Prediction of Rice Husk Gasification on Fluidized Bed Gasifier Based on Aspen Plus

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A biomass gasification model was developed using Aspen Plus based on the Gibbs free energy minimization method. This model aims to predict and analyze the biomass gasification process using the blocks of the RGibbs reactor and the RYield reactor. The model was modified by the incomplete equilibrium of the RGibbs reactor to match the real processes that take place in a rice husk gasifier. The model was verified and validated, and the effects of gasification temperature, gasification pressure, and equivalence ratio (ER) on the gas component composition, gas yield, and gasification efficiency were studied on the basis of the Aspen Plus simulation. An increasing gasification temperature was shown to be conducive to the concentrations of H_2 and CO, and gas yield and gasification efficiency reached peaks of 2.09 m^3/kg and 83.56%, respectively, at 700 °C. Pressurized conditions were conducive to the formation of CH_4 and rapidly increased the calorific value of syngas as the gasification pressure increased from 0.1 to 5 MPa. In addition, the optimal ER for gasification is approximately 0.3, when the concentrations of H_2 and CO and the gasification efficiency reach peaks of 23.65%, 24.93% and 85.92%, respectively.

Keywords: Biomass; Gasification; Aspen plus; Prediction

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INTRODUCTION

Currently, the depletion of fossil energy and the environmental problems, brought about by the high usage of fossil fuels during industrial development, has motivated a search for an ideal clean and renewable energy technology. As the fourth largest energy source in the world, behind only coal, oil, and natural gas, biomass is able to concentrate solar energy in a cheap and efficient way, to reduce greenhouse gases and other harmful gas emissions, and to easily be converted to conventional fuels. Biomass has a huge potential consumption globally (Chen *et al*. 2004; Kuprianov *et al*. 2011).

Biomass gasification is a thermochemical process that can effectively convert biomass to chemical energy in the form of gas fuel. With the help of gasification agents such as steam, air, and oxygen, a mixture known as biogas containing CO, H_2, and low-molecular weight hydrocarbon is produced in a biomass conversion process (Yuan *et al.* 2005). Rice husks are the largest part of the by-products left during rice processing. With the growing of rice in more than 75 countries around the world, the output of rice is approximately 6 billion tons in the world, which produces almost 2 billion tons of rice husk, calculated as 20% of the rice weight (Lu *et al.* 2005). The annual output of rice husk in China reached about 32 million tons in 1996, which is the most in the world. In the present work rice husk was selected as the raw material for the reasons that this material has a giant reserve and is available at low price. Furthermore, because of advantages such as lower investment costs, simple operation, and easy implementation in an auto-thermal conversion system, our developed model chooses air as a gasifying agent. The air gasification experiments were carried out using the rice husk in a fluidized bed reactor (Jia *et al.* 2007; Srinath and Reddy 2011), and the factors affecting the gasification process were analyzed to obtain the optimized parameters in the process.

The complexity and the variability of the gasification process results in a complex structure of the gasification device in the experiments. Meanwhile, the process is limited by the field test conditions and the gasification devices, through which it is difficult to entirely grasp the gasification characteristics. However, the analysis and the prediction of the simulation method can effectively compensate for the inherent limitation in the experimental system. Thus, developing a gasification model is helpful for optimizing the gasification process (Niu *et al.* 2013).

