INVESTIGATION OF GASIFICATION REACTIONS AND H_2/CO RATIO ANALYSIS FOR RICE HUSK AIR GASIFICATION SIMULATION

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The syngas generated during gasification process is used for industrial process heat, power generation and chemical feedstock production. Gas composition, H_2/CO and CO/CO_2 ratios are the deciding factors for producer gas end-use. In the present work, rice husk air gasification simulation is carried out by non-stoichiometric equilibrium model using Gibbs free energy minimization by FactSage 6.3 software. Influence of gasification reactions and effect of temperature, equivalence ratio on gas composition was studied. The analysis was carried out to study the effect of operating conditions on producer gas heating value and gasification efficiency at various H_2/CO ratios.Increase in temperature improves H_2 and CO formation in water gas reaction and reduction of H_2 formation was observed in water gas shift reaction due to endothermic behavior change. For all the values of the equivalence ratio (ϕ),an increase in temperature reduces the H_2/CO and enhances the CO/CO_2 ratio due to higher CO formation.For a particular ϕ value, maximum gas heating value and gasification efficiency were obtained at low H_2/CO ratio.

Keywords: rice husk, air gasification, gasification simulation, H_2/CO ratio, gas heating value, gasification efficiency.

1. Introduction

Biomass gasification is the thermo-chemical conversion process used to generate producer gas/syngas from solid biomass fuels.Rice husk is largely produced in India; every year 24 million tons of rice husk is produced [1]. The main constituents of producer gas are CO, CO_2 and H_2; gas calorific value and gasification efficiency depends on the composition of producer gas. The gas composition of producer gas depends on fuel property, gasifying medium and operating conditions[2, 3]. High formation of combustible gases leads to improved calorific value and gasification efficiency. Syngas was commonly used in the petrochemical and metallurgical process and H_2/CO ratio of the syngas decides its final use.Syngas with a high H_2/CO ratio is used for hydrogen production; syngas with H_2/CO ratio value of 2 is used for raw material production such as synthetic fuels, oxo-alcohols, ethylene glycol, acetic acid and dimethyl ether. H_2/CO ratio is not a constraint for syngas use in boilers and turbines; CO/CO_2 ratio is the index scale for process completion [4, 5].

Ardebili et al. [4] investigated the influence of operating parameters on low temperature gasification using a mathematical model for raw material production. Fischer-Troph (FT) process is used to convert hydrogen and carbon monoxide mixture into liquid hydrocarbon, the requirement of syngas for the FT process depends on high formation of H_2 and CO. The H_2/CO ratio and CO/CO_2 ratio were used to evaluate the effectiveness of the process. Carbonaceous materials are gasified with steam and oxygen to improve the H_2/CO ratio at low temperature conditions for the conformity of the FT process. Tristantini et al. [5] carried out the feasibility study of syngas with different H_2/CO ratios for FT synthesis using Co/γ-$Al_2O_3$and Co-Re/γ-Al_2O_3.A gas mixture with a low H_2/CO ratio decreased the CO conversion and slightly increased the hydrocarbon formation. The conversion of methane into syngas is known as steam reforming reaction, syngas with high H_2/CO ratio is produced during steam reforming and less CO_2 formation takes place compared to the gasification process.The complete conversion of methane is not possible in reforming reaction due to equilibrium constraints. Cao et al. [6] injected methane into the pyrolysis and gasification zone of the co-gasification reactor to modify the H_2/CO ratio of the syngas for the coal gasification process. Coal gasification reactivity correlated to the gas yield and H_2/CO ratio of syngas depends on the H/C ratio of the coal.

Buragohain et al. [7] have reported optimization of biomass (sawdust, rice husk, bamboo dust) air gasification and air-steam gasification in terms of feedstock for Fischer-Tropsch synthesis and decentralized power generation. Non-stoichiometric equilibrium model (SOLGASMIX) was used for optimization by FactSage software. H_2& CO content in producer gas and H_2/CO ratio were assessed for FT synthesis. The heating value of gas mixture and overall efficiency were analyzed with relevance to decentralized power generation. The gas heating value remained constant after 700°C and operating the gasifier above 700°C leading to energy loss. Low and high equivalence ratio resulted in significant unconverted carbon and CO_2 formation respectively.Syngas is commonly produced from steam reforming of natural gas, partial oxidation of heavy oils and gasification of coal and biomass [8].Syngas production is an important step in the production of derived products; it influences the cost and impact on the environment. Hydrogen and carbon monoxide formation plays an important role in the gasification process. Syngas with H_2/CO ratio of 2 is required for methanol production and FT synthesis; synthesis of ethanol, dimethyl ether and oxo-alcohols requires H_2/CO ratio of 1[7].

In the present work, the producer gas composition of rice husk air gasification was determined at different operating conditions using the non-stoichiometric equilibrium model.Influence of gasification reactions and effect of temperature, equivalence ratio on gas composition was studied.H_2/CO ratio, CO/CO_2 ratio were evaluated; an analysis was carried out to find a suitable operating condition for maximum H_2/CO ratio attainment. Gas heating value and gasification efficiency values were calculated at different H_2/CO ratios for the simulation of rice husk air gasification process.

2. Methodology

Simulation of rice husk air gasification was carried out by non-stoichiometric equilibrium model using FactSage software, Gibbs free energy minimization was employed to find out the gas composition. Many researchers - Li et al. [9], Altafini et al. [10] and Yoshida et al.[11] used equilibrium model to predict the performance of commercial gasifiers and closer agreement with experimental result was experienced.

Equilibrium composition of producer gas was determined by finding the number of atoms (C, H & O) for reactants and the number of carbon, hydrogen and oxygen atoms in each product.Gibbs free energy of formation for each product was evaluated at individual temperature.The minimum thermodynamic potential equation was developed for each product using the Lagrange multiplier method, given by Eq. (1). Many researchers – Zainal et al. [3], Lan et al. [12] and Syed Shabbar & Isam Janajreh [13] used Lagrange multiplier method to determine final gas composition of non-stoichiometric equilibrium model. The material balance equation was developed for each element, represented by Eq. (2). Products minimum thermodynamic potential equations and material balance equations are solved simultaneously along with a mole fraction equation for finding the mole composition of each product.

$$\Delta G^\circ + RT \ln x_j + \sum_k(\lambda a) = 0 \tag{1}$$

$$\sum_j(xa) = A_k / n_T \tag{2}$$

Where, ΔG° - Gibbs free energy of formation[kJ], R - Gas constant in [kJ $kmol^{-1}K^{-1}$], T -Temperature in [K], x - Mole fraction of gas components, λ - Lagrange multiplier, A_k - Number of moles of particular element [kmol] and n_T-Total number of moles [kmol].

