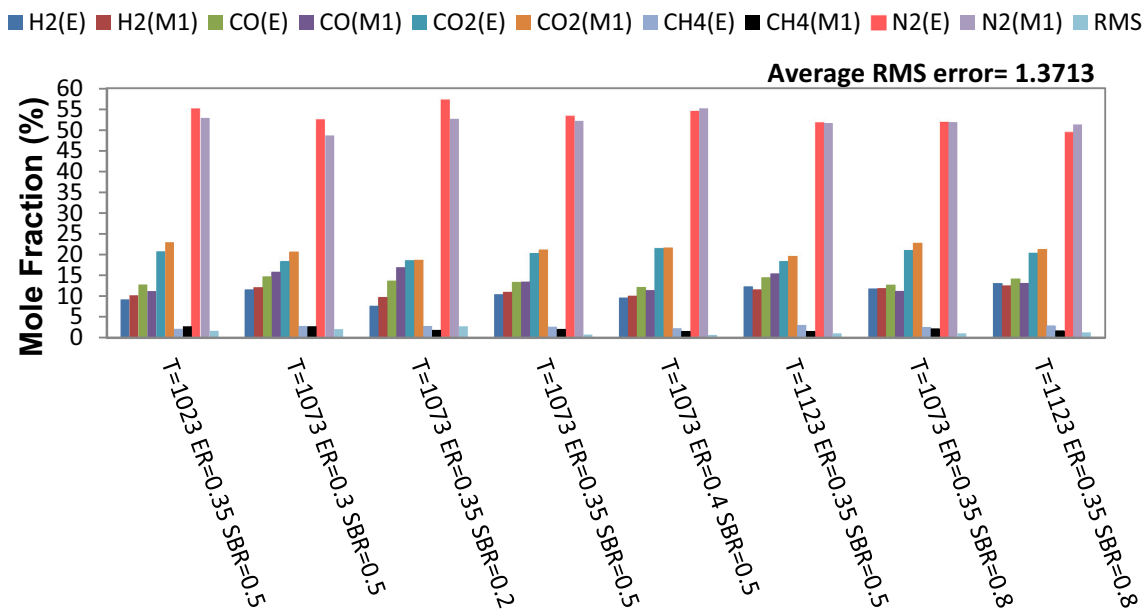
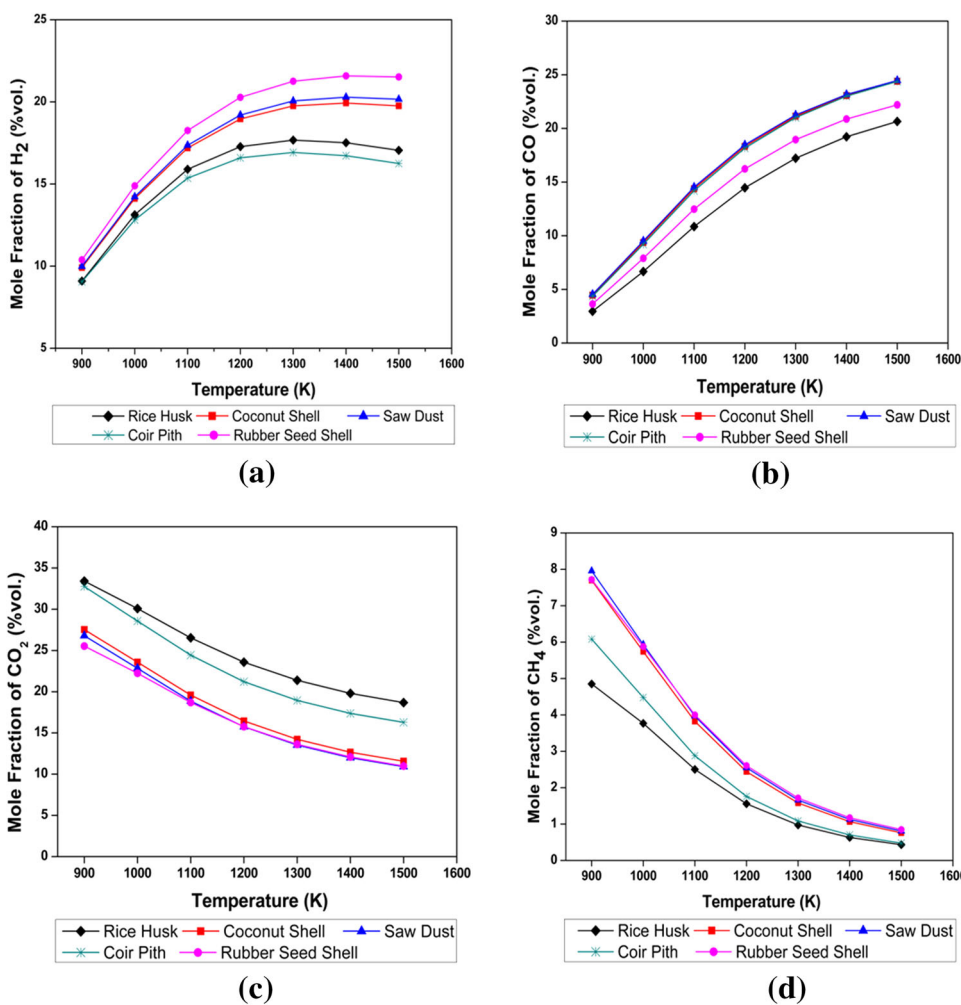