As a general-purpose process software, Aspen Plus has been widely used in coal combustion, gasification, conversion, and utilization (Lee *et al*. 1992; Backham *et al*. 2004), but the gasification of various types of biomass has not been applied on a large scale. In recent years, the simulation of biomass gasification has been presented using Aspen Plus in various reactors and by obtaining a series of research findings. Mathieu and Dubuisson (2002) simulated sawdust gasification process with air using Aspen Plus in a fluidized bed reactor and analyzed the influence of air temperature, air oxygen content, and operating pressure on gasification. Gao *et al*. (2008) used Aspen Plus to establish an interconnected fluidized bed model based on non-catalytic rice straw gasification with limestone and discussed the effects of gasification temperature and stream-to-biomass ratio on the process. Similarly, Zhang *et al.* (2009) established an interconnected fluidized bed model using Aspen Plus to simulate the straw gasification; they analyzed the effects of gasification temperature, pressure, and stream-to-biomass ratio on the yield of methanol. Nikoo and Mahinpey (2008) constructed a biomass gasification model with Aspen Plus in a fluidized bed and discussed the effects of gasification temperature, equivalence ratio, partial average size, ER, and stream-to-biomass ratio on gasification results. However, because of the neglect of incomplete carbon conversion, there are some deviations between simulated and experimental results. The overall trend in the experimental and predicted results, however, was identical, demonstrating that the Aspen Plus software can be used for simulation of biomass gasification. Chen *et al*. (2007) developed an air gasification model in a fluidized bed with Aspen Plus and compared peanut shell gasification results to verify the accuracy and reliability of the model. Aspen Plus software can thus be applied to the simulation of agricultural waste gasification.

In previous simulations, the Gibbs free energy reactor module often has been used to simulate biomass gasification. Considering thermodynamic equilibrium and ignoring the kinetic factors resulted in a greater deviation in the actual gas-solid two-phase diffusion from assuming ideal conditions in simulations.

This paper is based on the Gibbs free energy minimization principle (Wang *et al*. 2004) including restricted equilibrium parts, which adjusts the model prediction to agree with the experimental value. By comparing the simulation and experimental results, we are able to verify the accuracy and reliability of gasification. The presented simulation model is credibly used to analyze the effects of gasification temperature, pressure, and equivalence ratio on the process.

EXPERIMENTAL

Gasification Modeling

Biomass particle gasification in a gasifier is a complex process, which includes heat transfer and mass transfer (Gao *et al*. 2008). After entering the high temperature fluidised bed gasifier, biomass firstly produces gas, char, and tar during pyrolysis. Then, in the dense-phase zone, char is subjected to a redox reaction, and tar undergos a second pyrolysis reaction. Finally, gas produced by gasification experiences a reforming reaction in the dilute-phase zone.

The overall process of conversion of the rice husk to gases and energy is divided into three stages in the model, which include pyrolysis, combustion, and gasification. Here, the term pyrolysis will be taken to mean the thermal decomposition of biomass prior to oxidation. The term combustion will refer to reactions involving consumption of oxygen gas, and the term gasification will refer to further redox reactions after preliminary gasification. Meanwhile, it is hypothesized that phase transitions during a gasification process are stable processes. With these assumptions, the equilibrium model is established based on the Gibbs free energy minimization principle. The model simulating the processes in Aspen Plus is based on the following assumptions (Cardoso 1989; Hyvärinen and Oja 2000; Fermoso *et al*. 2010; Gordillo and Annamalai 2010):

1. The gasifier remains stable in operation, and all the parameters are unrelated to time;
2. Elements including O, H, N, and S are completely transferred into a gas phase, and C undergos an incomplete transformation with changes of conditions;
3. Ash in biomass is considered an insert substance and does not participate in the gasification process;
4. The gasification agent mixes with biomass particles instantly in the furnace;
5. All gas phase reactions react fast and reach equilibrium;
6. Biomass particles have temperature uniformity and the temperature gradient is zero; and
7. Pressure in the gasification furnace is constant and the pressure gradient is zero.

Based on the simplifying assumptions given above, an air-steam gasification fluidized-bed model is built (Fig. 1).

