Experimental Study on Co-gasification Characteristics of Biomass and Plastic Wastes

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Co-gasification of biomass (rice straw) and polyethylene (PE) was conducted in a lab-scale entrained-flow gasifier. The influences of PE proportion, reaction temperature, and equivalence ratio on producer gas composition, gasification index, and tar yield were investigated. In addition, the effects of dolomite and sodium carbonate (Na_2CO_3) catalysts on the co-gasification process were also examined. Increased PE proportion led to an increased lower heating value (LHV) of producer gas as well as an increase in tar yield. In addition, a higher reaction temperature could improve both gas quality and gasification indices significantly. An equivalence ratio (ER) of 0.25 led to a relatively high LHV and low tar yield. Na_2CO_3 showed a better tar removal efficiency than dolomite. Dolomite increased the LHV of producer gas, while Na_2CO_3 decreased the LHV. The difference in the catalyst proportion did not cause any significant change in the gas composition and gasification indices. The producer gas with the highest LHV and lowest tar yield was obtained by the co-gasification of 80% (w/w) straw, 20% (w/w) PE, and 3% (w/w) dolomite.

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INTRODUCTION

The energy crisis and environmental pollution are two serious concerns in today's world. Sources of renewable energy have been explored to decrease these problems and to protect the environment. In China, straw wastes have not been utilized in a proper way, and their disposal has caused air pollution. Moreover, the consumption of plastics continues to increase every year. Consequently, the amount of plastic wastes has also increased, and their disposal causes serious white pollution, particularly in rural areas.

Ekmann *et al*. (1998) reported that energy recovery from wastes may be an economically attractive source of energy as a part of an integrated waste management plan. To find new alternative fuels and new technologies to reduce the negative environmental impact of waste accumulation, the co-gasification of biomass and plastic wastes has been widely studied (Pinto *et al*. 2003; Pohorely *et al*. 2006; Wilk and Hofbauer 2013). Air gasification of a fraction of mixed plastic wastes was conducted by Kim *et al*. (2011) to yield a clean and high-calorie producer gas. Some researchers reported that the co-gasification of biomass and coal was feasible (Chmielniak and Sciazko 2003; Kumabe *et al*. 2007; Kern *et al*. 2013). Fluidized bed steam gasification has been demonstrated by Pinto *et al*. (2002) to be a possible way of converting biomass and plastic waste into fuel gases. The proportion of polyethylene (PE) in PE/biomass mixtures ranged from 0 to 60%, and the gasification temperature was 835 °C (Pinto *et al*. 2002). Gasification of mixtures

of PE and woodchips, in which the proportion of PE varied from 0 to 100%, had been investigated by Ahmed *et al*. (2011). They found that a small proportion of biomass material to PE could result in a higher energy yield than that obtained with 100% PE.

In previous studies, tar generated in the gasification was found to be a serious problem, leading to decreased energy utilization efficiency and equipment damage (Rabou *et al*. 2009; Dabai *et al*. 2014). However, the use of catalysts can reduce tar in gasification (Brage *et al*. 2000; Devi *et al*. 2003). The catalysts used in biomass gasification are divided into three groups: earth metal, alkali metal, and nickel catalysts (Basu 2010). Dolomite is widely used as an earth metal catalyst in biomass gasification because it is very effective for the disposal of tar and is inexpensive and widely available. Furthermore, dolomite is more effective than calcite and magnesite (Delgado *et al*. 1996). Alkali metal catalysts can reduce methane in the producer gas through a reforming reaction. Potassium is notorious for agglomeration in fluidized beds, which decreases the catalytic benefit. Nickel catalysts are highly effective and work best in the secondary reactor, but they are more expensive than dolomite. This study is focused on dolomite and alkali metal catalysts because they are effective, cheap, and easily available.

Most previous co-gasification experiments using biomass and plastic have been carried out in fluidized beds, where potassium is notorious for agglomeration. Furthermore, the gasification temperatures adopted in previous studies were not relatively high (Pinto *et al*. 2002; Ahmed *et al*. 2011), and only one type of catalyst was used.

