Experimental Investigation of Biomass Gasification in a Fluidized Bed Reactor

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This paper aims to catch the influence of various operating conditions and catalyst addition on the property of gas product and tar evolution. The gasification of three local biomass samples (sawdust, peanut shell, and wheat straw) was performed using a fluidized bed gasification reactor, and the gas product and liquid tar were analyzed with gas chromatography (GC). First, the influence of biomass property, gasification temperature, and air equivalence ratio was investigated. The biomass feeding rate was set at ∼2.37 kg/h; the furnace temperature variant was between 750 and 850 °C; and the equivalence ratio (ER) was 0.15−0.35. It can be observed that a lower heating value (LHV) of gas product from sawdust is higher than peanut shell and straw, while the tar content is also much higher than the other two samples, which might be attributed to the high volatile content. At 800 °C, with the increase of ER, the gas yield increased rapidly from 1.14 to 1.93 m^3/kg, while the LHV decreased from 7.09 to 3.26 MJ/m^3. Meanwhile, the variation of ER also showed a great effect on tar species. With the increase in temperature, combustible gas content, gas yield, and LHV all increased significantly, while the tar content decreased sharply from 13.24 to 6.53 g/m^3, which indicated that high temperature was favorable for biomass gasification. Then, three additives (dolomite, magnesite, and olivine) were introduced into the gasification process as catalyst for tar cracking. It is great for the upgrading of gas product quality, and tar removal efficiencies are all above 50%. It is significant for the development of biomass gasification technology.

1. Introduction

Biomass is an ideal renewable energy with advantages of lower sulfur, CO_2 neutral emission, and abundance resources. Biomass resource is huge in China, with 650 million tons of straw and 300 million tons of forest (sawdust and woodchip) wastes generated in 2000, and the energy contained equals 500 million tons of standard coal; furthermore, it increased greatly annually.[1] It is quite urgent and necessary to search an efficient and clean technology to convert biomass resources to energy.

Gasification, converting biomass to gaseous fuel at high temperatures (700−900 °C) with air, oxygen, or steam, is a promising technology.[2,3] The gasification behavior was affected by variant parameters.[4,5] Narvaez et al.[6] reported that, with temperature increasing from 700 to 900 °C, H_2 and CO yield increased greatly, while CO_2 vol % decreased from 16 to 13%. However, low heating value and high tar content of gas product are still blocking the use and commercialization of biomass gasification.

To remove tar and improve the overall efficiency of the system in producing a high quality of fuel gases, various chemicals, including dolomite,[7,8] magnesite,[9] olivine,[10,11] K_2CO_3, Na_2CO_3, Ni/Al_2O_3, etc.,[12−14] were introduced in as catalysts, to increase the economic feasibility of the biomass gasification process in the gas product. As the low cost of minerals, such as dolomite, magnesites, and olivine, they were used widely in the industry biomass gasification unit.[15] Rapagna et al.[11] studied the catalytic activity of olivine and dolomite, and high tar removal efficiency (90%) was observed.

However, the gasification property of local biomass resources in a fluidized bed gasifier was rarely reported. Therefore, the gasification of a local typical biomass sample (sawdust, peanut shell, and wheat straw) was performed in a fluidized bed reactor, and the influence of variant operating conditions, including ER (0.15−0.35), furnace temperature (750−850 °C), and natural minerals (dolomite, magnesite, and olivine) on the property of fuel gas and tar removal were investigated in depth.