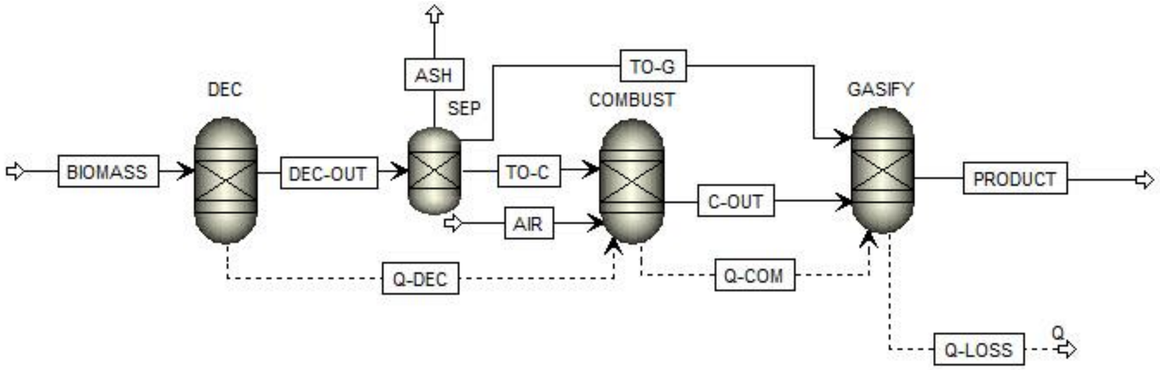

Fig. 1. Flow chart of biomass gasification system

Combined with the principle of biomass gasification, the biomass gasification process is simulated based on the pyrolysis module (Ryield), combustion module (RGibbs) (Plus 2003), and the gasification modules (RGibbs). In the model, the stream of biomass is defined as "unconventional composition." By adding elemental analysis and industry analysis, it is transformed into a routine component to input.

As shown in the model, raw biomass materials (BIOMASS) first enter the pyrolysis module (DEC, abbreviated from DECOMPOSITION); then pyrolysis products (DEC-OUT), including C, H, O, N, S, and ash, are formed. These products are separated by a follow-up separation module (SEP, abbreviated from SEPARATION) into a gas section (TO-G, abbreviated from GAS produced by SEP), fixed carbon (TO-C, abbreviated from FIXED CARBON produced by SEP), and ash (ASH). Ash (ASH) is discharged from the separation module (SEP). Fixed carbon (TO-C) and heat of pyrolysis (Q-DEC, abbreviated from HEAT RELEASE of DECOMPOSITION) enter the first RGIBBS reactor module (COMBUST, abbreviated from COMBUSTION) with the participation of a gasification agent, *i.e.*, air (AIR), to undergo incomplete combustion. Heat generated during pyrolysis (Q-DEC) enters the first RGIBBS reactor module (COMBUST). Gas section (TO-G) and products (C-OUT, abbreviated from C produced by COMBUST) from the incomplete combustion in the first RGIBBS reactor module (COMBUST), as well as parts of combustion heat released (Q-COM, abbreviated from HEAT RELEASE of COMBUSTION), enter the second RGIBBS reactor module (GASIFY, abbreviated from GASICIFATION) to undergo gasification. Some heat loss (Q-LOSS, abbreviated from HEAT LOSS) in this process flows to the second RGIBBS reactor module (GASIFY).

The main reactions in the model (Franco *et al*. 2003) are as follows:

1. Oxidizing reaction:

$$C+O_2=CO_2 \quad (1)$$

$$2C+O_2=2CO \quad (2)$$

2. Gas reforming reaction:

$$C+CO_2=2CO \quad (3)$$

$$C + H_2O =CO+H_2 \quad (4)$$

$$C +2H_2O =CO_2+2H_2 \quad (5)$$

$$CO + H_2O =CO_2+H_2 \quad (6)$$

$$CH_4 + H_2O = CO + 3H_2 \quad (7)$$

3. Methanation reaction:

$$C + 2H_2=CH_4 \quad (8)$$

According to chemical reaction kinetics (Xu 2004), the rate of gasification is mostly affected by heterogeneous reactions between carbon particles and the gasification agent. The overall velocity of reaction of the gas-solid two-phase is related to the gas diffusion velocity from gas to surface of solid carbon particles. Additionally, it is also under the influence of the chemical reaction speed. According to the Arrhenius equation, when the temperature rises, the velocity of heterogeneous-phase chemical reactions accelerates. However, the diffusion speed of the gas-solid two-phase is relatively slow. Consequently, because of the effects of the low diffusion rate in a gas-solid two-phase system, the model was unable to reach the chemical equilibrium assumed in the Gibbs free energy minimization method, resulting in a deviation between experimental values and simulation values. Based on the partial equilibrium steps included in the Gibbs free energy minimization principle, the process is able to approximate the conditions of the RGIBBS reactor module to close the gaps between practical and ideal reactions for the gas-solid reaction. By setting respective balanced approaching temperatures in gas-solid reactions (1) to (5), the model is modified to make gasification a better approximation of the actual situation.

RESULTS AND DISCUSSION

Model Verification

To verify the reliability of the model, the simulation results of rice husk gasification in the air were compared to the experimental data (Yao 2008). The raw materials in the test were the rice husk, and its proximate analysis and ultimate analysis are shown in Table 1 (Yao 2008). This model is used to simulate the bio-gas from air gasification when the equivalence ratio ranges from 0.22 to 0.25, at 600 °C gasification temperature, with 0.1 MPa gasification pressure.

Table 1. Proximate and Ultimate Analysis of Rice Husk

Proximate analysis (wt% on air dry basis)				Ultimate analysis (wt% on air dry basis)					LHV (MJ/kg)
Moisture	Ash	Volatiles	Fixed carbon	Carbon	Hydrogen	Oxygen	Nitrogen	Sulfur	
12.8	15.9	57.3	13.9	37.7	4.3	28.6	0.4	0.05	14.28

As can be seen in Fig. 2, the tendency of changes in the gas component composition, gas yield, gas calorific value, and gasification efficiency in the gasification simulation results with equivalence ratio ER (Yao 2008) were in accordance with test results. Among these results, the simulated results of the content of CO and CO_2 were consistent with the experimental results in all the experimental ranges.

The content of H_2 had a higher value compared to the experimental value, while the content of CH_4 was lower than the real results. In addition, the simulation results of gas yield in the whole test range were in agreement with the experimental value; however, the predictive value of gas calorific and the gasification efficiency of bio-gas was relatively low.

Reasons for the higher forecast value of H_2 contents include the following: ①The model ignored hydrocarbon content, including C_nH_m, when the model is established on the basis of the Gibbs free energy minimization principle. According to the equilibrium of H elements and chemical equilibrium, the content of H_2 is higher than the experimental value. ②In addition, water contained in biomass leads to the production of H_2 and O_2 in the pyrolysis module (DEC), which can act as H_2O in the RGibbs reactor and result in an increase in H_2 production.