Product composition of water gas reaction, water gas shift reaction and Boudouard reaction were determined for the temperature range 200–1200°C using FactSage software. Influence of gasification reactions, effect of temperature and equivalence ratio on gas composition was studied. H_2/CO ratio and CO/CO_2 ratio were determined based on the gas composition at different simulation conditions. Gas heating value and gasification efficiency were determined with reference to H_2/CO ratio.

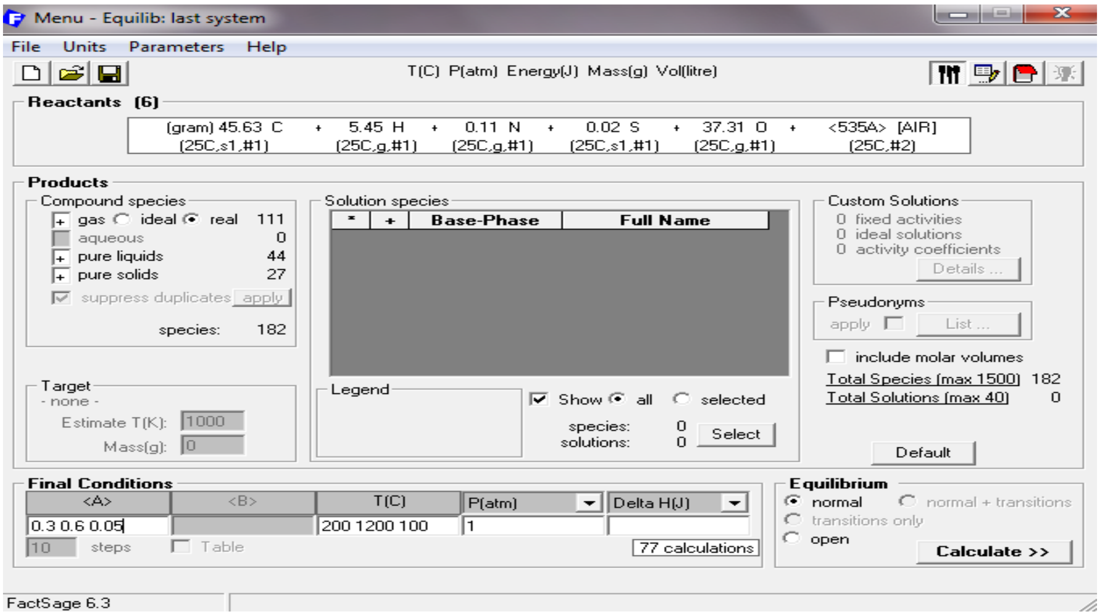
2.1. Gasification Simulation Procedure

Non-stoichiometric equilibrium model based on Gibbs free energy minimization using the Lagrange multiplier iterative method was adopted in FactSage software. The objective function of the model was subjected to mass balance and non-negativity of the number of moles to find the equilibrium composition of final products. Equilib module was selected from the calculate section of the FactSage software main window. Rice husk elemental composition, air quantity, temperature and pressure of the gasification process were the input detail. The elemental composition of rice husk is taken from Karmakar et al. [8], shown in Tab. 1.

Table 1. Proximate and ultimate analysis of rice husk

S.No	Fuel	Proximate Analysis [Wt.%]				Ultimate Analysis [Wt.%]				
		Volatile Matter	Fixed Carbon	Moisture	Ash	C	H	N	O	S
1	Rice husk	55.54	14.99	9.95	19.52	49.07	3.79	0.63	46.42	0.09

Air quantity required for gasification of rice husk was established by stoichiometric calculations. Rice husk elemental composition entered along with the air quantity in the reactant window of the software. The atmospheric condition was taken as the initial condition and the most stable phase was selected for the reactants. Simulation conditions were specified at the menu window, the temperature range was 600–800°C and the equivalence ratios were 0.25, 0.35 and 0.45. The menu window of FactSage software is shown in Fig. 1. From the gas composition result, H_2/CO ratio, CO/CO_2 ratio, gas heating value and gasification efficiency were determined.


Menu - Equilib: last system

T(C) P(atm) Energy(J) Mass(g) Vol(litre)

Reactants (6)

(gram) 45.63 C (25C,s1,#1) + 5.45 H (25C,g,#1) + 0.11 N (25C,g,#1) + 0.02 S (25C,s1,#1) + 37.31 O (25C,g,#1) + <535A> [AIR] (25C,#2)

Products

Compound species

gas ○ ideal ● real 111

aqueous 0

pure liquids 44

pure solids 27

☑ suppress duplicates

species: 182

Target - none -

Estimate T(K): 1000

Mass(g): 0

Custom Solutions

0 fixed activities

0 ideal solutions

0 activity coefficients

Pseudonyms apply ☐

☐ include molar volumes

Total Species (max 1500) 182

Total Solutions (max 40) 0

☑ Show ● all ○ selected

species: 0

solutions: 0

Final Conditions

<A>	<B>	T(C)	P(atm)	Delta H(J)
0.3 0.6 0.05		200 1200 100	1	

10 steps ☐ Table

77 calculations

Equilibrium

● normal ○ normal + transitions

○ transitions only

○ open

FactSage 6.3

Figure 1. FactSage Software Menu Window

2.2. Method of Data Processing

Gasification efficiency indicates the amount of energy transferred to the gas mixture from the rice husk. Gasification efficiency is calculated from the gas yield, lower heating value of gas mixture and rice husk. The gas yield represents the volume of gas produced per unit mass of biomass. Gasification efficiency calculated from Eq. (3)

$$\eta = (LHV_{gas} / LHV_{rice\ husk}) \times Y \times 100\ [\%] \tag{3}$$

Where, LHV_{gas}- Lower heating value of gas mixture [kJ Nm^{-3}], $LHV_{rice\ husk}$ - Lower heating value of rice husk[$kJkg^{-1}$], and Y- Gas yield [Nm^3kg^{-1}].

