Effects of Metal Salt Catalysts on Fluidized Bed Gasification Characteristics of Source-Collected Combustible Solid Waste

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Effects of metal salt catalysts and gasification temperature on the gasification characteristics of combustible solid waste were studied, based on the source-classified waste in a fluidized bed gasifier, to provide guidance for utilizing the source-classified waste effectively. The results showed that the gasification characteristics of combustible solid waste, such as paper and sawdust, improved noticeably after adding NaCl, K_2CO_3, or sodium dodecyl benzene sulfonate (LAS). Adding NaCl to sawdust increased the yield of CO and CH_4, while the gasification was inhibited gradually with increasing addition. Adding NaCl promoted the generation of H_2 in paper gasification. NaCl played a catalytic role only when it exceeded a certain value. Adding K_2CO_3 increased the yield of H_2 noticeably in sawdust and paper gasification. The catalytic effect of K_2CO_3 on sawdust was better than that on paper. Similar results were obtained for LAS in producing H_2 in gasification. The carbon conversion efficiency and the gasification efficiency were increased with additional LAS. Moreover, the catalytic effect of K_2CO_3 was superior to that of LAS by comparison. As the temperature rises, the activity of the metal salt catalyst is enhanced, but it is inhibited if the temperature is too high.

Keywords: Metal salt; Catalyst; Combustible solid waste; Fluidized bed; Gasification

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INTRODUCTION

As urbanization continues to accelerate, the development of municipal solid waste (MSW) production is also rising (Zhang *et al*. 2015). Various waste disposal technologies have been studied, and some of them are in progress (Okumura *et al*. 2014). Refuse-derived fuel (RDF) is one of a number of latest development trends, which has the advantages of combustion stability, low secondary pollution, ease of transporting and storage, *etc*. (Reza *et al*. 2013; Krüger *et al*. 2014).

Gasification, a thermal conversion process, is applied to solid fuels. It shows higher energy efficiency and wider applicability as compared with incineration technology (Hwang *et al*. 2014; Butterman *et al*. 2014; Materazzi *et al*. 2015). Fluidized bed gasification technology with the characteristics of high efficiency and producing low pollution has become the direction for the current sustainable development research. However, there are many parameters that affect the gasification products, such as gasifying agents and steam to feedstock ratio, or the equivalent ratio of air. Many reports have shown their effects on gasification (Kumar *et al*. 2009; Arena *et al*. 2010; Wang *et al*. 2010; Han *et al*. 2013). For steam gasification, temperature is another important factor (Li *et al*. 2014). In addition, the catalyst is also a significant factor.

Zhou *et al*. (2006) perform air and steam coal partial gasification tests in a fluidized bed gasifier and analyse the effects of the catalysts limestone, sodium carbonate, and dolomite on the coal gasification gas components, calorific value, gas yield, and carbon conversion rate. The results show that adding catalyst can improve the quality of the gas, the gas yield, and the carbon conversion rate. The catalytic effect of sodium carbonate is preferable. The effect of limestone rank is second only to the effect of sodium carbonate, and the effect of dolomite was the worst.

At present, alkali metals and alkaline earth metals are two areas of focus. Research shows that the alkali metals K and alkaline earth metals Ca of catalytic substances can affect the steam gasification of coal char in a fixed bed reactor. The catalytic activity of K is higher than that of Ca. Coal char gasification reactivity increases with an increase of K and Ca additive content, and the load saturation for K and Ca is both 10% (Yang *et al*. 2008). Meanwhile, other research shows that metal salts can promote the pyrolysis of biomass in catalytic pyrolysis experiments with K_2CO_3 and dolomite mixed in various proportions when these mixed catalysts are added into rice straw after pre-treatment (Chen *et al*. 2013). The alkali metal compound in the coal char of solid phase has good flow ability, so it may have a better catalytic activity (Karimi and Gray 2011). Yan and Zhang (2012) reported on the effect of Na_2CO_3 on the specific surface area variation in the catalytic gasification process of anthracite. From the above-mentioned research, it is found that adding dispersed Na_2CO_3 catalyst can improve the carbon active sites and increase the gasification rate, resulting in more microspores opening, cross-linking, and the reduction of micropore surface area. Sugiura *et al*. (2007) study the organic waste (sludge) gasification characteristics in the melt 62-Li_2CO_3/38-K_2CO_3. The findings indicate that the molten salt metal ions have a catalytic effect on combustion and gasification, which can increase the reaction rate.