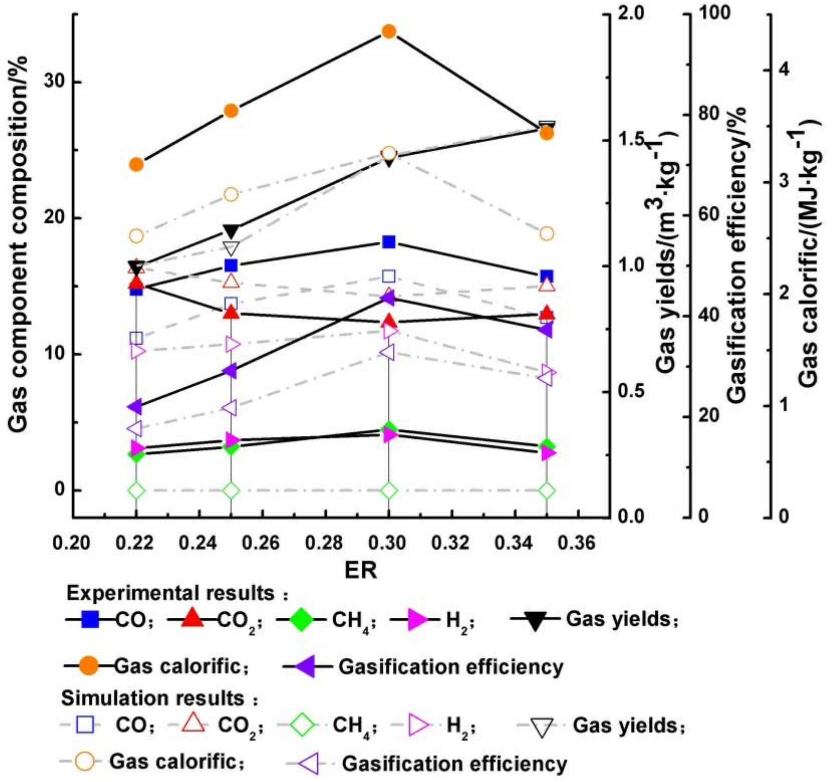

Fig. 2. Comparison of model with experimental data

Based on the conservation of atoms and energy when the RGibbs reactor module is established, with increasing production of H_2, the rest of the H elements for CH_4 generation decrease correspondingly. Meanwhile, limiting conditions of the gas-solid reaction in this gasification model was an effective modification, but restrictions related to the equipment and the effects of factors in the actual test made the actual reaction deviate greatly from the balanced state in reaction (7), which caused high rates in CH_4 decomposition and led to a lower simulation value of CH_4.

This situation is similar to the simulation results in the literature (Schuster *et al*. 2001; Mathieu *et al*. 2002; Zhang 2009), in which Schuster *et al.* (2001) regarded the reasons for the results above as: reactions in actual tests are unable to reach equilibrium; thus, methane and hydrocarbons released in biomass pyrolysis increase, eventually leading to incomplete equilibrium concentrations of CO, CO_2, and H_2.

It can be clearly seen that syngas was in low CH_4 content which has high calorific value. In addition, ignoring C_2H_2, C_2H_4, and C_2H_6, which have high calorific values, causes a low calorific value of synthesis gas. According to the calculation formula of gasification efficiency:

$$\eta\,(\%) = \frac{WQ_{LHV}^{g}}{Q_{LHV}} \times 100\% \tag{9}$$

Gasification efficiency is related to W (dry gas yield), Q^{g}_{LHV} (the calorific value of gas gasification products) and Q_{LHV} (lower heating value of biomass material). From the equation, W is consistent with the experimental values and low Q^{g}_{LHV} decreases gasification efficiency, but is still consistent with the tendency of the test results.

The Effects of Gasification Temperature

Figure 3 shows how the gasification temperatures ranging from 400 to 1500 °C affect the results of the process when the feed quantity of rice husk is 1 kg/h, entering when the air amount is 1.38 kg/h with a temperature of 25 °C and pressure of 0.1 MPa. The simulation shows that, with the gasification temperature increasing from 400 to 700 °C, the contents of H_2 and CO increase while the contents of CO_2 and CH_4 decrease. Thus, gasification efficiency and gas yield gradually increase, which is consistent with a previous report (Basu 2006). A possible reason to explain the result is that the reforming reaction of CH_4, Boudouard reaction, water gas reaction, and the reverse reaction of methanation were accelerated when the gasification temperature increases. Therefore, CH_4 and CO_2 gradually convert to CO and H_2 (Cardoso 1989), which leads to faster increase of H_2 contents than CO contents.

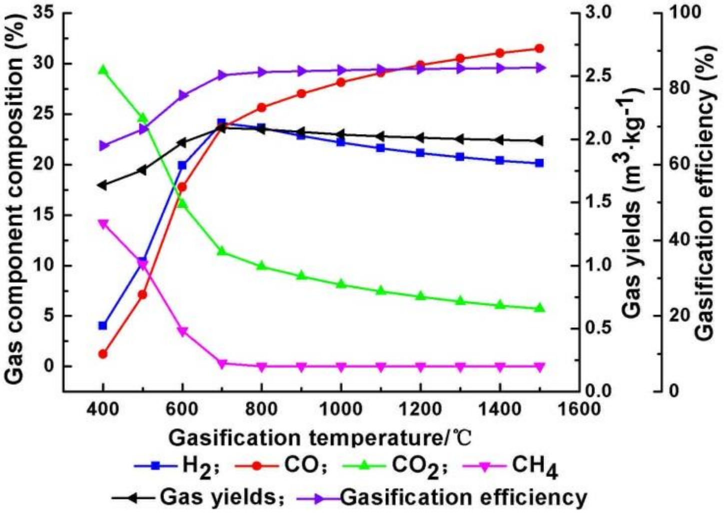

Fig. 3. Effect of gasification temperature on gas composition, gas yield, and gasification efficiency