Gas lower heating value is determined from Eq. (4) given by Cheng et al. [14].Rice husk heating value found from Eq. (5) given by Proll and Hofbauer [15], gas yield value calculated from Eq. (6) given by Ngo et al. [16]

$$LHV_{gas}= (CO\% \times 126.36 + H_2\% \times 107.98 + CH_4\% \times 358.18) \tag{4}$$

Where, CO, H_2& CH_4 represents carbon monoxide, hydrogen and methane respectively in mole percentage.

$$LHV_{rice\ husk} = (34835\ C + 93870\ H - 10800\ O + 6280\ N + 10465\ S) \tag{5}$$

Where, C, H, O, N & S represents fuel bound carbon, hydrogen, oxygen, nitrogen and sulphur contents respectively in mass fraction.

$$\text{Gas yield} = Volume_{gas} / Mass_{rice\ husk} \tag{6}$$

Where, $Volume_{gas}$ - Volume of gas mixture [m^3], and $Mass_{rice\ husk}$ - Mass of rice husk [kg]

2.3. Model Validation

In rice husk air gasification simulation, the simulation parameters were T=600 – 800°C and ϕ=0.25-0.45. Simulation results were compared with the experimental value of Karmakar et al [8].The simulation result has an average RMS error of 3.59.The difference in gas composition of simulation result with literature value is shown in Fig. 2. For extensive details, refer to the author's previous publications [17, 18].

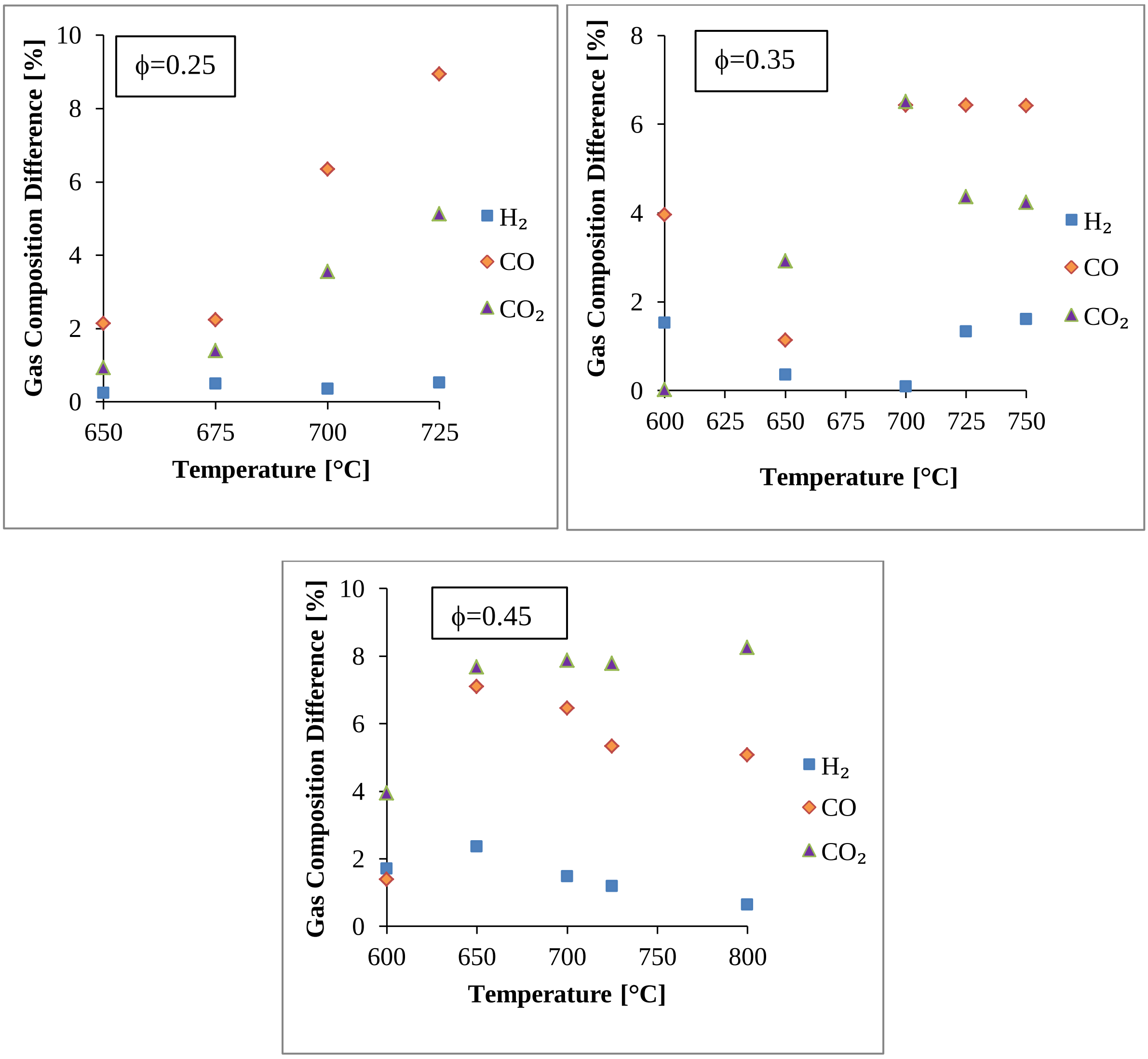


Figure 2. Gas composition simulation result validation

3. Result and Discussion

Gasification is a complex process,simultaneous chemical reactions occur during gasification. The final composition of the gas mixture depends on the individual chemical reactions that occur during gasification [19, 20].Water-gas reaction, water gas shift reaction and Boudouard reactions are predominant reactions for combustible gas production [21-23].With the exception of the water gas shift reaction, the other two reactions are endothermic.