Until now, most of the studies that have been published about this topic focus on the effect of catalyst additions on biomass and coal gasification. In China, the composition of MSW is very complex, and food waste accounts for a large proportion. Thus, we separate the MSW into dried waste and water-containing waste. Small amounts of sodium chloride in food waste can permeate the solid waste, which may affect the MSW gasification. Meanwhile, potassium carbonate is recognized as the best catalyst with good stability and low cost. To the best of our knowledge, there are few studies on the gasification of a blended catalyst and MSW.

In our study, a comprehensive experimental mechanism has been carried out for combustible solid waste air-steam gasification in a fluidized bed reactor. The main objective of this research is to determine the influence of metal salt catalysts (NaCl and K_2CO_3) on the composition of gasification products and gasification characteristics at various temperatures. The novelty is that we compare effects of organic catalyst and inorganic catalyst on biomass fuels. This research can provide reference for the further efficient use of refuse-derived fuel.

EXPERIMENTAL

Two kinds of typical waste components were selected: sawdust and paper, whose particle sizes are less than 10 mm. Various proportions of sodium chloride (NaCl), potassium carbonate (K_2CO_3), and sodium dodecyl benzene sulfonate (LAS) were added to the two-component materials using the spray method after being dissolved in distilled water. Then, the air-steam gasification process was performed in a fluidized bed. The ultimate and proximate analyses of materials are shown in Table 1. The elemental analyses were conducted by using the elemental analyzer Eurovector EA3000 (Redavalle, Italy).

Table 1. Ultimate and Proximate Analyses of Materials

Sample	Ultimate analysis (%)						Proximate analysis (%)				Lower calorific value (MJ/kg)
	C	H	O	N	S	Cl	M_{ad}	A_{ad}	V_{ad}	FC_{ad}	
Paper	33.55	4.23	40.49	0.13	0.07	0.05	3.04	10.36	75.93	10.69	10.72
Sawdust	43.98	5.35	43.96	0.13	0.03	0.04	4.15	4.39	76.78	14.74	15.49

Experimental Instrument

The main instrument in this experiment was a fluidized bed waste gasification reactor, of which the maximum processing capacity was 20 kg/h. The gasifier's principal part had an inner diameter of 0.108 m, a height of 4 m, and stainless heat resisting furnace tube of austenitic chromium nickel stainless steel, and its maximum temperature was 1200 °C. Bauxite with the average particle size of 60- to 80-mesh was used as a bed material in these experiments. The steam was generated by a steam generator, whose maximum saturation vapor pressure was 0.4 MPa and maximum temperature was 140 °C. According to the characteristics of the experimental materials, the fluidized velocity of the bed material was set to 0.36 m/s and fluidized flow was 5 m^3/h. A schematic diagram of the entire experimental device is shown in Fig. 1.