Though the water gas reaction among all reactions is able to generate CO_2, the chemical equilibrium constant to generate CO_2 is smaller than the chemical equilibrium constant to generate CO of 7.50 at 700°C. The total rate of all reactions to generate CO is smaller than that to generate CO_2, as a result of which increasing temperature is more conducive to generating CO in gasification. The formation of combustible gas production leads to an increase in the gas calorific value and the gas yield in this period; meanwhile, gasification efficiency increases constantly. When the gasification temperature is above 700 °C, the contents of CO_2, and H_2 all decrease, especially for the content of H_2 gas, because of the effects of water gas reaction and the reverse water gas shift reaction. Additionally, rising temperature leads to an increasing concentration of CO and decreasing concentration of CO_2 in the Boudouard reaction until the temperature reaches 1000 °C, at which point the reaction only generates CO (Mathieu *et al*. 2002; Kuo *et al*. 2012). For these reasons, the overall concentration of CO grows faster than its decreasing rate. Therefore, CO concentration increases, while CO_2 concentration and gasification yields undergo a slow decline and gasification efficiency remains unchanged. In the heating process, the chemical equilibrium constant of methanation reaction becomes reduced, as a consequence of which CH_4 concentration is almost reduced to zero (Kuo *et al*. 2012). In the gasification process, H_2 concentration, gas yield, and gasification efficiency reaches peaks of 24.11%, 2.09 m^3/kg, and 83.56% at 700 °C, respectively. When the gasification temperature rises to 1000 °C, simulation results show that CO has the highest concentration in bio-gas and becomes the most combustible component. Detournay *et al*. (2011) conducted a high-temperature gasification in a fluidized bed, showing that the volume fraction of CO, 38%, is larger than the volume fraction of H_2, 33%, at 900 °C, which is close to the simulated results.

The Effects of Gasification Pressure

Gasification pressure is a very important factor in the gasification process. Figure 4 shows the effects of the change in gasification pressure from 0.1 to 5 MPa on the results of gasification when 1 kg/h rice husk enters at 700 °C with 1.38 kg/h air at 25 °C and 0.1 MPa.

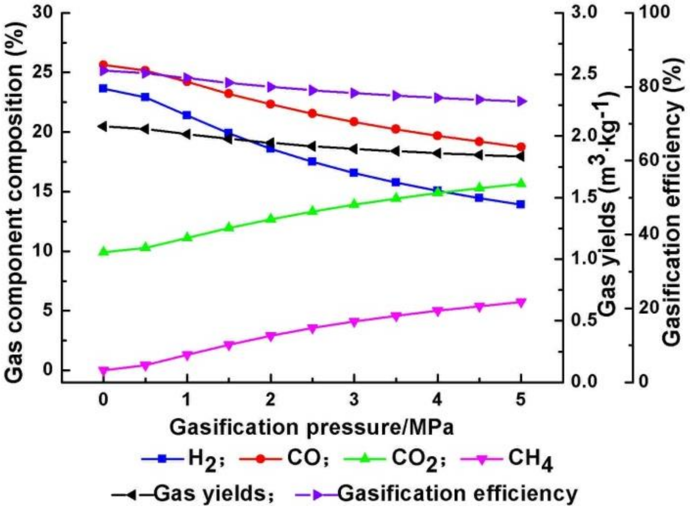

Fig. 4. Effect of gasification pressure on gas composition, gas yield, and gasification efficiency

