# INVESTIGATION OF GASIFICATION REACTIONS AND H<sub>2</sub>/CO RATIO ANALYSIS FOR RICE HUSK AIR GASIFICATION SIMULATION

#### by

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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,  $\phi$ , an increase in temperature reduces the  $H_2/CO$  and enhances the  $CO/CO_2$  ratio due to higher CO formation. For a particular,  $\phi$ , value, maximum gas heating value and gasification efficiency were obtained at low  $H_2/CO$  ratio.

Key words: rice husk, air gasification, gasification simulation,  $H_2/CO$  ratio, gas heating value, gasification efficiency

## 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 millionns of rice husk is produced [1]. The main constituents of producer gas are CO, CO<sub>2</sub> and H<sub>2</sub>. 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<sub>2</sub>/CO ratio of the syngas decides its final use. Syngas with a high H<sub>2</sub>/CO ratio is used for hydrogen production, syngas with H<sub>2</sub>/CO ratio value of two is used for raw materi-

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al production such as synthetic fuels, oxo-alcohols, ethylene glycol, acetic acid and dimethyl ether. The  $H_2/CO$  ratio is not a constraint for syngas use in boilers and turbines and  $CO/CO_2$  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  $H_2$  and CO mixture into liquid HC, the requirement of syngas for the FT process depends on high formation of H<sub>2</sub> and CO. The H<sub>2</sub>/CO ratio and CO/ CO<sub>2</sub> ratio were used to evaluate the effectiveness of the process. Carbonaceous materials are gasified with steam and oxygen to improve the H<sub>2</sub>/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  $Co/\gamma$ -Al<sub>2</sub>O<sub>3</sub> and Co-Re/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub>. A gas mixture with a low H<sub>2</sub>/CO ratio decreased the CO conversion and slightly increased the HC formation. The conversion of CH<sub>4</sub> into syngas is known as steam reforming reaction, syngas with high H<sub>2</sub>/CO ratio is produced during steam reforming and less CO<sub>2</sub> formation takes place compared to the gasification process. The complete conversion of CH<sub>4</sub> is not possible in reforming reaction due to equilibrium constraints. Cao et al. [6] injected  $CH_4$ 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<sub>2</sub>/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 FT synthesis and decentralized power generation. Non-stoichiometric equilibrium model (SOLGASMIX) was used for optimization by FactSage software. The H<sub>2</sub> and CO content in producer gas and H<sub>2</sub>/ 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<sub>2</sub> 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. The H<sub>2</sub> and CO formation plays an important role in the gasification process. Syngas with H<sub>2</sub>/ CO ratio of two is required for methanol production and FT synthesis; synthesis of ethanol, dimethyl ether and oxo-alcohols requires H<sub>2</sub>/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. The H<sub>2</sub>/CO ratio, CO/CO<sub>2</sub> ratio were evaluated, an analysis was carried out to find a suitable operating condition for maximum H<sub>2</sub>/CO ratio attainment. Gas heating value and gasification efficiency values were calculated at different H<sub>2</sub>/CO ratios for the simulation of rice husk air gasification process.

#### 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, and O) for reactants and the number of C,  $H_2$  and O 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 Shabbarand 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:

$$\Delta G^{\circ} + \mathbf{R}T \ln x_j \sum_{k} (\lambda a) + = 0 \tag{1}$$

$$\sum_{i} (xa) = \frac{A_k}{n_T} \tag{2}$$

where  $\Delta G^{\circ}$  [kJ] is the Gibbs free energy of formation, R [kJkmol<sup>-1</sup>K<sup>-1</sup>] – the gas constant, T [K] – the temperature, x [kmol] – the lar element, and  $n_T$  [kmol] – the total number of moles.

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. The H<sub>2</sub>/CO ratio and CO/CO<sub>2</sub> ratio were determined based on the gas composition at different simulation conditions. Gas heating value and gasification efficiency were determined with reference to H<sub>2</sub>/CO ratio.

### 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.

S No.	Fuel	Proximate analysis [wt.%]				Ultimate analysis [wt.%]				
		Volatile matter	Fixed carbon	Moisture	Ash	С	Н	N	0	S
1	Rice husk	55.54	14.99	9.95	19.52	49.07	3.79	0.63	46.42	0.09

Table 1. Proximate and ultimate analysis of rice husk

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				
File Units Parameters Help				
D 😅 🖬	T(C) P(atm) Energy(J) Mass(g) Vol(litre)	TT 🚽 🕒 📧		
Reactants (6)				
(gram) 45.63 C (25C,s1,#1)	+ 5.45 H + 0.11 N + 0.02 S + 37.31 O + (25C,g,#1) (25C,g,#1) (25C,s1,#1) (25C,g,#1)	<535A> [AIR] (25C,#2)		
Products				
Compound species	Solution species	Custom Solutions		
	• • Base-Phase Full Name	0 fixed activities 0 ideal solutions 0 activity coefficients Details		
species: 182		apply List		
- none -	Legend Show • all C selected	Total Species (max 1500) 182 Total Solutions (max 40) 0		
Estimate T(K): 1000 Mass(g): 0	species: 0 Select	Default		
Final Conditions	E	quilibrium		
<a> <b>           0.3 0.6 0.05        </b></a>	T(C)         P(atm)         C           200 1200 100         1         C           77 calculations         C	normal C normal + transitions transitions only open Calculate >>		
FactSage 6.3				