3.1. Influence of Gasification Reactions on Gas Composition

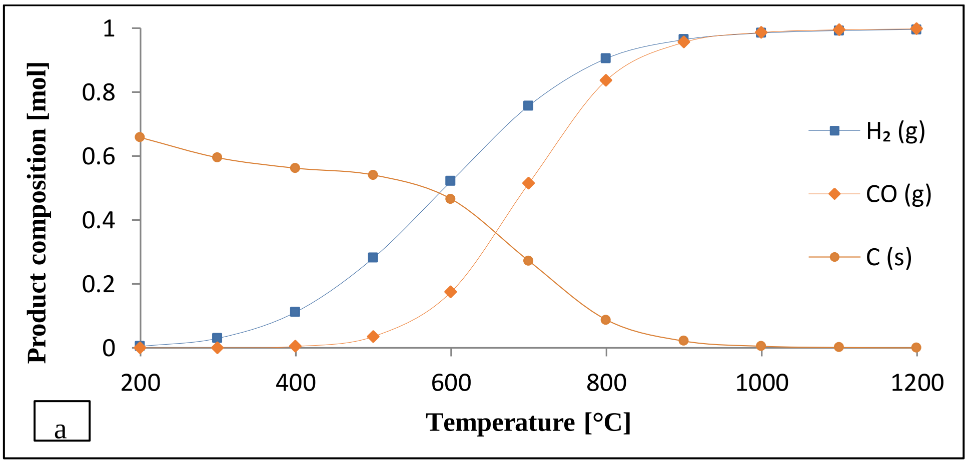
An increase in temperature favours the formation of H_2 and CO contents due to the improvement of the endothermic reactions [24, 25].Char formed during gasification reacts with steam released during the drying stage to produce H_2 and CO. Formed CO further reacts to form CO_2 and H_2.

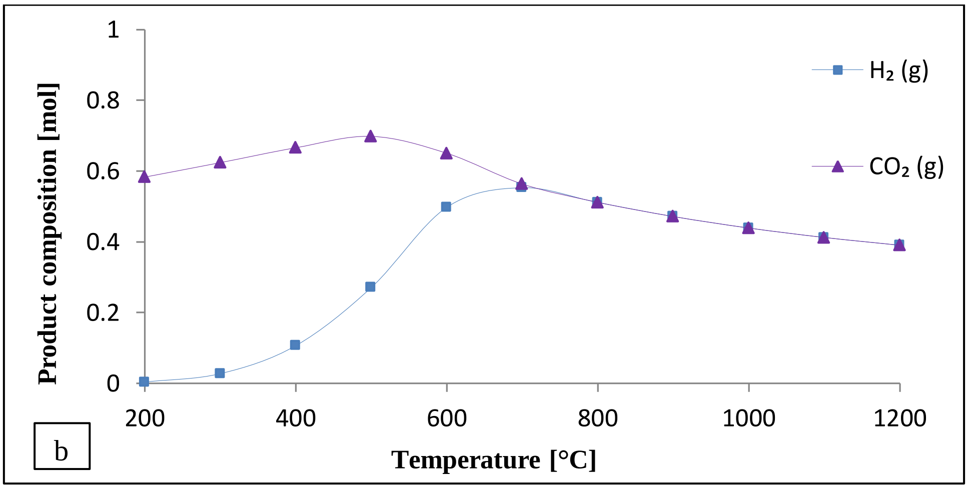
The produced CO_2 reduced into CO while reacting with the bed char. Hydrogen formation depends on water gas and water gas shift reaction. Carbon monoxide improvement occurs due to water gas and Boudouard reaction [24-26]. The reactions proceed until the char conversion occurs.Water gas reaction, water gas shift reaction and Boudouard reaction are indicated by Eq. (7), Eq. (8)&Eq. (9) respectively. The variation of gasification reactions product composition with temperature is shown in Fig.3.

$$C + H_2O \rightarrow CO + H_2 \tag{7}$$

$$CO + H_2O \rightarrow CO_2 + H_2 \tag{8}$$

$$C + CO_2 \rightarrow 2CO \tag{9}$$





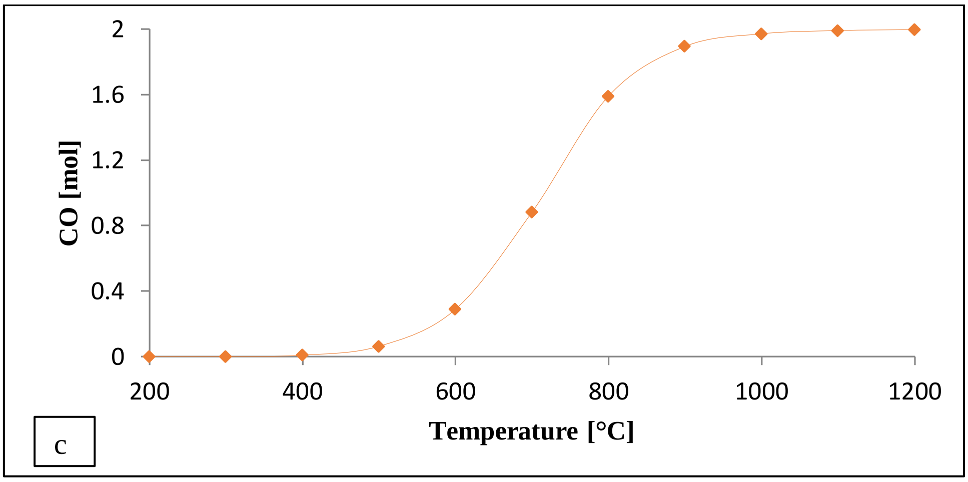


Figure 3. Product Composition Variation with Temperature (a) Water Gas Reaction, (b) Water Gas Shift Reaction and (c) Boudouard Reaction

A reduction in H_2 formation was observed in water gas shift reaction, the decrease of H_2 formation after 700°C was due to the endothermic behaviour change. The change in equilibrium of water gas shift reaction was counteracted by reversing ofreaction,H_2 reduction is justified by Le Chatelier's principle [25]. Enthalpy change of water gas shift reaction is shown in Fig. 4. From 600°C onwards the reactions becomes endothermic and effective reverse reaction starts from 700°C.