In this study, the effects of PE proportion, equivalence ratio (the equivalence ratio is the ratio of the actual air-fuel ratio to the stoichiometric air-fuel ratio), reaction temperature, and the type of catalyst on co-gasification were investigated in a lab-scale entrained-flow reactor, and tar yields under different reaction conditions were analyzed. The proportion of PE ranges from 0 to 30% because there is much more straw waste than plastic waste in Chinese rural areas. This study aims to obtain producer gas with higher lower heating value (LHV) and lower tar yield, which is conducive to the utilization of energy and the elimination of plastic waste in rural areas.

EXPERIMENTAL

Materials

Rice straw and PE were used as the materials for entrained-flow gasification experiments. Both were pulverized and then sieved into particles with the size of less than 0.3 mm. The results of ultimate and proximate analysis of the samples are listed in Table 1. Dolomite ($CaCO_3.MgCO_3$) and sodium carbonate (Na_2CO_3) were chosen as catalysts in this study.

Table 1. Ultimate and Proximate Analysis of Straw and PE (as-received basis)

	Ultimate analysis (wt.%)					Proximate analysis (wt.%)				
	C	H	N	S	O	Moisture	Ash	Volatile	Fixed carbon	LHV(kJ/kg)
Straw	30.26	5.53	0.62	0.22	36.85	12.42	14.10	61.05	12.43	14395
PE	83.96	11.02	-	0.38	3.99	0.65	-	99.35	-	46753

Methods

The gasification plant used in this study is shown in Fig. 1. The plant consisted of a feeding system, an entrained-flow gasification reactor based on a Badzioch-type reactor (Badzioch and Hawksley 1970; Biagini *et al*. 2005), a gas supply system, a tar collection system, and other auxiliary facilities. The reaction tube height was 600 mm, and the inner diameter was 48 mm. The reactor was heated by 12 globars and surrounded by heat-insulating materials. A proportion integration differentiation (PID) control system was used to regulate the temperature in the reactor.
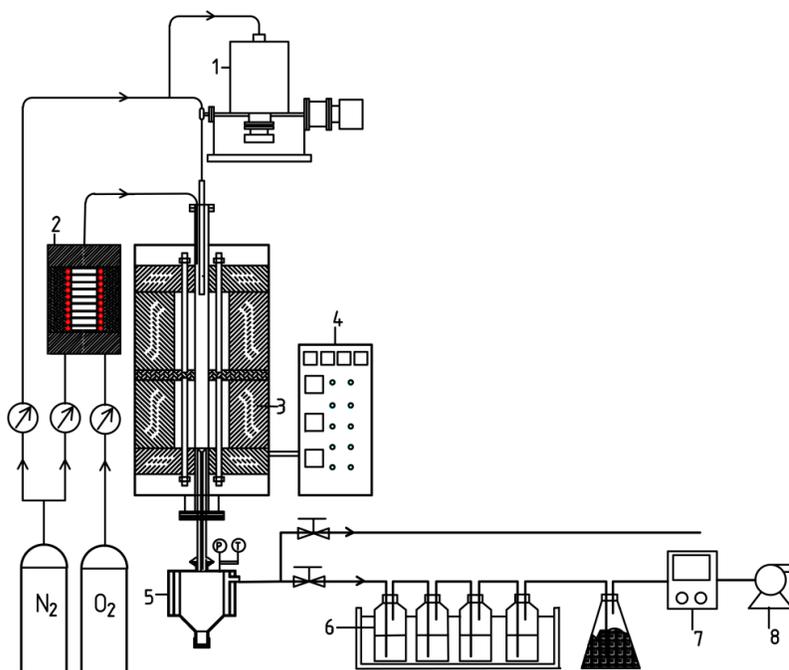
Fig. 1. Sketch of the experimental system; (1) feeder, (2) preheater, (3) entrained-flow reactor, (4) temperature controller, (5) hopper, (6) tar collection system, (7) gas flow meter, and (8) pump

After the reactor tube reached the selected temperature, the gasifying agent was introduced into the reactor. The gasifying agent included the feed gas (N_2) and the secondary gas (N_2 and O_2). Then, the biomass was fed into the reactor with the feed gas at a constant rate of 4 g/min.

Reaction products from the reactor entered a hopper and filter, where char and ash were collected. Tar contained in the flue gas was collected in the tar collection system using a cold trapping method (Claes and Chen 1997). Finally, three bags of the producer gas were collected as sample gases. To avoid the condensation of tar before tar collection, especially

for heavy tar condensation, the temperature of the hopper was kept above 220 °C. The experimental conditions are listed as follows: feed rate of 4 g/min, feed gas rate of 2 L/min, secondary gas rate of 4 L/min, preheating temperature of 500 °C, reaction temperatures of 800, 900, 1000, 1100, and 1200 °C, and ERs of 0.15, 0.20, 0.25, and 0.30.