Figure 1. FactSage software menu window

# 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:

$$\eta = \frac{\text{LHV}_{\text{gas}}}{\text{LHV}_{\text{rice husk}}} \times Y \times 100\%$$
(3)

where  $LHV_{gas}$  [kJ Nm<sup>-3</sup>] is the lower heating value of gas mixture,  $LHV_{rice husk}$  [kJkg<sup>-1</sup>] – the lower heating value of rice husk, and Y [Nm<sup>3</sup>kg<sup>-1</sup>] – the gas yield.

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]:

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

where CO, H<sub>2</sub>, and CH<sub>4</sub> represents carbon monoxide, hydrogen, and methane, respectively in mole percentage:

$$LHV_{ricehusk} = (34835C + 126.36H - 10800O + 6280N + 10465S)$$
(5)

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

$$Gas_{yield} = \frac{Volume_{gas}}{Mass_{rice husk}}$$
(6)

where Volume<sub>gas</sub> [m<sup>3</sup>] is the volume of gas mixture and Mass<sub>rice husk</sub> [kg] – the mass of rice husk.

#### Model validation

In rice husk air gasification simulation, the simulation parameters were  $T = 600-800^{\circ}$ C and  $\phi = 0.25-0.45$ . 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; (a)  $\phi = 0.25$ , (b)  $\phi = 0.35$ , and (c)  $\phi = 0.45$ 

## **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.

# 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<sub>2</sub> and  $H_2$ . The produced CO<sub>2</sub> reduced into CO while reacting with the bed char. The  $H_2$  formation depends on water gas and water gas shift reaction. The CO improvement occurs due to water gas and Boudouard reaction [24-26]. The reactions proceed until the char conversion occurs. Water gas reaction, water gas



Figure 3. Product composition variation with temperature; (a) water gas reaction, (b) water gas shift reaction, and (c) Boudouard reaction

shift reaction and Boudouard reaction are indicated by eqs. (7)-(9), respectively. The variation of gasification reactions product composition with temperature is shown in fig. 3.

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

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

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



Figure 4. Effect of temperature on enthalpy change of water gas shift reaction



Figure 5. Gas composition at different conditions; (a)  $\phi = 0.25$ , (b)  $\phi = 0.35$ , and (c)  $\phi = 0.45$ 

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 of reaction,  $H_2$  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.

#### 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. The  $H_2$  and CO contents were improved with temperature raise, CO<sub>2</sub> and CH<sub>4</sub> contents were reduced. The H<sub>2</sub> 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  $\phi = 0.35$  and  $\phi = 0.45$ , a slight decrease of the H<sub>2</sub> content was observed after 725 °C and 650 °C, respectively. The H<sub>2</sub> 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.

# 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  $\phi$  value from 0.25-0.45 at 650 °C decreased the H<sub>2</sub> and CO content by 5.28% and 2.14%, respectively. At 700 °C, H<sub>2</sub>, 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<sub>2</sub> and CO content was noticed at 725 °C.

### The H<sub>2</sub>/CO ratio and CO/CO<sub>2</sub> ratio analysis

At  $\phi = 0.25$ , increase in temperature decreases the H<sub>2</sub>/CO ratio due to the higher formation of CO than H<sub>2</sub>. 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  $\phi$  =0.35. At  $\phi$  = 0.25, the temperature change from 650-725 °C leads to 32.32% of H<sub>2</sub>/CO ratio reduction, 22.05% and 8.13% reduction was observed for  $\phi = 0.35$  and 0.45, respectively. At 650 °C, the increase in  $\phi$ value decreases H<sub>2</sub>/CO ratio due to the higher reduction of H<sub>2</sub> than CO. At 700 °C and 725 °C,  $H_2$ /CO ratio decreased for  $\phi$  value change from 0.25-0.35, further increase in  $\phi$  value improves  $H_2/CO$  ratio due to the higher CO reduction. The H<sub>2</sub>/CO ratio and CO/CO<sub>2</sub> ratio of rice husk air gasification is shown in fig. 7.

Increase in temperature enhances CO/ CO<sub>2</sub> ratio for all the equivalence ratios due to the increment of CO content and the corresponding reduction of CO<sub>2</sub> content [4, 5, 30-32]. At  $\phi = 0.25$ , maximum CO/CO<sub>2</sub> ratio of 4.74 was attained for 725 °C. The temperature change from 650-725 °C leads to 2.31 times of CO/CO<sub>2</sub> ratio improvement. At 650 °C increase in  $\phi$  value initially augments the CO/CO<sub>2</sub> ratio and then decreases due to CO and CO<sub>2</sub> reduction with equivalence ratio. At 700 °C and 725 °C, the CO/CO<sub>2</sub> ratio decreases with the equivalence ratio due to the reduction of CO and the corresponding increase of the CO<sub>2</sub> content.