The line graph above shows that, with increasing gasification pressure, the contents of H_2 and CO decrease, while the contents of CO_2 and CH_4 increase and gasification efficiency and gas yield slowly decrease. It is estimated that H_2 and CO concentrations are reduced by 12.15% and 8.95%, respectively, while CO_2 and CH_4 concentrations are increased by 7.42% and 7.30%, respectively, which causes a decrease of gas yield and gasification efficiency by 0.30 m^3/kg and 10.27%, respectively. The constant reduction of H_2 concentration and the constant increase in CH_4 concentration content may occur for the following reasons: ① According to Le Chatelier's principle, pressurization causes the balance of the steam reforming reaction of CH_4 to move in the direction to reduce volume, which gives rise to a slowing down of the positive reaction and acceleration of the reverse reaction. Consequently, some H_2 and CO are consumed to generate CH_4. ②Pressurization makes the balance of the methanation reaction move in the direction of the positive reaction, and H_2 is constantly consumed into CH_4.

Similarly, ① the major reaction is in the direction to generate CO_2, which has relatively little increase in volume under pressure. The reaction to generate CO is restrained, so that the content of CO decreases while the content of CO_2 increases. ② Because of the restraint of the positive direction of the Boudouard reaction during pressurization, CO_2 consumption is reduced, which results in increasing CO_2 concentration and reducing CO concentration. The results are close to the gasification experiment conducted by Detournay *et al* (2011) on a fluidized bed reactor using oak and momiki.

According to the equation of state, the gas volume is greatly reduced with increasing pressure, resulting in a corresponding decline in dry gas yield. Although H_2 concentration decreases, the content of CH_4, which has a high calorific value, increases. Therefore, the overall gas calorific value increases, which means pressurization is beneficial for CH_4 generation using agricultural waste as the raw material to produce high-calorific value syngas.

The Effects of Equivalence Ratio

Equivalence ratio is a very important factor when biomass air gasification is conducted in a fluidized bed. Figure 5 demonstrates how the results of gasification change according to a variation of equivalence ratio (ER) from 0.22 to 0.48 during gasification at 700 °C and 0.1 MPa. It can be seen from Fig.5 that when ER is lower than 0.3, with increasing air ER, the volume fractions of H_2 and CO increase, while CO_2 and CH_4 concentrations decrease.

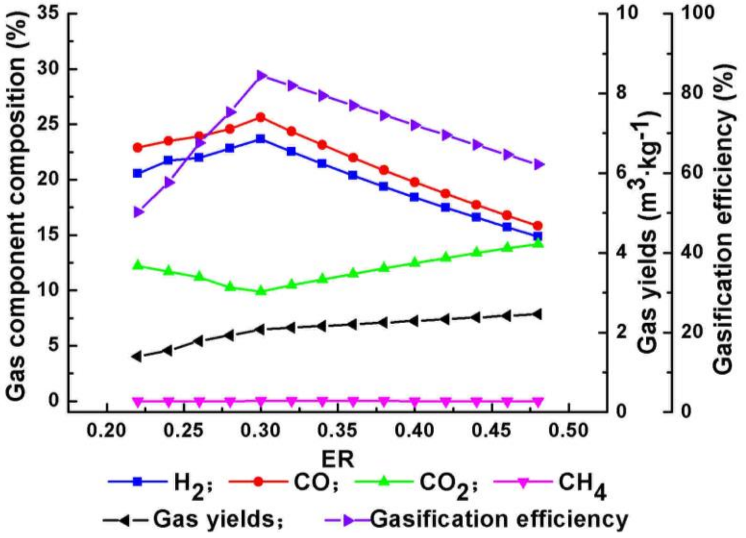

Fig. 5. Effect of equivalence ratio on gas composition, gas yield, and gasification efficiency