The sample gas obtained in the gasification was analyzed by a GC-9160 (Shanghai Precision & Scientific Instrument; Shanghai, China) gas chromatograph to detect the concentrations of CO, H_2, CO_2, O_2, N_2, CH_4, and C_xH_y (C_2H_4 and C_2H_6). The yield of each gas could be calculated with N_2 as a tracer according to the data obtained from the chromatography (Qin *et al*. 2012).

Gasification tar yield, representing the mass of tar (mg) obtained from 1 g of material, can be calculated according to Eq. 1,

$$\text{Tar}_p = \frac{m_{\text{tar}}}{m \cdot t} \tag{1}$$

where Tar_p is the tar yield (mg/g); m_{tar} is the mass of tar collected (mg), m is the material feeding rate (g/min), and t is reaction time (min).

RESULTS AND DISCUSSION

The effects of PE content in the feedstock, reaction temperature, ER, and various catalysts on the gasification process were studied.

Effects of PE Proportion in the Feedstock
In this experiment, the PE proportion in the feedstock was varied from 0 to 30% m/m with a constant ER of 0.25 and gasification temperature of 1000 °C. As shown in Fig. 2, different proportions of PE in the feedstock caused the changes in the gas composition. When the proportion of PE in the feedstock was increased from 0 to 30%, similar trends for CO and H_2 were observed. The concentration of H_2 increased from 10.34% to 13.67%, and CO increased from 11.39% to 13.33%. The effect of PE proportion on the concentration of C_xH_y was significant. The concentration of C_xH_y increased from 0.78% to 2.45% as the PE proportion increased from 0 to 30%. The increased H_2 product was caused by cracking reactions of PE molecules, which also caused an increase in other gases.
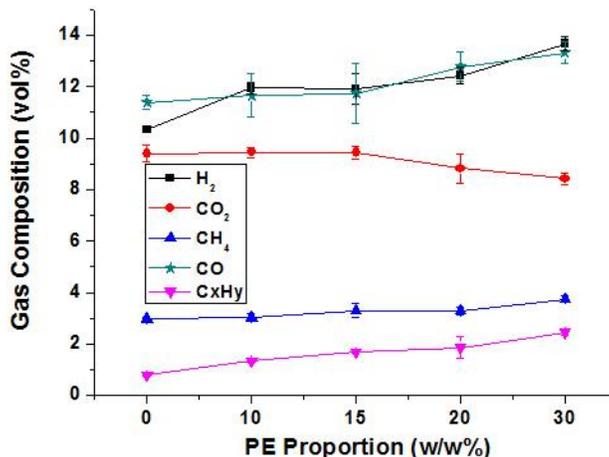
Fig. 2. Effects of PE proportion on gas composition

At high temperatures, the thermal energy supplied to the polymer is much higher than the covalent bond strength energy. Therefore, the bond breakage occurs throughout the molecule and the polymer is decomposed into small molecular fractions, which may include mostly hydrogen, methane, ethane, ethane (PE monomer), and heavier combinations of these units, some of which might be further cracked. The composition of the biomass, especially its hydrogen-to-carbon (H/C) ratio, has an important bearing on the pyrolysis yield. The H/C ratio is changed when PE is added into the feedstock.

The results were consistent with previous results (Pinto *et al*. 2002; Aznar *et al*. 2006), with the exception of the change in CO concentration in the current study. Pinto *et al*. (2002) found that, compared with the 0% PE proportion, the use of 20% PE led to an increase of approximately 73% H_2 concentration and a decrease of only around 27% CO formation. For the higher amounts of PE, the concentration of these gases remained almost constant. However, in the study by Aznar *et al*. (2006), a slight decrease in CO was observed when the PE content was increased. These opposing trends in CO concentration observed in different studies are primarily attributable to different gasification temperatures. The gasification temperatures in previous studies (Pinto *et al*. 2002; Aznar *et al*. 2006) were 835 °C and 850 °C, respectively. In this experiment, the gasification temperature was 1000 °C, which was much higher than those in previous studies. As an endothermic reaction, the reaction shown as (2) will be promoted when the temperature is relatively high. Therefore, the concentration of CO increased as PE content increased.