Fig. 2 Comparison between experimental and modified model results. *E* experimental result, *M1* modified model result

Fig. 3 Effect of temperature on syngas composition (SBR = 1, ER = 0.25): **a** H₂ mole fraction, **b** CO mole fraction, **c** CO₂ mole fraction, **d** CH₄ mole fraction



same CO mole fraction for the entire temperature range. From Fig. 3b, c, it is clear that CO and CO₂ concentrations, respectively, increase and decrease with temperature.

This variation indicates the reversal of water gas shift reaction in gasification process. Decrease in CH₄ concentration with temperature, as shown in Fig. 3d, is due to the effect of shifting of exothermic hydrogasification reaction towards the reactant side.

Effect of ER on product gas composition

Figure 4 shows the effect of ER on different gas constituents in the product. All the gas constituents except CO₂ decrease with ER. This is due to shifting of the process more towards combustion at higher ER values.

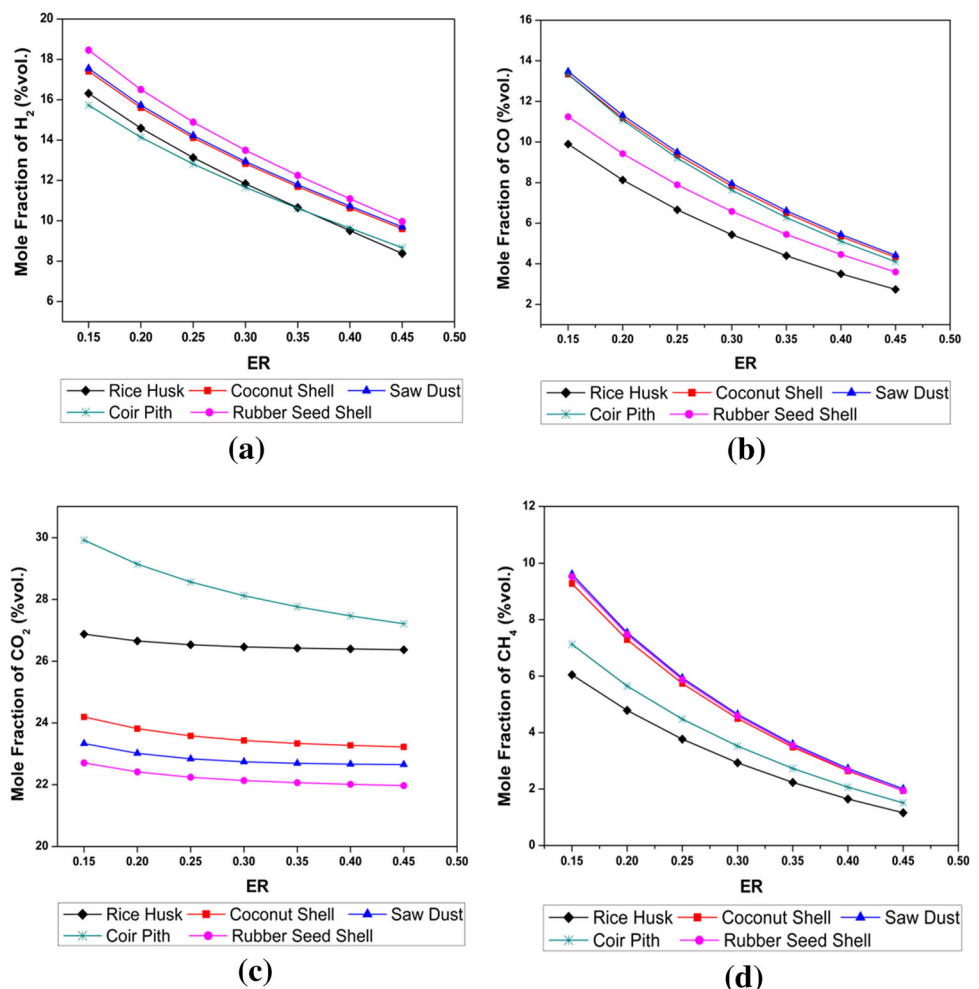
Even though the number of moles of CO₂ increases, there is a decrease in CO₂ mole fraction with ER. This is attributed to the higher rate of increase of total number of moles of gas constituents, due to the addition of N₂, compared to the increase in number of moles of CO₂. The