Minimal ER leads to incomplete gasification, which leads to the formation of a large amount of char and produces syngas with a low heat value (Chen *et al.* 2009). With increasing air intake volume, combustible components in biomass quickly burn and release plenty of heat, which promotes the incomplete pyrolysis of volatiles. Meanwhile, incomplete pyrolysis makes the increase in the input quantity of the gasification agent play a dominant role, leading to an increase in H_2 and CO production and a decrease in CO_2 production. Because of the increase in air volume, the large amount of N_2 makes the CH_4 concentration decrease. An increasing amount of gasification agent gives rise to the increasing gas yield with the increase of ER. Oxygen included in the gasification agent accelerates pyrolysis reactions and reduction reactions, which further promotes the pyrolysis of tar involved in the pyrolysis products. There is plenty of generated combustible gas, which increases gas yield and the calorific value of syngas, and gasification efficiency is sharply increased.

The simulation results show that optimal ER for husk gasification is 0.3, when the contents of H_2 and CO and gasification efficiency reach maximum values of 23.65%, 24.93%, and 85.92%, respectively. When ER is above 0.3, the pyrolysis reaction is fundamentally complete, with continued combustion of the combustible components CO, H_2, and CH_4. On the one hand, with the increase of O_2, CO and O_2 undergo an oxidation reduction to generate CO_2. On the other hand, an increase in the amount of O_2 promotes the complete reaction of fixed carbon and O_2, which leads to a decrease in CO concentration and an increase in CO_2 concentration. The increase in ER causes the intense combustion of combustible components releasing heat. From one side, a large amount of heat promotes a water-gas reaction in an endothermic positive direction, while a large amount of fixed carbon reacts with O_2, leaving a small part of water to participate in the water-gas reaction, leading to less production of H_2. From another aspect, a part of H_2 concentration reacts with O_2 and generates steam, resulting in the eventual reduction of H_2 concentration. In this process, because of the more thorough reaction in gasification, a second pyrolysis of tar is strengthened and the more combustible components are produced by an oxidation reaction to increase the calorific value of syngas. Meanwhile, a large amount of N_2 brought on by air dilutes the calorific value of bio-gas. These two aspects cause the calorific value of syngas to decrease with increasing ER. The slow increase in gas yield eventually causes a decline in gasification efficiency. When the ER reaches a very high level, excessive complete combustion generates CO_2 and H_2O at the expense of CO and H_2 (Kuo *et al*. 2012).

CONCLUSIONS

1. As seen from the comparison of predictions and the actuality, the tendency of changes in the gas component composition, gas yield, gas calorific value, and gasification efficiency in the gasification simulation results with equivalence ratio ER were in accordance with test results.
2. From the model, when the gasification temperature increased, the CO content in combustible syngas increased while the content of CO_2 and CH_4 decreased. However, the H_2 content and gas yield increased initially and then decreased. The gasification efficiency increased and then remained at a stable value. The H_2 content, gasification efficiency, and gas yield reached peaks of 24.11%, 83.56%, and 2.09 m^3/kg, respectively, at 700 °C.
3. As is simulated, with increasing gasification pressure, the contents of CO_2 and CH_4 continued to increase, the H_2 and CO contents decreased, and gasification efficiency and gas yield were slightly reduced. The H_2 and CO contents were reduced by 12.15% and 8.95%, respectively, and the CO_2 and CH_4 contents increased by 7.42% and 7.3%, respectively. Gas yield and gasification efficiency decreased by 0.30 m^3/kg and 10.27%, respectively. Pressurized conditions are conducive to the formation of CH_4 and rapidly increased the calorific value of syngas.
4. Predicted by the model, the optimal ER for gasification is approximately 0.3. Under these conditions, H_2 and CO concentrations and gasification efficiency reached their highest values, 23.65%, 24.93%, and 85.92%, respectively. When the ER was relatively small, hydrocarbon pyrolysis was not completely carried out; when the ER was relatively high, the introduction of a large amount of N_2 and O_2 made the combustion components undergo a severe combustion reaction. In addition, the increase in N_2 content caused a reduction of the combustible components of H_2 and CO and a decrease in gasification efficiency.

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