2. Sample and Experimental Method

2.1. Sample. The biomass samples used are the local typical agricultural and forest wastes: sawdust, peanut shell, and wheat

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Table 1. Physical and Chemical Property of Biomass Samples

	sawdust	peanut shell	wheat straw
particle size (mm)	≤1	≤3	≤1
stack density (g/cm^3)	0.11	0.13	0.20
low heating value (MJ/kg)	15.41	16.28	13.67
Ultimate Analysis (wt %, on Air-Dried Basis)			
C	45.76	43.53	38.09
H	6.74	6.54	6.15
O	37.85	34.04	37.31
N	0.07	2.24	0.70
S	0	0.12	0.06
Proximate Analysis (wt %, on Air-Dried Basis)			
fixed carbon	15.82	17.99	14.54
volatile	74.60	68.48	67.77
moisture	6.11	8.84	4.13
ash	3.47	4.69	13.56

Table 2. XRF Analysis of Biomass Sample Ash (on Ash Basis wt %)

sample	K_2O	Na_2O	Al_2O_3	Fe_2O_3	CaO	TiO_2	SiO_2
sawdust	10.45	1.50	4.97	1.21	17.54	0.77	52.66
peanut shell	16.84	1.71	6.78	4.66	10.78	1.35	49.61
wheat straw	15.58	1.40	0.76	5.63	8.33	1.65	55.39

straw. The sample was ground and sieved, and the particle size was between 1 and 3 mm. The proximate and ultimate analyses were analyzed using TGA2000 (Las Navas, Spain) and CHNS analyzer (EL-2, Vario, Germany), respectively. The low heating value (LHV) was measured using a bomb calorimeter (Parr 6300, Parr Instrument Company, Moline, IL). The result is shown in Table 1. It can be observed that the biomass samples showed a higher volatile and low N and S content, the heating value is ~15 MJ/kg. They are an ideal renewable energy resource, with minor environmental pollution. The inorganic matters in biomass samples were analyzed using X-ray fluorescence spectroscopy (XRF) (results shown in Table 2). They are on an oxide basis as a percent of the ash mass. The main inorganic elements in biomass samples are Si, K, Na, Ca, Al, Fe, etc. It was found that sawdust showed the highest CaO, while wheat straw showed the highest SiO_2 and peanut shell showed the higher K_2O.

To find a suitable tar cracking catalyst for industrial biomass gasification running, three natural minerals (dolomite, magnesite, and olivine) were introduced as catalysts for tar cracking. They were taken from some refractory factory without pretreatment in low quality at low cost. The main components of dolomite are MgO (31%) and CaO (52%) after calcined at 1050 °C. The main components of magnesite are MgO (47.81%), CaO (2.35%), and CO_2 (49.84%). The main components of olivine are MgO (44%), SiO_2 (38%), and Fe_2O_3 (8%). There are still some other minor inorganic matters in the three additives. The particle size of minerals was ground to less than 1 mm and dry-mixed with the original biomass sample prior to feeding. The weight ratio of mineral/biomass material is 0.1.

2.2. Experimental Setup and Method. The gasification experiment was performed in a fluidized bed (FB) gasification system as shown in Figure 1. It consisted of a feedstock feeder, electrical furnace, and stainless-steel cylinder (inside diameter of 60 mm and height of 3 m) with an air distributor, cyclone separator, liquid condenser, gas cleaner, and gas product analyzer. The biomass was fed with a screw feeder continuously at ~2.37 kg/h into a hot FB gasifier, which was heated a to selected temperature previously; simultaneously, the whole gas line (FB and three cyclone separator) was heated to 300 °C, in case tar was condensed along the gas line. Then, the biomass sample was taken through a series of complicated physical and chemical reactions, which included heating, drying, pyrolysis, gasificiation, etc.[16] Biomass samples cracked to hydrogen-rich fuel gas (H_2, CO, CO_2, CH_4, etc.) and solid char with some liquid tar. The gas product was cleaned by a cyclone separator and then passed through the liquid condenser and gas-cleaning unit (referred to in Figure 2); finally, it was analyzed using a gas analyzer online. After gas product and solid char were separated with a cyclone separator, the gas phase was taken to gas and tar sampling equipment, as shown in Figure 2. It was set up following a previous report.[17] Liquid tar was condensed and solved in acetone solution, and the yield was measured by weighing the bottle before and after each trial. The volume of gas product was checked using a gas flow meter.