Figure 6. Effect of equivalence ratio on gas composition



Figure 7. The H<sub>2</sub>/CO ratio and CO/CO<sub>2</sub> ratio at different conditions; (a)  $\phi = 0.25$ , (b)  $\phi = 0.35$ , and (c)  $\phi = 0.45$ 



Figure 8. Gas heating value and gasification efficiency at different H<sub>2</sub>/CO ratio; (a)  $\phi = 0.25$ , (b)  $\phi = 0.35$ , and (c)  $\phi = 0.45$ 

# 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<sub>2</sub>/CO reduction. At  $\phi = 0.25$ , the increment of heating value was in the range of 0.48-0.58 MJ/Nm<sup>3</sup>. Maximum heating value augment was observed for a temperature change from 650-675 °C. At  $\phi = 0.35$ , the heating value improved by 2.01 MJ/Nm<sup>3</sup> for a temperature change from 600-750 °C. At  $\phi = 0.45$ , the heating value was improved by 1.08 MJ/Nm<sup>3</sup> for the total temperature change. Gas lower heating value and gasification efficiency value at different H<sub>2</sub>/CO ratio is shown in fig. 8.

At  $\phi = 0.25$ , maximum efficiency augment was observed for the temperature change from 675-700 °C. Increment of efficiency was in the range of 6.3%-6.68%. At  $\phi = 0.35$  and  $\phi = 0.45$ , efficiency was improved by 25.68% and 14.46%, respectively, for the total temperature change. At  $\phi = 0.35$  and  $\phi = 0.45$ , 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<sub>2</sub>/CO ratio values were obtained at the different operating conditions. The H<sub>2</sub>/CO ratio value of 0.5 was obtained at 725°C and  $\phi = 0.25$ , 700 °C and  $\phi = 0.35$  with corresponding CO/CO<sub>2</sub> ratio of 4.74 and 2.84, respectively. Similarly, the H<sub>2</sub>/CO ratio value of 0.56 and 0.64 was reached under different operating conditions. The

operating condition with higher CO/CO<sub>2</sub> ratio has higher gas heating value and gasification efficiency. The same value of the CO/CO<sub>2</sub> ratio occurred at 650 °C for  $\phi = 0.25$  and 0.45, higher H<sub>2</sub>/CO ratio was attained at  $\phi = 0.25$  with greater gas heating value and inferior gasification efficiency. The inferior gasification efficiency was due to the low gas yield at  $\phi = 0.25$ .

#### Conclusion

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. The  $H_2$  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. The  $H_2$  and CO reached a maximum value of 17.97 % and 35.53 %, respectively, at 725 °C for Venugopal, D., *et al.*: Investigation of Gasification Reactions and H<sub>2</sub>/CO Ratio ... THERMAL SCIENCE: Year 2023, Vol. 27, No. 1B, pp. 881-890

 $\phi = 0.25$ . Increase in the equivalence ratio reduces the combustible gas content of the producer gas. Increase in temperature decreased the H<sub>2</sub>/CO ratio and improved CO/CO<sub>2</sub> ratio for all equivalence ratios. The maximum H<sub>2</sub>/CO ratio of 0.93 was attained at 600 °C and  $\phi = 0.35$  and CO/CO<sub>2</sub> ratio of 4.74 was attained at 725 °C and  $\phi = 0.25$ . For operating conditions with same H<sub>2</sub>/CO ratio value, better gas heating value and gasification efficiency were attained at higher CO/CO<sub>2</sub> ratios. Maximum gasification efficiency of 58.57% was attained at 725 °C for  $\phi = 0.25$  with LHV = 6.54 MJ/Nm<sup>3</sup>. For all the equivalence ratios, increase in temperature enhances the gas lower heating value and gasification efficiency with corresponding H<sub>2</sub>/CO reduction.

#### Nomenclature

- $A_k$  number of moles of particular
- element, [kmol]

 $C(s) \quad - \, carbon, [mol]$ 

- CO(g) carbon monoxide, [mol]
- $\Delta G^{\circ}$  gibbs free energy of formation, [kJ]
- H<sub>2</sub>(g) hydrogen, [mol]
- $\Delta H$  enthalpy change, [kJmol<sup>-1</sup>]
- $LHV_{gas}$  lower heating value of gas
- mixture, [kJNm<sup>-3</sup>]
- $LHV_{rice \ husk}$  lower heating value of
- rice husk, [kJkg<sup>-1</sup>]
- $n_T$  total number of moles, [kmol]
- R gas constant,  $[kJkmol^{-1}K^{-1}]$

- T temperature, [K]
- x mole fraction of gas components
- Y = gas yield, [Nm<sup>3</sup>kg<sup>-1</sup>]

#### Greek symbols

- $\lambda$  Lagrange multiplier
- $\phi$  equivalence ratio

#### Acronyms

- R gas constant, [kJkmol<sup>-1</sup>K<sup>-1</sup>]
- FT Fischer-Troph
- RMS root mean square

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