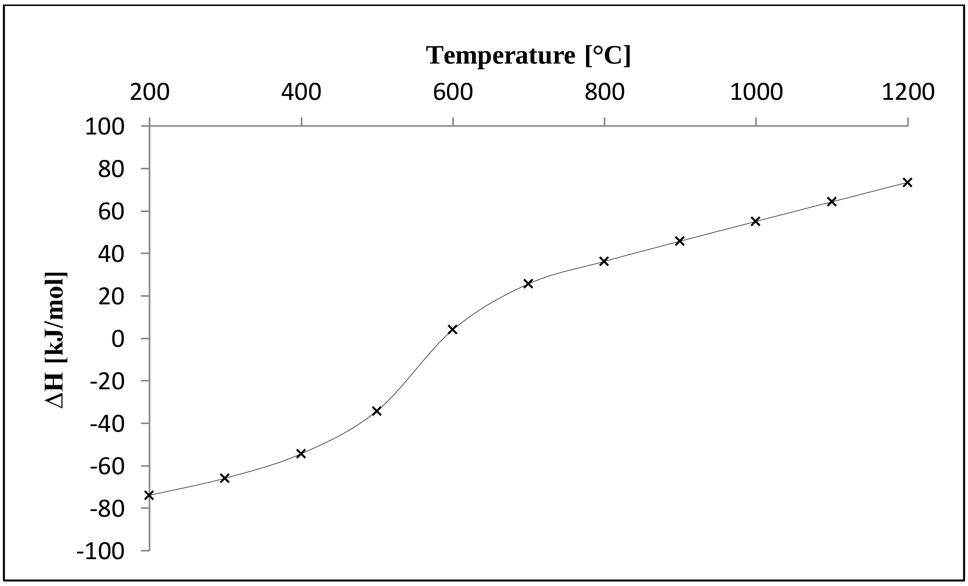


Figure 4. Effect of Temperature on Enthalpy Change of Water Gas Shift Reaction

3.2. Effect of Temperature on Gas Composition

The temperature of the rice husk air gasification varied from 650 – 725°C for the equivalence ratio of 0.25. H_2 and CO contents were improved with temperature raise; CO_2 and CH_4 contents were reduced.Hydrogen and CO reached a maximum value of 17.97% and 35.53% respectively at 725°C. An increase in temperature reduced the methane content by thermal cracking and reforming reactions [22, 23].An increase in temperature reduced the CO_2 content due to the Boudouard reaction and the reverse water gas shift reaction [25, 26]. For ϕ=0.35 and ϕ=0.45,a slight decrease of the H_2 content was observed after 725°C and 650°C respectively. Hydrogen reached a maximum value of 14.55%, 11.69% and CO reached 29.66%, 22.85% at $\phi=0.35$,$\phi=0.45$ respectively, shown in Fig. 5.