The gas product was measured with a quad-channel Microgas chromatograph (GC) with a thermal conductive detector (Agilent 3000, Germany, Agilent Technologies, Inc.) online, with helium as a carrier gas. The four channels are MolSieve 5A PLOT, plot U, Al_2O_3, and OV-1, respectively. The applied columns were molecular sieve 5A (for H_2, CH_4, and CO) at 110 °C, plot U (for CO_2, C_2H_4, C_2H_6, and C_2H_2) at 100 °C, and plot Al_2O_3 (for C_3H_6 and C_3H_8) at 140 °C.

The main component of tar was measured by a gas chromatograph−mass spectrometer (GC−MS Finnigan TRACE). The detecting method was described as follows. The column used was a capillary column DB-1301 (30 m × 0.25 mm inside diameter, 0.25 μm film thickness) with helium as a carrier gas with a constant flow of 0.5 mL/min. The GC initial oven temperature was held at 50 °C for 5 min and then programmed to increase to 250 at 10 °C/min. The oven temperature was held at 250 °C for 20 min. The mass spectrometer ion source was at 200 °C. MS was operated in electron ionization mode with 70 eV ionization potential, and a m/z range from 30 to 500 was scanned. The identification of the peaks is based on computer matching of the mass spectra with the National Institute of Standards and Technology (NIST) library. However, tar species were just analyzed semi-quantitatively, because the composition of tar is very complicated and it is difficult to specify the components and determine yield in detail. However, the peak height/area can also show the variant tendency of the main species of tar under variant conditions. However, to catch the influence of catalyst addition on tar property, the yield of the main light species from different biomass gasification with and without catalyst was analyzed quantitatively.

3. Results and Discussion

3.1. Influence of Biomass Material on Biomass Gasification. The main gas products are CO, CO_2, H_2, CH_4, and C_2H_4, with trace C_{2+} (<1%). The main combustible gas product distribution of gas products from different biomass gasification is shown in Figure 3 (800 °C, ER = 0.25). Because the content of $C_{2,\ 3}$ is too low, it was not shown here. The LHV of the gas product was calculated following previous literature[18] and shown in Figure 3. It can be observed that sawdust showed higher H_2 and CO content, while that of wheat straw is quite low. It might be attributed to the variant chemical components of different biomass samples, because sawdust showed higher volatile and H content but wheat straw showed lowest volatile and H content. At the same time, the LHV of gas products verified greatly with different biomass materials. The order of LHV of the gas product from three biomass sample gasifications is as follow: sawdust > peanut shell > wheat straw. It is consistent with that of the volatile content and the organic elements C and H of biomass materials. The tar yield of different biomass gasifications is showed in Table 3. It can be observed that the values are all above 6 g/kg of biomass sample and sawdust showed the highest tar yield, while peanut shell is much lower. It might be attributed to the higher O content of sawdust, while peanut shell was the lowest one.

To classify the components in detail of liquid tar, tar from

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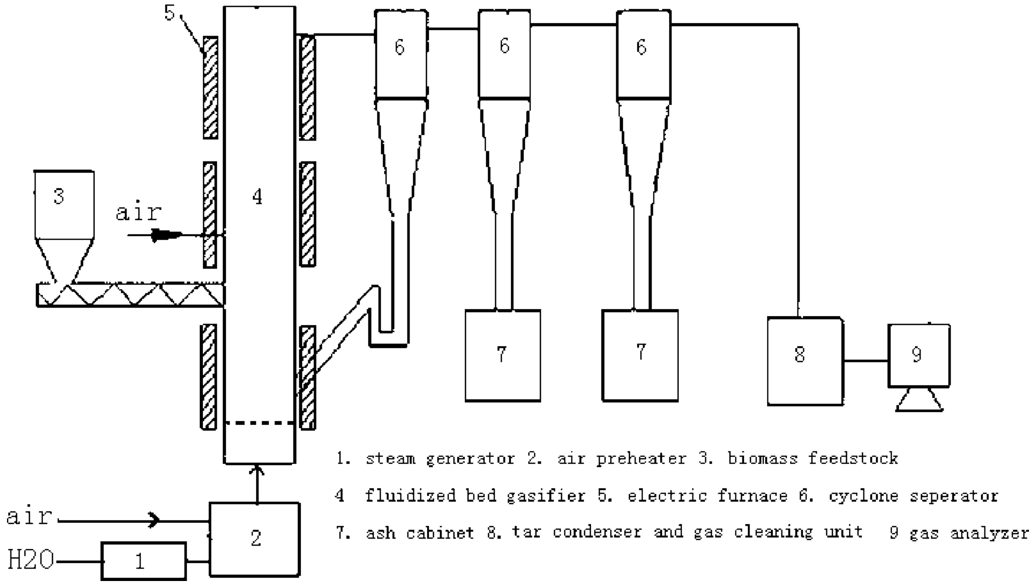