increase in number of moles of CO₂ with ER is shown in Fig. 5.

Effect of SBR on product gas composition

The influence of steam addition on product gas composition is depicted through Fig. 6. Increase in H₂ concentration with SBR is due to the effect of water gas shift reaction. The increase in H₂ mole fraction with SBR is found to be insignificant beyond a SBR value of 2.5 for all the feed stocks except coir pith, for which it decreases slightly beyond SBR = 2.5. The decrease and increase in CO and CO₂ concentrations with SBR is depicted through Fig. 6b, c, respectively. This is due to the effect of water gas shift reaction. Influence of methanation reaction on gasification process is reflected in the decrease of CH₄ mole fraction with SBR as shown in Fig. 6d. Similar effects of decrease in CO and increase in H₂ and CO₂ concentrations with SBR are observed in the literature [35] at a temperature of 988 K and ER of 0.12.

Fig. 4 Effect of ER on syngas composition ($T = 1000$ K, SBR = 1): **a** H₂ mole fraction, **b** CO mole fraction, **c** CO₂ mole fraction, **d** CH₄ mole fraction



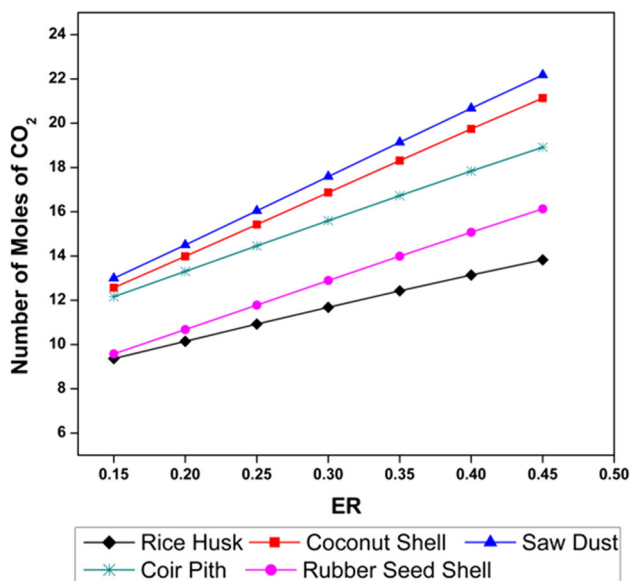


Fig. 5 Effect of ER on number of moles of CO₂ ($T = 1000\text{ K}$, $\text{SBR} = 1$)

Fig. 6 Effect of SBR on syngas composition ($T = 1000\text{ K}$, $\text{ER} = 0.25$): a H₂ mole fraction, b CO mole fraction, c CO₂ mole fraction, d CH₄ mole fraction