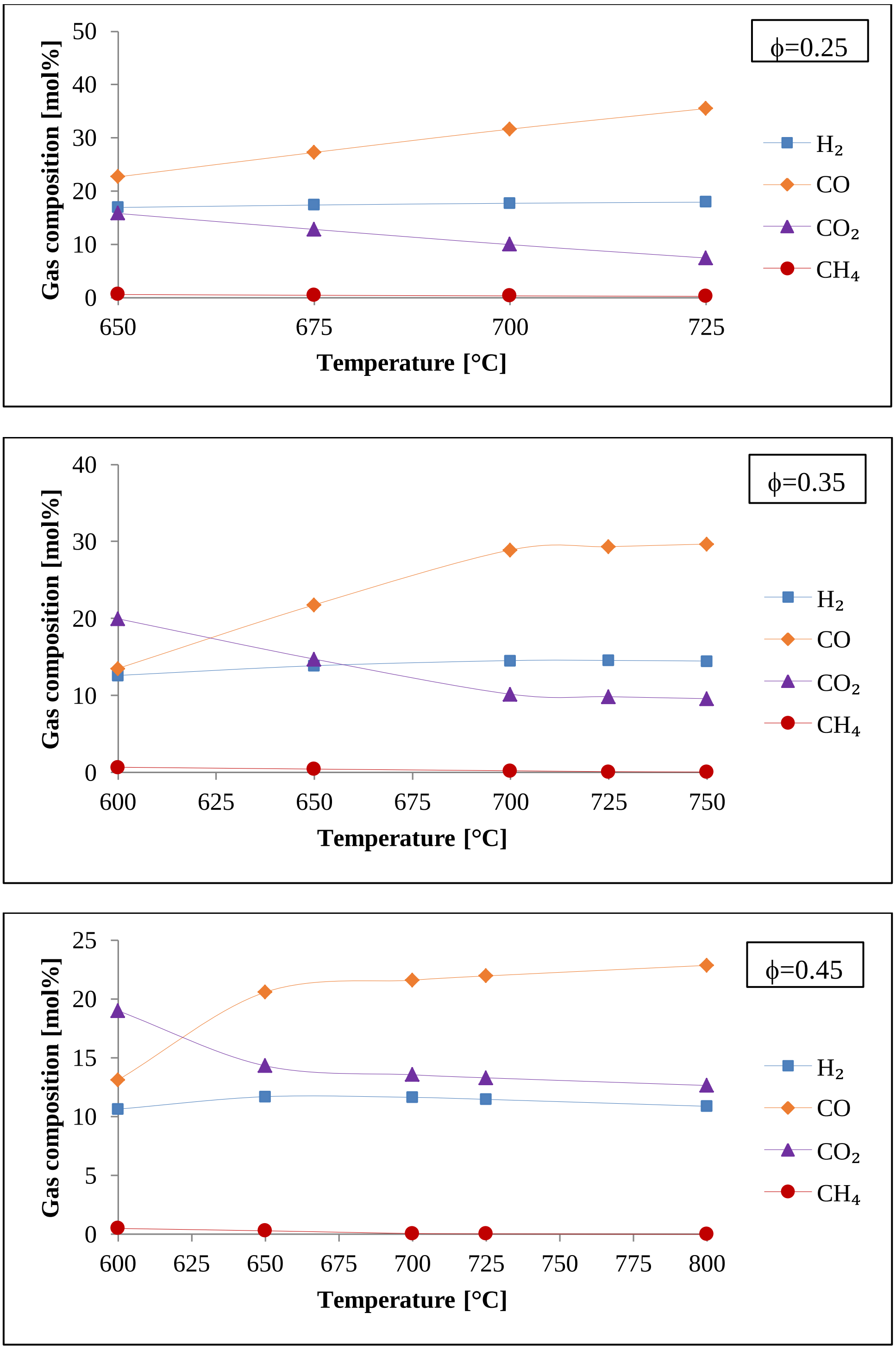


Figure 5. Gas composition at Different Conditions

3.3. Effect of Equivalence Ratio on Gas Composition

The lower limit of the equivalence ratio was decided by the quantity of gasifying medium required to burn the rice husk and to produce sufficient heat for endothermic reactions [8, 25].The effect of the equivalence ratio on gas composition was analyzed for 650°C, 700°C and 725°C is shown in Fig. 6.A high equivalence ratio during gasification leads to the combustion of rice husk. An increase in the equivalence ratio caused a decrease in the combustible gases due to the oxidation reaction [27-31]. Augmenting ϕ value from 0.25 to 0.45 at 650°C decreased the H_2 and CO content by 5.28% and 2.14% respectively. At 700°C, H_2 and CO content were decreased by 6.13% and 10.29% respectively; at 725°C, the reduction was 6.51% and 13.58% respectively. The prevalent reduction of H_2 and CO content was noticed at 725°C.

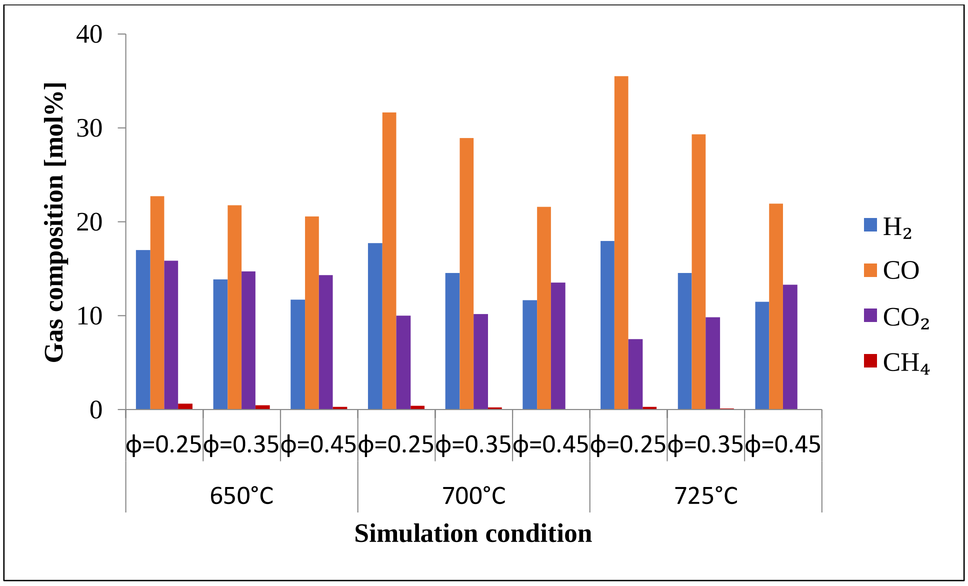


Figure 6. Effect of Equivalence Ratio on Gas Composition

3.4. H_2/CO Ratio and CO/CO_2 Ratio Analysis

At ϕ=0.25,increase in temperature decreases the H_2/CO ratio due to the higher formation of CO than H_2.The decreasing trend is applicable to other equivalence ratio conditions. The maximum H_2/CO ratio of 0.93 was attained at 600°C for ϕ=0.35. At ϕ=0.25, the temperature change from 650°C to 725°C leads to 32.32% of H_2/CO ratio reduction; 22.05% and 8.13% reduction was observed for ϕ=0.35 and 0.45 respectively. At 650°C, the increase in ϕ value decreases H_2/CO ratio due to the higher reduction of H_2 than CO.At 700°C and 725°C, H_2/CO ratio decreased for ϕ value change from 0.25 to 0.35; further increase in ϕ value improves H_2/CO ratio due to the higher CO reduction. H_2/CO ratio and CO/CO_2 ratio of rice husk air gasification is shown in Fig. 7.

Increase in temperature enhances CO/CO_2 ratio for all the equivalence ratios due to the increment of CO content and the corresponding reduction of CO_2 content [4, 5, 30-32]. At ϕ=0.25, maximum CO/CO_2 ratio of 4.74 was attained for 725°C.The temperature change from 650°C to 725°C leads to 2.31 times of CO/CO_2 ratio improvement.At 650°C increase in ϕ value initially augments the CO/CO_2 ratio and then decreases due to CO and CO_2 reduction with equivalence ratio. At 700°C and 725°C, the CO/CO_2 ratio decreases with the equivalence ratio due to the reduction of CO and the corresponding increase of the CO_2 content.