Figure 1. Working flow of the fluidized biomass gasification system.

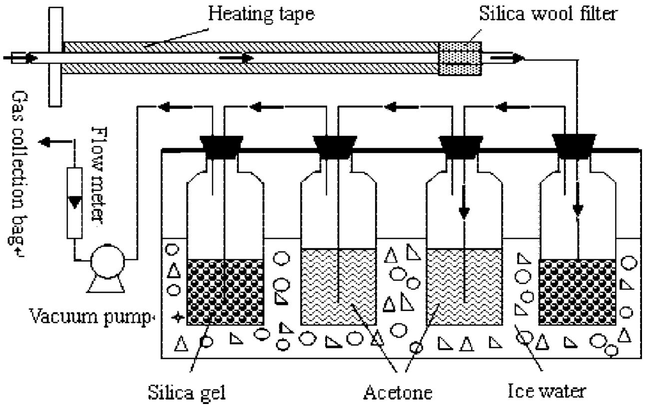

Figure 2. Profile of the gas and tar sampling system.

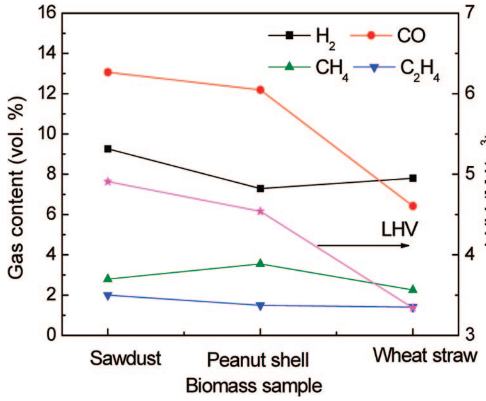

Figure 3. Gas products and LHV from the gasification of different biomass samples.

biomass gasification was investigated using GC−MS. One typical GC−MS chromatogram showing total ion count (TIC) versus retention time is given in Figure 4. It can be observed that the component of tar is very complicated, with more than 20 GC peaks shown. The main components of tar are benzene, phenol, naphthalene, and indene, with some C_{18+} hydrocarbon. The total tar in the product gas could be defined as the summation of the light and heavy tar. The molecular weight of the light tar is between 78 and 202, while that of heavy tar was larger than 202.[19] From GC spectra and MS library, it can be known that the light tar is simple with methylbenzene (4.2 min), styrene (7.07 min), phenol (8.9 min), indene (10.7 min), and naphthalene (12.6 min) and the retention time is less than 20 min, while that of heavy tar is much more complex, with large mounts of GC peaks, with a retention time larger than 20 min, and the components are acid, alcohol, etc., with a larger molecule (C_{18+}). From GC spectra, it can be known that the value of light tar species of sawdust is much higher than that of the other two biomass samples. The yield of light tar and heavy tar from three biomass gasifications are shown in Table 3. The light tar amount was obtained from the results from GC−MS and the summary of main light species. The heavy tar was determined by the difference of total tar yield and light tar yield. It can be found that the amount of light tar is higher than heavy tar for the three biomass samples, while the yield of light and heavy sawdust both showed a higher value than wheat straw and peanut shell. The ratio of light tar/heavy tar showed a different trend, with peanut shell showing a high value, while sawdust showed the lowest one. It might be attributed to the fact that the higher alkali metals (K_2O, etc.) in peanut shell ash and wheat straw promoted the cracking of tar; hence, the light part is relatively higher.