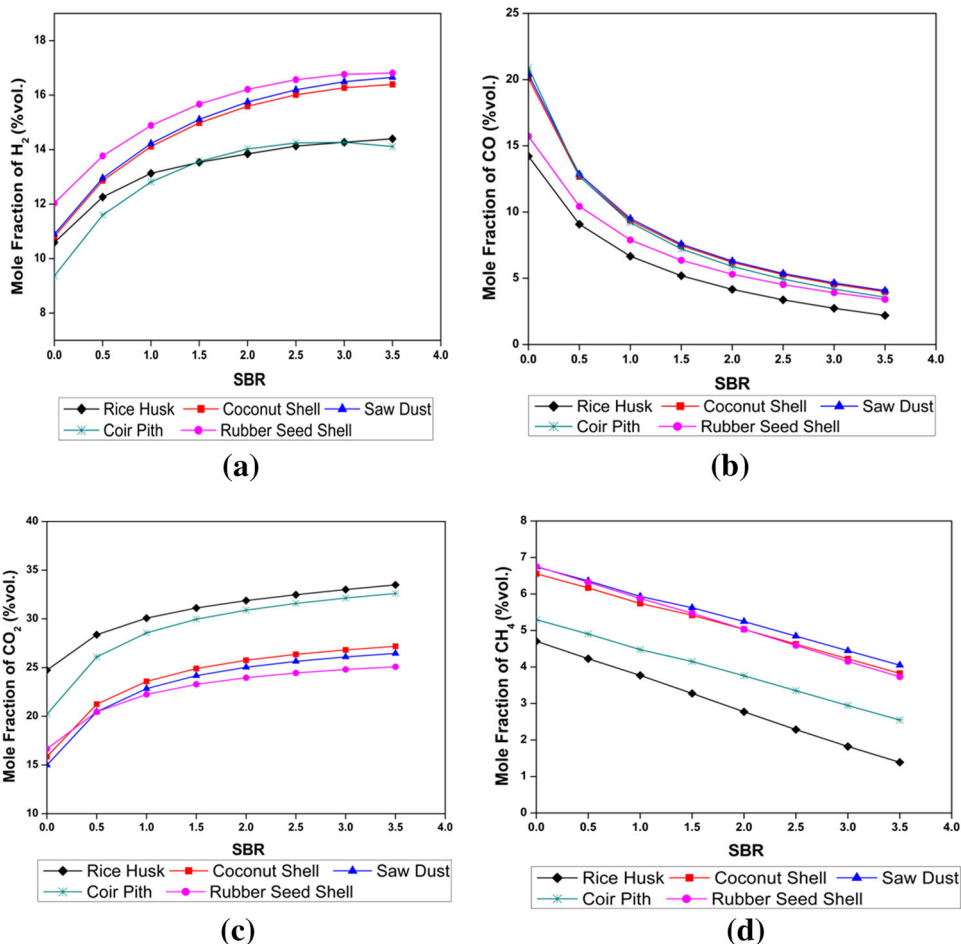


Figure 7 shows the decrease of LHV of syngas produced from all the feed stocks with ER. This is due to the shifting of partial oxidation to combustion at higher ER values. Lower heating value of syngas decreases as the feed stock is changed in the order saw dust, coconut shell, rubber seed shell, coir pith and rice husk.

Effect of temperature, SBR and ER on efficiency

Influence of temperature, SBR and ER on gasification efficiency is illustrated through Fig. 8. The increase in gasification efficiency with temperature is due to the hike in values of lower heating values of syngas at higher temperatures.

Efficiency is found to decrease with both SBR and ER. This decrease with ER is due to the reduced lower heating value of the product gas, whereas the reason for efficiency degradation with SBR is due to the increased energy input in the form of steam.

Comparison of gasification performance of different biomass samples

Comparison of gas composition, lower heating value and gas yield for different biomasses is shown in Table 3.