$$C + CO_2 \rightarrow 2\ CO + 172\ \text{kJ/mol} \tag{2}$$

Different media may have important influence on the gas composition. The water-gas shift reaction is probably one of the most important reactions for reaching final gas composition. This reaction may decrease the concentration of CO in the other two studies.

$$\text{Water- gas shift}\quad CO + H_2O \rightarrow CO_2 + H_2 \tag{3}$$
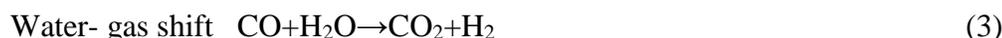
In addition, Aznar *et al*. (2006) and Pinto *et al*. (2002) chose pine as a feedstock, which had a carbon concentration of approximately 50%. However, the carbon concentration of straw in this study was approximately 30%. Compared with the carbon concentration of pine, the carbon concentration of feedstock in this study increased more significantly with the increase in PE content. The difference in the H/C ratio of feedstock could also influence the concentration of CO in producer gas.
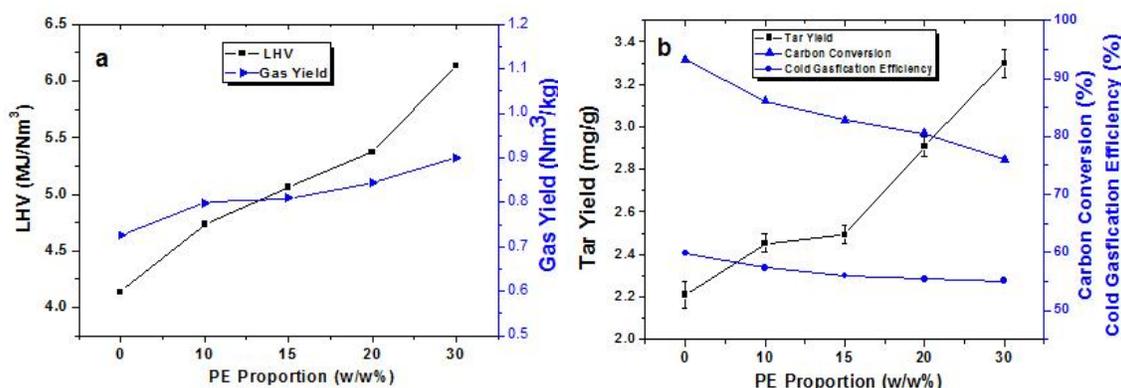
Fig. 3. Effects of PE proportion on gasification indices: (a) LHV and gas yield, (b) carbon conversion, cold gas efficiency, and tar yield

As shown in Fig. 3, the PE proportion had a noticeable effect on LHV and gas yield. At a PE proportion of 30%, the LHV of producer gas was 48.25% higher than that produced from single straw. LHV is related to gas composition, where an increase in LHV is caused by the increases in the concentrations of combustible components such as H_2, CO, CH_4, and C_xH_y. The producer gas yield also increased by 24.10% as the PE proportion was increased from 0 to 30%. This is due to the higher volatile content of PE relative to straw.

With increasing PE proportion, the cold gas efficiency decreased slightly from 59.83% to 55.11%, and carbon conversion decreased from 93.29% to 76.6%. Holocellulose and lignin in biomass mostly consist of weak R-O-R ether bonds (phenols, aldehydes, ketones), which can be easily released (Lapuerta *et al*. 2008). PE does not devolatilize, but is decomposed thermally through radical reactions. These reactions may start at higher temperatures than devolatilization, but they are faster than char gasification. The carbon concentration of PE (C_{ar} = 83.96%) is much higher than that of straw (C_{ar} = 30.26%). Therefore, PE may require more time to convert C into CO, CH_4, and C_xH_y. However, residence time of PE in reactor is not long enough, resulting in the decrease in carbon conversion.

Compared with pure straw, the mixed feedstock can produce more tar during gasification. At the PE content of 30%, the tar yield increased by approximately 50%. More tar is produced during PE pyrolysis, which leads to an increase in tar yield in the gasification of the mixtures with the higher PE proportion.