As mentioned above, sawdust showed higher combustible gas product and tar yields. In the following report, sawdust was chosen as a typical biomass sample to investigate the influence of the gasification temperature, ER, and catalyst addition on biomass gasification behavior.

3.2. Influence of ER on Biomass Gasification. The distribution profiles of the gas product from sawdust gasification under variant ERs are shown in Figure 5. The gasification temperature was set at 800 °C. The profiles of gas yield and LHV from sawdust gasification under different ERs are shown in Figure 6.

It can be observed that H_2, CO, and CH_4 content decreased greatly with ER increasing, while CO_2 content increased consequently. It might be caused by the fact that the combustible gas was burned, CO and CH_4 reacted with O_2 forming CO_2, and more char and hydrocarbon formed CO_2 directly with excessive air supplied. Consequently, the gas yield increased

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Table 3. Tar Yield of Biomass Gasification with Catalyst Addition (mg/kg of Biomass)

	sawdust				peanut shell				wheat straw			
tar composition	original[a]	dolomite	magnesite	olivine	original	dolomite	magnesite	olivine	original	dolomite	magnesite	olivine
TLT[a]	6104	3016	3283	3532	4421	3062	3044	2983	5107	2996	3187	2605
THT[a]	4816	209	232	451	1811	175		225	2585	335	254	395
TT[a]	10 920	3225	3515	3983	6232	3237	3044	3208	7692	3331	3441	3000
η[a]		70.5	67.8	63.5		48.1	48.1	51.5		56.7	55.3	61.0

[a] Original, without catalyst addition; TLT, the yield of light tar; THT, the yield of heavy tar; η, tar conversion rate (%).

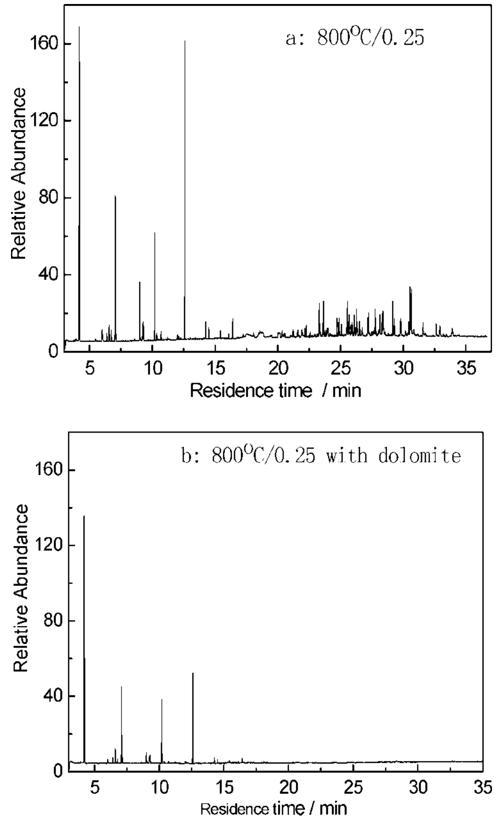

Figure 4. GC−MS chromatogram total ion of tar from sawdust gasification.