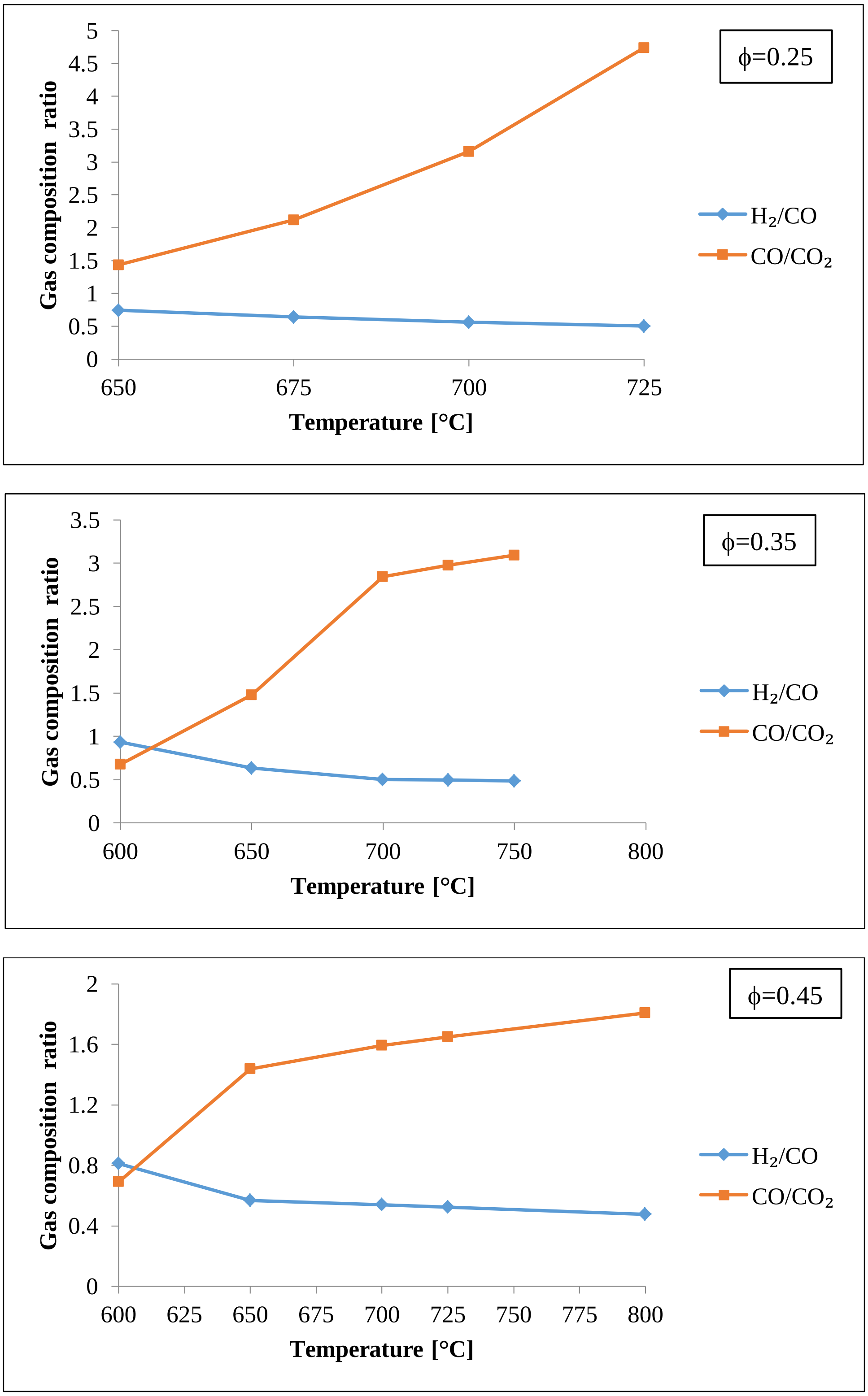


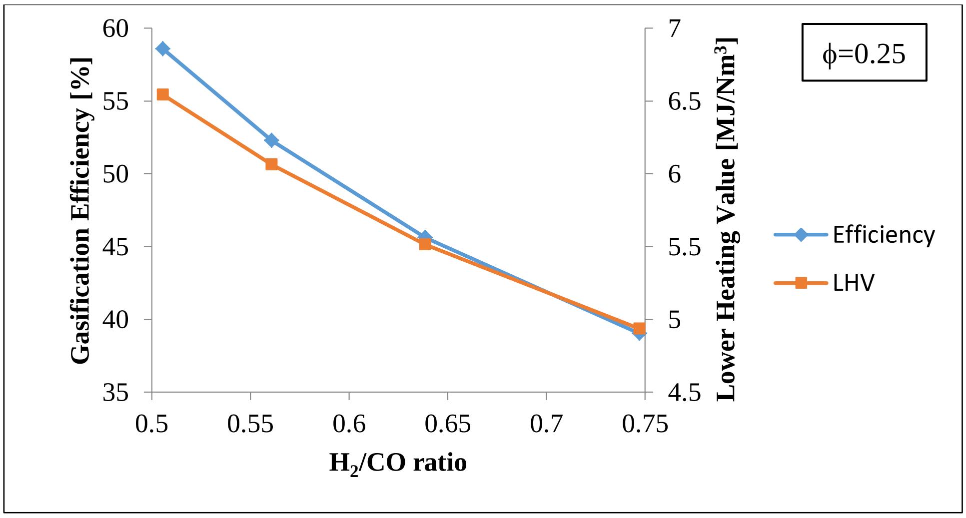
Figure 7. H_2/CO Ratio and CO/CO_2 Ratio at Different Conditions

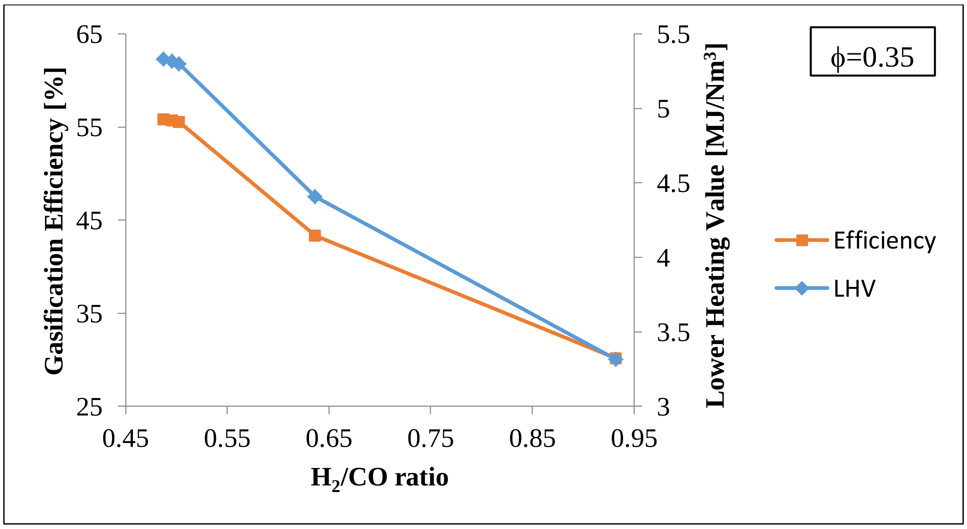
3.5. Gas Heating Value and Gasification Efficiency Analysis

For all the equivalence ratios, increase in temperature enhances gas lower heating value and gasification efficiency with corresponding H_2/CO reduction. At ϕ=0.25, the increment of heating value was in the range of 0.48-0.58 MJ/Nm^3; maximum heating value augment was observed for a temperature change from 650°C to 675°C. At ϕ = 0.35, the heating value improved by 2.01 MJ/Nm^3 for a temperature change from 600°C to 750°C.At ϕ = 0.45, the heating value was improved by1.08 MJ/Nm^3 for the total temperature change. Gas lower heating value and gasification efficiency value at different H_2/CO ratio is shown in Fig. 8.