Effect of Temperature

Reaction temperature is an important factor of the gasification performance of both PE and straw. In this experiment, the proportion of PE was 20%; ER was 0.25; the temperature ranged from 800 to 1200 °C. As shown in Fig. 4, when the reaction temperature increased from 800 to 1200 °C, the concentrations of H_2 and CO increased from 5.15% and 6.89% to 23.36% and 17.10%, respectively. On the contrary, CO_2 concentration decreased sharply, from 13.23% to 4.16%. Concentrations of CH_4 and C_xH_y showed a slight decrease with the increase in temperature.
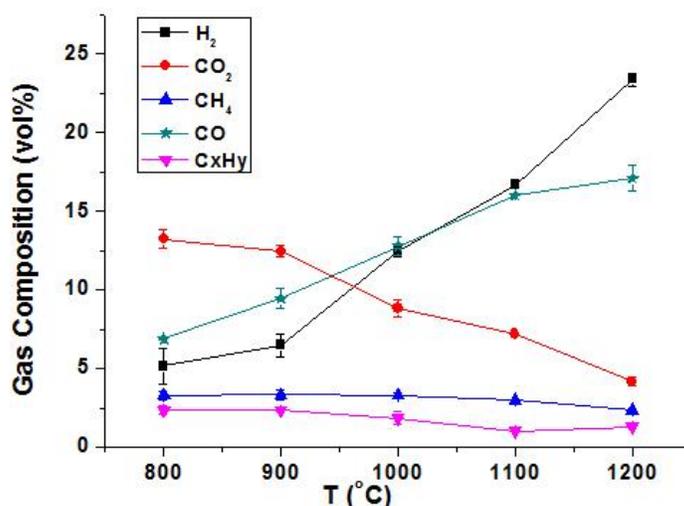
Fig. 4. Effects of temperature on gas composition

Reactions (2) and (4 through 6) are endothermic reactions, which are promoted by high temperature. Therefore, the concentrations of CO and H_2 increased with temperature rise. This also accounts for the decreases in CO_2, CH4, and C_xH_y.

$$C_mH_n + mCO_2 \rightarrow mCO + n/2H_2 \tag{4}$$
$$C + H_2O(g) \rightarrow CO + H_2 + 131kJ/mol \tag{5}$$

$$C_mH_n + mH2O \rightarrow mCO + (n+2m)/2 \tag{6}$$

Marked effects of temperature on LHV and gas yield can be observed in Fig. 5. When the temperature increased from 800 to 1200 °C, the LHV increased by 52.62% due to the increases in concentrations of CO and H_2 in the producer gas. At the same time, the gas yield increased from 0.59 Nm^3/kg to 1.25 Nm^3/kg. Both the rate of solid-gas reactions and producer gas yield increased with the increase in temperature.

When the temperature reached 1200 °C, the cold gasification efficiency jumped to 79.13% and the carbon conversion reached 88.94%. On the contrary, the tar yield declined quickly with temperature rise. The tar yield at 1200 °C decreased by 65.74% compared with that at 800 °C. The trend obtained in this experiment is consistent with previous results (Aznar *et al*. 2006; Rechulski *et al*. 2012). The decreasing tar yield trend can be interpreted as follows: as endothermic reactions, the tar cracking and reforming reactions are strengthened with temperature rise.
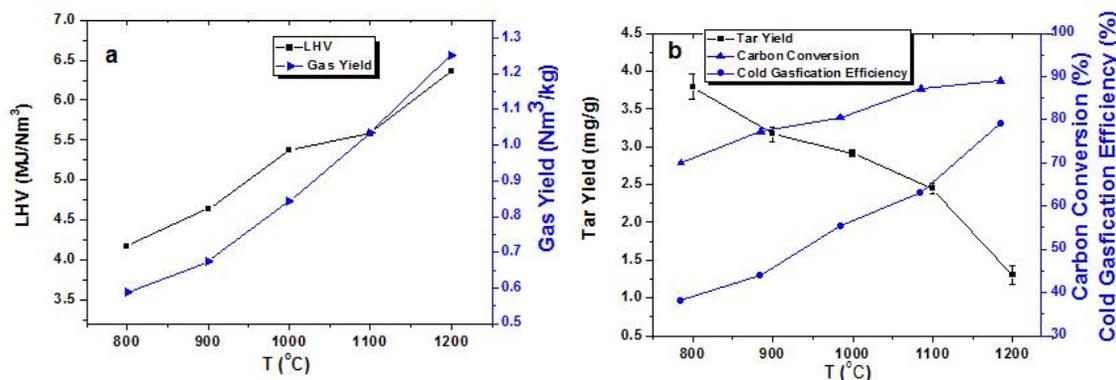
Fig. 5. Effects of temperature on gas indices: (a) LHV and gas yield, (b) carbon conversion, cold gas efficiency, and tar yield