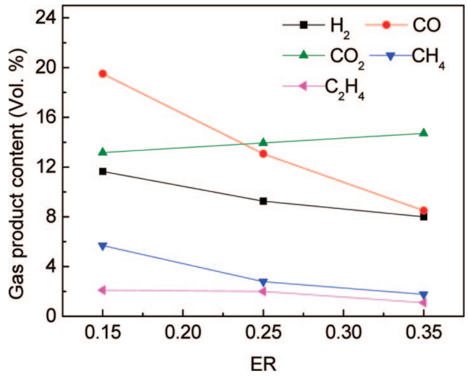

Figure 5. Gas products from sawdust gasification under different ERs.

rapidly from a 1.14 m^3/kg rise to 1.93 m^3/kg with ER increasing, while LHV decreased largely from 7.09 to 3.26 MJ/m^3. Because the CO_2 yield was increased greatly with O_2 content increasing at a price of CO, H_2, CH_4, etc. combustible gas diminishing. Meanwhile, the N_2 part in the gas product increased straightly with ER increasing; hence, the concentration of the product gas was diluted. Although the total yield of the gas product increased, the LHV decreased. No positive influence was shown on biomass gasification with ER increasing. It might be attributed that O/C in sawdust is quiet high (~0.6). Because

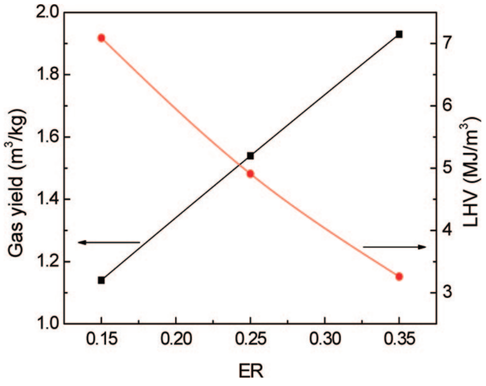

Figure 6. LHV and gas yield of sawdust gasification under different ERs.

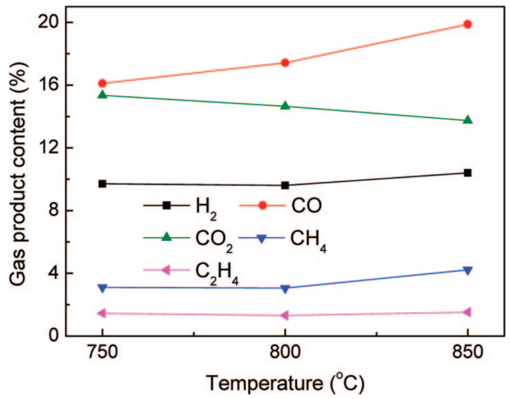

Figure 7. Profiles of gas products of sawdust gasification at different temperatures.

ER = 0.15, it is almost enough for C in the gasification system converting to CO. Hence, with ER increasing further, CO, H_2, etc. combustible gas should be combusted gradually with CO_2 and H_2O formed.

ER also displayed a great effect on tar property. First, with the air supplied increasing, the tar yield decreased largely, because more hydrocarbon was combusted with enough air supplied. Simultaneously, the main light species of tar varied greatly the changing trend of light species of tar. The yield of naphthalene increased significantly with ER increasing, while that of methylbenzene showed a different tendency. Because ER increased from 0.15 to 0.25, it increased slightly; however, it decreased obviously with ER increasing further. The part of indene showed a similar tendency with methylbenzene. However, styrene production declined linearly with ER increasing. The part of the oxygenated compound (phenol) decreased greatly with ER increasing.

3.3. Influence of Temperature on Biomass Gasification. The profiles of gas product distribution, gas yield, and LHV from sawdust gasification under different temperatures with ER set at 0.15 constantly are shown in Figures 7 and 8. As shown in Figure 7, the content of CO, CH_4, and H_2 all increased, while that of CO_2 showed a contrast trend. C_2H_4 did not show an obvious change. Because temperature is lower than 800 °C, the

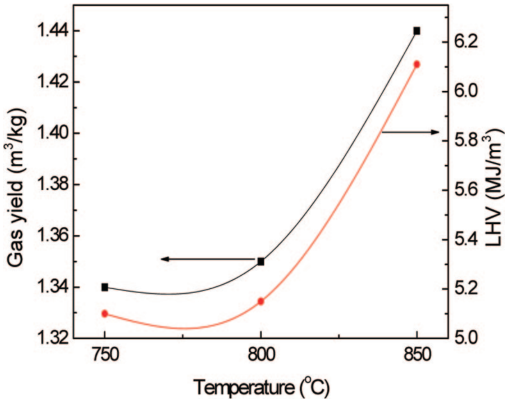

Figure 8. Total yield and LHV of gas products from sawdust gasification at different temperatures.