At ϕ=0.25, maximum efficiency augment was observed for the temperature change from 675°C to 700°C;increment of efficiency was in the range of 6.3%-6.68%. At ϕ = 0.35 and ϕ = 0.45, efficiency was improved by 25.68% and 14.46% respectively for the total temperature change. At ϕ = 0.35 and ϕ = 0.45, maximum increase in heating value and efficiency occurs at initial temperature change;gas heating value and gasification efficiency value stabilizes after 700°C.

From the observation of gasification results, the same H_2/CO ratio values were obtained at the different operating conditions. H_2/CO ratio value of 0.5 was obtained at 725°C & ϕ = 0.25, 700°C & ϕ = 0.35 with corresponding CO/CO_2 ratio of 4.74 and 2.84 respectively. Similarly, the H_2/CO ratio value of 0.56 and 0.64 was reached under different operating conditions.The operating condition with higher CO/CO_2 ratio has higher gas heating value and gasification efficiency. The same value of the CO/CO_2 ratio occurred at 650°C for ϕ = 0.25 and 0.45, higher H_2/CO ratio was attained at ϕ = 0.25 with greater gas heating value and inferior gasification efficiency.The inferior gasification efficiency was due to the low gas yield at ϕ = 0.25.





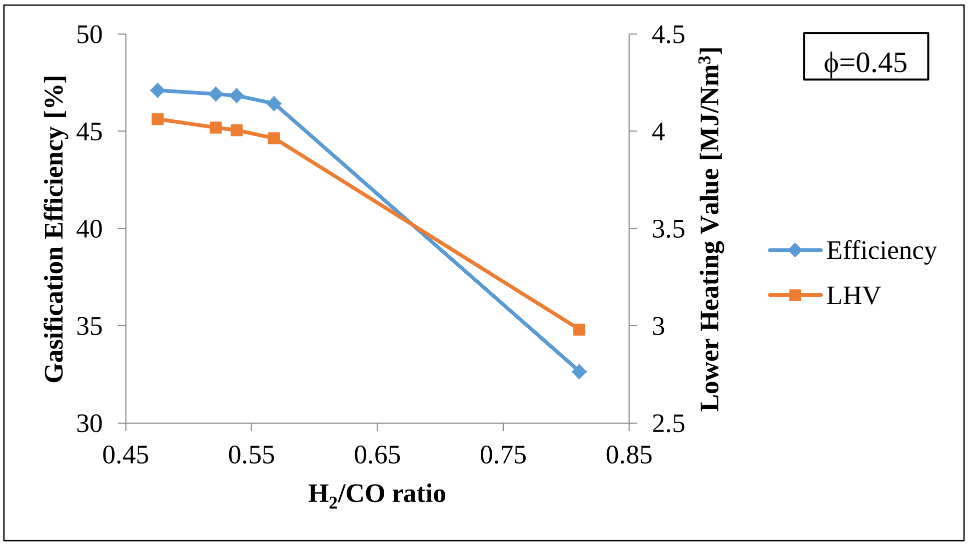


Figure 8. Gas Heating Value and Gasification Efficiency at Different H_2/CO Ratio

4. Conclusions

Rice husk air gasification simulation was carried out by a non-stoichiometric equilibrium model based on Gibbs free energy minimization using FactSage 6.3 software. The Influence of gasification reactions and effect of operating conditions on final gas composition was studied.Increase in temperature favours the formation of H_2 and CO contents due to the improvement of endothermic reactions.Hydrogen formation depends on water gas and water gas shift reaction, CO improvement occurs due to water gas and Boudouard reaction. A reduction in H_2 formation after 700 °C was observed in water gas shift reaction due to the endothermic behaviour change.H_2 and CO reached a maximum value of 17.97 % and 35.53 % respectively at 725 °C for $\phi = 0.25$. Increase in the equivalence ratio reduces the combustible gas content of the producer gas. Increase in temperature decreased the H_2/CO ratio and improved CO/CO_2 ratio for all equivalence ratios. The maximum H_2/CO ratio of 0.93 was attained at 600 °C & $\phi = 0.35$ and CO/CO_2 ratio of 4.74 was attained at 725 °C & $\phi = 0.25$. For operating conditions with same H_2/CO ratio value, better gas heating value and gasification efficiency were attained at higher CO/CO_2ratios.Maximum gasification efficiency of 58.57 % was attained at 725 °C for $\phi = 0.25$ with LHV = 6.54 MJ/Nm^3. For all the equivalence ratios, increase in temperature enhances the gas lower heating value and gasification efficiency with corresponding H_2/CO reduction.

Nomenclature

A_k - Number of moles of particular element, [kmol]
$\Delta G°$ - Gibbs free energy of formation, [kJ]
ΔH - Enthalpy change, [kJ mol^{-1}]

LHV_{gas}- Lower heating value of gas mixture, [kJ Nm^{-3}]
$LHV_{\text{rice husk}}$ - Lower heating value of rice husk, [kJ kg^{-1}]
n_T -Total number of moles, [kmol]

R- Gas constant, [kJ kmol^{-1} K^{-1}]
T- Temperature, [K]
x - Mole fraction of gas components

Y - Gas yield, [Nm^{3}kg-^{1}]

Greek symbols

λ- Lagrange multiplier

ϕ- Equivalence ratio

Acronyms

FT - Fischer-Troph

RMS - Root mean square

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