Effect of ER

ER is another crucial factor in gasification. The effect of ER was studied in the range from 0.15 to 0.30 with a constant PE proportion of 20% at a gasification temperature of 1000 °C.
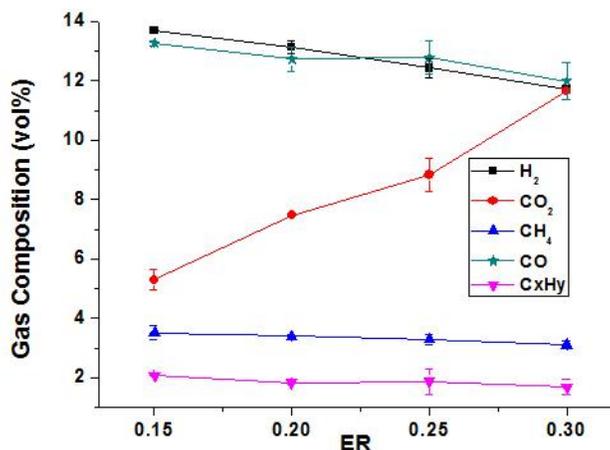
Fig. 6. Effects of ER on gas composition

As shown in Fig. 6, an increase in ER caused a decrease in all combustible gas contents. The concentration of H_2 decreased from 13.70% to 11.72%, and the concentration of CO decreased from 13.27% to 11.98%. However, the concentration of CO_2 increased rapidly from 5.28% to 11.66%. At a higher ER, more active oxidization reactions would occur, causing the increases in CO_2 and H_2O at the expense of CO and H_2.

The gasification indices are shown in Fig. 7. When the ER increased from 0.15 to 0.30, the gas yield showed little change; carbon conversion increased; LHV decreased by 13.38%; and cold gasification efficiency also decreased. When the ER increased from 0.15 to 0.30, the tar yield decreased rapidly from 5.39 mg/g to 0.97 mg/g. The higher ER allows more oxygen to react with volatiles in the flaming pyrolysis zone. The results are consistent with the findings of previous studies (Kinoshita *et al*. 1994; Yu *et al*. 2014). In the study by Kinoshita *et al*. (1994) with an ER of 0.27, phenols were nearly completely converted, and less tar was formed. However, although the higher ER reduced tar, it also reduced the quality of the gas.
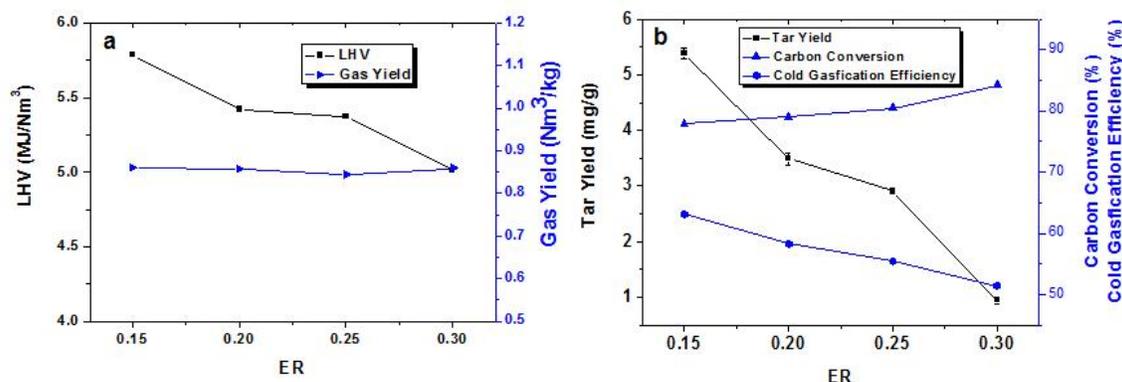
Fig. 7. Effects of ER on gas indices: (a) LHV and gas yield, (b) carbon conversion, cold gas efficiency, and tar yield