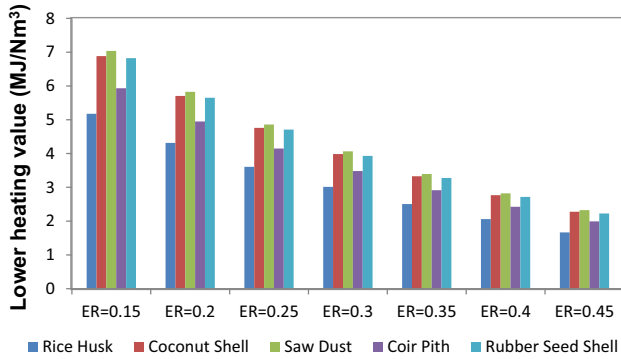


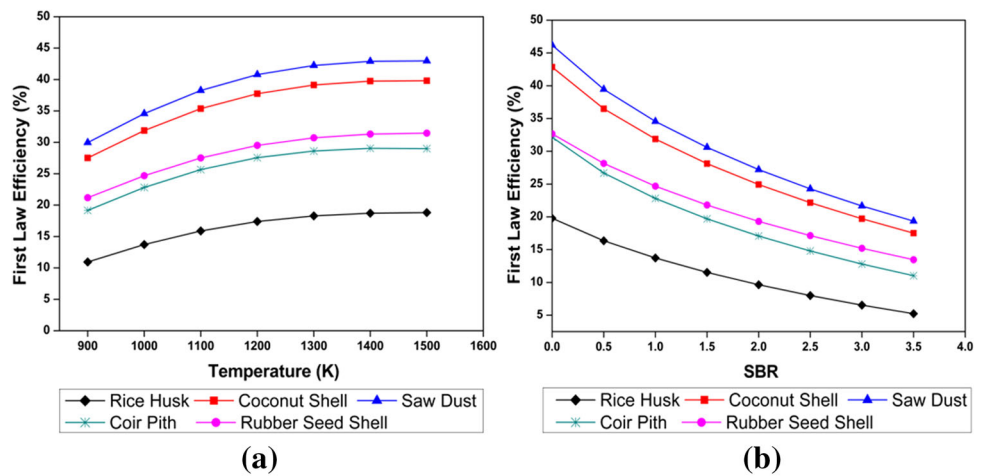
Fig. 7 Effect of ER on lower heating value of product gas ($T = 1000\text{ K}$, $SBR = 1$)

For $T = 1000\text{ K}$, $SBR = 1$ and $ER = 0.25$, rubber seed shell and coir pith have the maximum and minimum potential for hydrogen generation, respectively. It is also observed that rice husk, biomass with maximum oxygen content, yields lowest calorific value syngas and biomass with maximum carbon content, saw dust, contributes to highest gas yield under the same operating conditions. The biomass with largest hydrogen content is found to produce syngas with higher hydrogen concentration.

Regression analysis

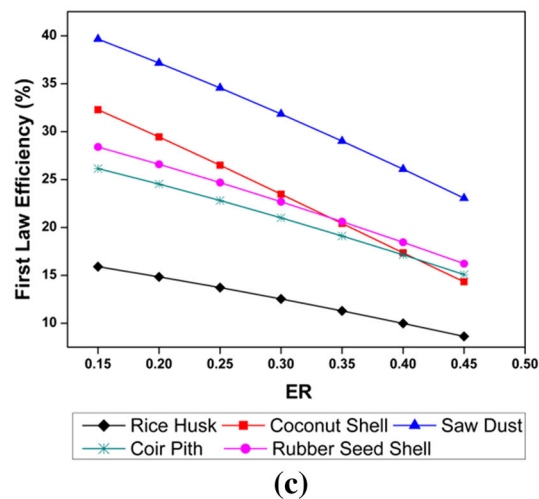
A regression analysis is performed by considering 392 values for each gas constituents, using statistical software Minitab 16, and correlations for the yield of different gas species in terms of gasification temperature, SBR and ER are formulated. The regression equations for the product yield of different biomasses are given in Table 4.