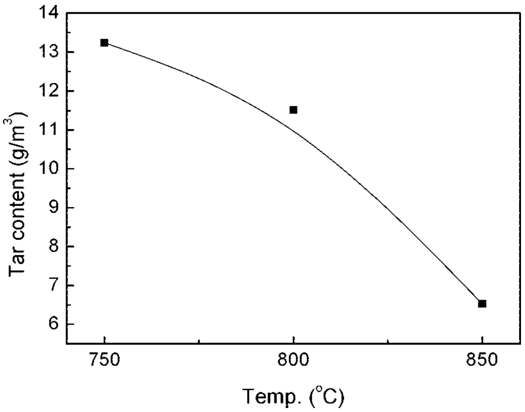

Figure 9. Tar content in gas product of sawdust gasification at different temperatures.

change extent is relatively tender; however, with temperature increasing further (>800 °C), the increasing extent was enhanced greatly. It might be attributed to the following reason. First, with temperature increasing, water shifting of carbon and hydrocarbon was enhanced greatly and, at the same time, the cracking and reforming of tar and char were accelerated largely. Consequently, CO, H_2, and CH_4 content increased, because a higher temperature is favorable for tar cracking with CO, H_2, CH_4, and other light hydrocarbon gas forming; hence, more gas product was formed.

From Figure 8, it can be observed that a higher temperature is favorable for the increase of gas yield and LHV. When the temperature is higher than 800 °C, with temperature increasing, the gas yield increased greatly from 1.35 to 1.44 m^3/kg, while LHV rose from 5.15 to 6.11 MJ/m^3. However, with a temperature lower than 800 °C, the increase trend was much tender. It was mainly attributed to the fact that char pyrolysis/gasification and tar cracking were preferred at high temperatures and the char conversion rate was enhanced greatly at higher temperatures. Hence, more combustible gas was formed, and the gas yield and LHV increased significantly. Consequently, the tar content reduced largely from 13.24 to 6.53 g/m^3 as the gasification temperature increased from 750 to 850 °C, as seen in Figure 9. The temperature displayed an important impact on tar cracking and reforming, which affects not only tar yield but also tar components. At higher temperature, more heavy tar cracked with a light gas product and tar species formed. Thus, in tar, the amount of light species increased at a price of heavy species diminishing.

3.4. Influence of Catalyst on Biomass Gasification. The property gas product releasing from sawdust gasification with/without dolomite addition is shown in Figure 10 (ER = 0.25).

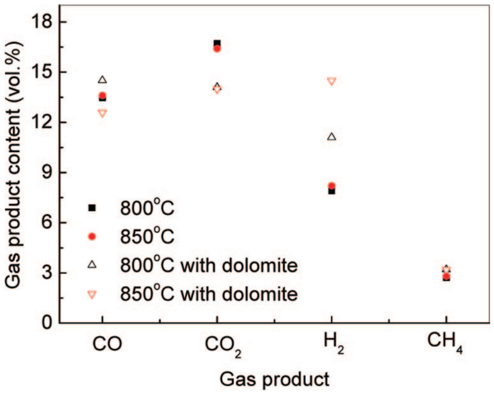

Figure 10. Gas product releasing from sawdust gasification with dolomite.

It can be observed that H_2 content was enhanced greatly with dolomite addition, while CH_4 increased slightly. With the temperature increasing, the increasing extent of H_2 was enhanced; it might be attributed to the fact that the catalyst for tar cracking and char reforming was much strong at higher temperature than lower temperature. CO showed a different trend at different temperatures with catalyst addition. At 800 °C, the content increased with dolomite addition, while it decreased greatly with the gasification temperature rising. The increase of H_2 might be the main contribution, because more H_2 was formed with tar cracking and char reforming.