Effects of Catalysts

The effects of the catalysts on the gas yield, LHV, cold gasification efficiency, and tar yield were analyzed. The feedstock consisted of 20% PE and 80% straw, and the feed rate was 4 g/min in this experiment. Catalyst amounts of 3% (w/w) dolomite, 6% (w/w) dolomite, 3% (w/w) Na_2CO_3, and 6% (w/w) Na_2CO_3 were respectively mixed with the feedstock under the ER of 0.25 at a gasification temperature of 1000 °C.
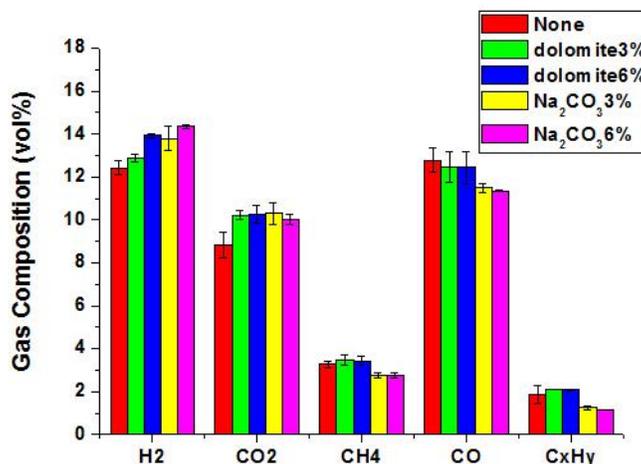
Fig. 8. Effects of catalyst on gas composition

The effects of catalysts on gas composition are shown in Fig. 8. When dolomite and Na_2CO_3 were respectively used as the catalyst in the gasification, the concentrations of H_2 and CO_2 increased gradually, while the concentration of CO showed a slight decrease. The effects of Na_2CO_3 were more obvious than those of dolomite. These catalysts promoted Reactions (4) and (6), causing an increase in H_2. Metallic ions were believed to be able to promote the generation of CO_2 and restrain the generation of CO. When the catalysts were mixed in the feedstock, partial metallic ions were on the surface of the feedstock, and other metallic ions were combined with alduronic acid in hemicellulose to produce glycuronate. Compared with alduronic acid, glycuronate releases carbonyls more easily, which leads to an increase in CO_2 generation (Tan *et al*. 2005). Dolomite increased the concentrations of CH_4 and C_xH_y due to reduction reactions of tar. Na_2CO_3 restrained the concentrations of CH_4 and C_xH_y because of a reforming reaction. Reforming, thermal cracking, and steam cracking are three major reactions that are responsible for tar destruction:

$$\left. \begin{array}{l} \text{- reforming} \\ \text{tars \quad - thermal cracking} \\ \text{- steam cracking} \end{array} \right\} \rightarrow CO_2 + CO + H_2 + CH_4 + ... + coke \downarrow$$
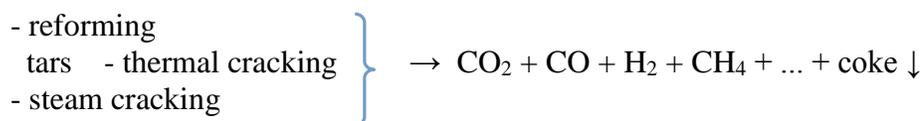
More coke was generated when Na_2CO_3 was used as the catalyst.

For both catalysts, the difference in the proportion did not cause a significant change in the gas composition.