Fig. 8 Effect of process parameters on efficiency ($SBR = 1$, $ER = 0.25$): a temperature, b SBR, c ER



(a)

(b)



(c)

Table 3 Comparison of gas composition, LHV and gas yield for different biomasses

Sl. no.	Biomass	Product gas composition (% dry basis)					LHV (MJ/Nm ³)	Gas yield (Nm ³ /h)
		H ₂	CO	CO ₂	CH ₄	N ₂		
1.	Rice husk	13.13	6.66	30.08	3.77	46.36	3.61	0.81
2.	Coconut shell	14.12	9.37	23.58	5.74	47.19	4.76	1.46
3.	Saw dust	14.22	9.50	22.84	5.94	47.50	4.86	1.58
4.	Coir pith	12.82	9.21	28.57	4.48	44.93	4.15	1.14
5.	Rubber seed shell	14.89	7.89	22.24	5.88	49.10	4.71	1.18

Operating conditions: $T = 1000$ K, $SBR = 1$ and $ER = 0.25$

Table 4 Regression equations for product yield of different biomasses

Sl. no.	Biomass	Regression equation (% vol.)	R ² (%)
1.	Rice husk	H ₂ = 4.183 + 0.014585 T - 31.38 ER + 1.225 SBR	80.64
		CO = -2.60234 + 0.0258743 T - 38.8078ER - 3.86095 SBR	90.94
		CO ₂ = 47.2267 - 0.02184 T + 0.977798 ER + 2.71341 SBR	88.83
		CH ₄ = 10.9034 - 0.00516197 T - 8.39252 ER - 0.467673SBR	71.61
2.	Coconut shell	H ₂ = 5.86767 + 0.015906 T - 38.4606 ER + 1.73205 SBR	85.69
		CO = -0.622431 + 0.0272745 T - 39.6843 ER - 3.88895 SBR	90.33
		CO ₂ = 38.5471 - 0.0217425 T + 6.95273 ER + 2.7336 SBR	87.05
		CH ₄ = 17.0464 - 0.00838929 T - 13.2497 ER - 0.287159 SBR	78.08
3.	Saw dust	H ₂ = 5.799 + 0.016287 T - 39.10 ER + 1.7577 SBR	86.09
		CO = -0.535347 + 0.0272512 T - 39.5014 ER - 3.88701 SBR	90.43
		CO ₂ = 37.4687 - 0.0215885 T + 7.64056 ER + 2.73769 SBR	87.20
		CH ₄ = 17.832 - 0.00877337 T - 13.9551 ER - 0.291162 SBR	78.85
4.	Coir pith	H ₂ = 5.704 + 0.012757 T - 32.43 ER + 1.6595 SBR	80.90
		CO = 0.500249 + 0.0272316 T - 41.7527 ER - 4.22765 SBR	89.02
		CO ₂ = 45.3196 - 0.0226152 T + 2.14251 ER + 2.96163 SBR	85.93
		CH ₄ = 12.6657 - 0.00630763 T - 9.49212 ER - 0.259665 SBR	73.95
5.	Rubber seed shell	H ₂ = 5.687 + 0.017935 T - 41.11 ER + 1.4814 SBR	86.82
		CO = -3.025 + 0.025818 T - 34.90 ER - 3.2488 SBR	91.22
		CO ₂ = 36.110 - 0.019936 T + 7.139 ER + 2.2694 SBR	87.70
		CH ₄ = 17.420 - 0.008346 T - 13.809 ER - 0.3868 SBR	78.20

Conclusions

A stoichiometric thermodynamic equilibrium model for air–steam gasification of biomass considering char conversion and tar formation was developed to compare the gasification performance of different locally available feed stocks. After comparing the model results with experimental results for product gas composition, its prediction accuracy was improved by incorporating suitable coefficients with equilibrium constants. The resulting quasi-equilibrium model was used to compare the gasification performance of different feed stocks such as rice husk, coconut shell, saw dust, coir pith and rubber seed shell in

terms of product gas composition, lower heating value, gas yield and gasification efficiency. Regression equations for the yield of different syngas constituents were developed for all the biomasses. For $T = 1000$ K, $SBR = 1$ and $ER = 0.25$, maximum mole fraction of hydrogen (14.89 %) was obtained from rubber seed shell with a lower heating value and gas yield of 4.71 MJ/Nm³ and 1.18 Nm³/h, respectively.

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