When the two spectra in Figure 4 are compared, it can be observed that tar species verified greatly and the peak of heavy tar diminished, while the peak of light tar also decreased greatly. It indicated that mineral addition played a great effect on tar removal. The tar yields of biomass samples with catalyst addition are listed in Table 3 (800 °C and ER = 0.25). It can be observed that, with catalyst addition, the tar yield reduced greatly; however, biomass samples showed different selectivity for catalyst addition. For sawdust, dolomite is the most efficient on tar cracking, with tar removal efficiency of ~70%, while for peanut shell and straw, it is olivine; however, the removal efficiency is a little bit low, about 50−60%. With catalyst addition, the yield of heavy tar and light tar both decreased obviously. It might be attributed to the fact that catalyst addition not only enhanced the cracking of heavy tar but also the reformal of light tar (refer to Figure 4). Most of the heavy tar and polycyclic aromatic hydrocarbons were cracked and converted to light gas products. The variant of catalysts of three additives might be attributed to the different amounts of the main components, while the difference between tar properties of three biomass might play the main contribution to the variant selectivity, because sawdust showed a higher tar yield but peanut shell showed the lowest one. It can be derived that catalyst addition showed a higher catalytic value for tar cracking because a higher tar yield was shown for original biomass gasification.

In comparison to previous work,[11,13] it can be known that tar removal efficiency here is much lower. It might be attributed to the following reasons. First, the catalyst addition is natural mineral, and the purity is quiet low. The mineral was mixed with a biomass sample previously. It was fed into a gasifier with a biomass sample during trial, but the catalyst is much heavy than biomass materials. It fell to the bottom of gasifier, while the temperature of the bottom is too low (~600 °C) for minerals being calcined. Furthermore, the volatile part in solid biomass samples evolved out and carried the upper part and exhaust out quickly in a hot gasifier. The contact chance of the solid catalyst with volatile is minimal; hence, tar removal efficiency and the catalytic value of mineral addition are much lower than other work. The cracking mechanism and the upgrading method of tar removal with mineral addition will be studied in depth in the near future.

4. Conclusions

The gasification properties of three local biomass samples were investigated using a fluidized bed reactor combined with micro-GC and GC−MS. The main conclusions can be derived as follows.

Gasification behaviors displayed a close relation with biomass type. Sawdust showed a higher combustible gas (H_2, CO, and CH_4) and tar yield, as the higher volatile and hydrogen and oxygen content. The LHV order of product gas from different biomass samples can be elaborated as follow: sawdust > peanut shell > wheat straw.

With ER increasing, the gas yield increased rapidly from 1.14 to 1.93 m^3/kg, while the heating value was decreased largely from 7.09 to 3.26 MJ/m^3, as more carbon was converted to CO_2. Meanwhile, the variation of ER also had an important effect on tar yield and tar species. The amount of methylbenzene and naphthalene increased greatly with ER increasing, while that of phenol and styrene production decline obviously.

The temperature is an important factor to the biomass gasification. With the temperature increasing, combustible gas content increased greatly, total gas yield and LHV both increased greatly, and tar content decreased sharply from 13.24 to 6.53 g/m^3. A high temperature is favorable for biomass gasification. It has a significant effect on the upgrading of gas quality and quantity.

Catalyst addition showed great catalytic value on biomass gasification. The releasing of light gas products (H_2, CH_4, and CO) was enhanced greatly with dolomite, olivine, and magnesite addition; however, the biomass sample showed variant adoptability. Tar removal efficiency varied from 48.1 to 70.5%, while sawdust gasification showed highest tar removal efficiency with dolomite addition. With regard to tar species, heavy tar components decreased as catalyst addition with more light gas products formed.

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