As shown in Fig. 9, dolomite can promote LHV, cold gasification efficiency, carbon conversion, and gas yield, while Na_2CO_3 presents a negative effect on the four indices. When 3% dolomite was added to the feedstock, LHV increased by 4.43% and carbon conversion increased from 80.50% to 87.08% due to the increase in CO_2, CH_4, and C_xH_y contents. Consequently, cold gasification efficiency increased from 55.41% to 58.96%. When 3% Na_2CO_3 was added into the feedstock, LHV decreased by 11.60% and cold gasification efficiency decreased from 80.49% to 75.51% due to the decreases in CO, CH_4, and C_xH_y contents. Compared to 3% contents, 6% of dolomite or Na_2CO_3 showed no significant difference in gasification indices. When 3% and 6% (w/w) dolomite was added, the tar yield was respectively decreased by 23.16% and 26.11%.
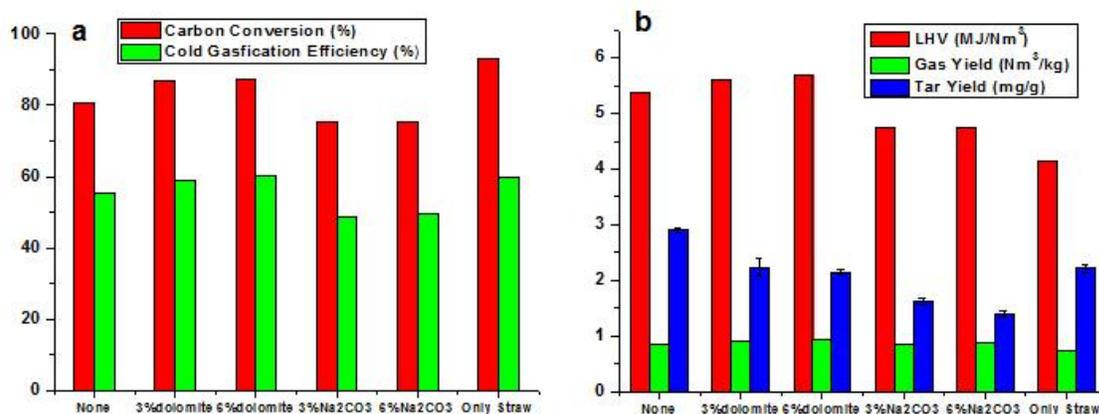
Fig. 9. Effects of catalysts on gas indices: (a) carbon conversion and cold gas efficiency, (b) LHV, gas yield, and tar yield

The higher proportion of dolomite led to a slight difference in tar yield. Na_2CO_3 showed more marked effects on tar yield than dolomite. When the Na_2CO_3 content increased from 3% to 6%, the decrease in tar yield only increased from 43.99% to 52.36%. Similar to dolomite, the increase in Na_2CO_3 did not reduce tar to a great extent. If economic efficiency is taken into account, 3% (w/w) catalysts in gasification are more suitable than 6% (w/w) catalysts.

Compared with pure straw gasification with an ER of 0.25 at a gasification temperature of 1000 °C, the co-gasification of 20% PE and 80% straw with 3% dolomite as the catalyst showed more valuable results. The LHV and gas yield increased by 35.42% and 24.31%, respectively, and the tar yield increased slightly. When 3% Na_2CO_3 was added to the co-gasification, although the tar yield decreased by 26.32%, the LHV and gas yield increased by 14.81% and 16.56%, respectively. Moreover, 3% dolomite had a more positive influence on the gasification indices than did both 3% and 6% Na_2CO_3.

CONCLUSIONS

1. The elimination of plastics through co-gasification was technically feasible. Co-gasification also increased the LHV of the producer gas significantly. LHV increased by 48.24% and gas yield increased by 24.10% when 30% PE was added into the feedstock. However, tar yield also increased with increasing PE proportion.

2. As the reaction temperature increased during co-gasification, the concentrations of H_2 and CO increased sharply and CO_2 decreased. The LHV, gas yield, carbon conversion efficiency, and cold gas efficiency increased with temperature rise. However, the tar yield was reduced significantly. High reaction temperatures were favorable to the gasification process.

3. In the experiments, a higher ER could reduce the tar yield, although the LHV also decreased and the gas yield remained nearly unchanged. In this experimental study, an ER of around 0.25 was an appropriate value.

4. With dolomite or Na_2CO_3 as the catalyst, the concentrations of H_2 and CO_2 increased gradually while the concentration of CO decreased slightly. However, dolomite increased the concentrations of CH_4 and C_xH_y, while Na_2CO_3 had the opposite effect.

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Dolomite could effectively increase LHV, cold gasification efficiency, carbon conversion, and gas yield. On the contrary, Na_2CO_3 had a negative effect on the four indices, although Na_2CO_3 had a greater influence on the reduction of tar than did dolomite.

5. The effects of different contents of dolomite and Na_2CO_3 on gasification indices or producer gas compositions showed no significant difference. If economic efficiency is taken into account, 3% (w/w) catalysts in gasification are more suitable than 6% (w/w) catalysts. Overall, dolomite catalyst had a greater influence on the gasification indices than did Na_2CO_3 catalyst.

ACKNOWLEDGMENTS

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