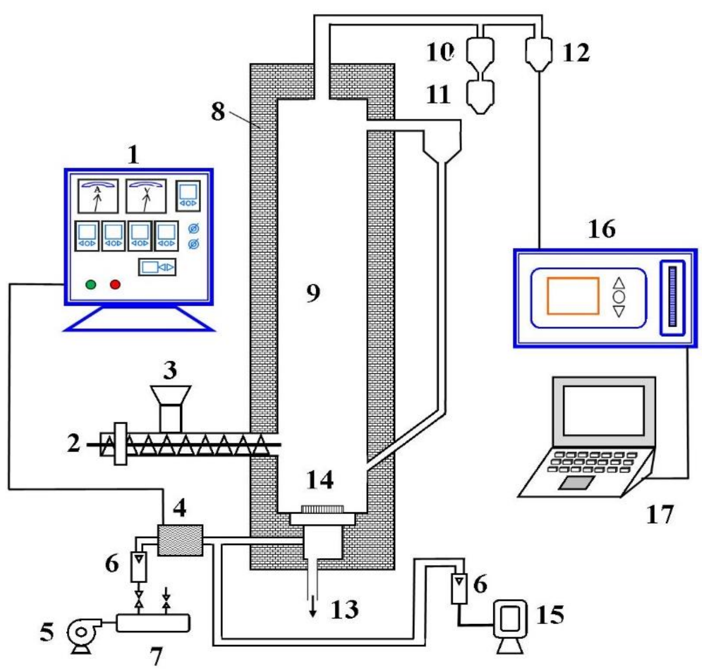

Fig. 1. Schematic of the fluidized bed reactor: 1-Temperature control cabinet; 2-Screw feeder; 3-Silo; 4-Air preheater; 5-Blower; 6-Rotor flow meter; 7-Mixed air filter; 8-Insulation layer; 9-Combustion furnace; 10-Cyclone separator 1; 11-Ash hopper; 12-Cyclone separator 2; 13-Slag discharge tube; 14-Air distribution plate; 15-Steam generator; 16-Online monitoring of flue gas analyzer; and 17-Computer

Experimental Procedure

To begin with, the power and the water valve master switches were turned on, followed by setting the required experimental temperature in the fluidized bed gasification reaction function instrument box, and then gasification experiments were performed at that temperature.

Then, the power switch of steam generator was turned on. And when the gasifier temperature was steady or when the temperature fluctuations were in the range of 15 ºC in the furnace of the measuring point, the spiral feeder power was switched on, and the transport of the reaction materials from the feed inlet was started.

Gasboard-3100-line infrared gas analysis meter made in Wuhan Cubic Optoelectronics Co., Ltd of China (Wuhan, China) was used to monitor the data from which the average data was taken.

RESULTS AND DISCUSSION

Effect of NaCl on Sawdust and Paper Gasification Characteristics

Figures 2 (a), (b), and (c) showed the effect of NaCl on sawdust and paper gasification characteristics when the air equivalence ratio (ER) was 0.4 and the ratio of steam to material (S/B) was 0.8 at gasification temperature of 850 °C. We added 0%, 0.1%, 0.3%, and 0.5% NaCl into sawdust and paper in this experiment. As shown in Fig. 2, the total yield of combustible fuel gas (CO, CH_4, and H_2) generated by sawdust gasification improved after adding NaCl, which reflects the fact that CO and CH_4 contents noticeably increased, while the H_2 content decreased slightly. The total amount of fuel gas was improved noticeably. In this process, the yield of combustible fuel gas increased from 46.48% to 52.30%, mostly because adding NaCl could deepen the secondary cracking of the sawdust lignin to generate tar.

Meanwhile, the easy bond-breaking of carboxyl and carbonyl into small-molecule compounds, as well as hydrocarbon chain reactions, also promoted the generation of coke (Na and Baoxia 2016). These cokes reacted with CO_2 to produce CO according to Eq. 6 and with H_2 to produce CH_4 at high temperatures according to Eq. 10. However, the increase of NaCl inhibited the generation of CO. This phenomenon may be due to the fact that excessive catalyst covers the surface of sawdust to block the aperture, resulting in a reduction of the contact area between sawdust and reaction gas. In this study, NaCl promoted the release of carbon-containing gas noticeably, so the carbon conversion efficiency was noticeably increased, from 73.2% to a maximum of 97.3%. In addition, the gasification efficiency also increased because the yield of fuel gas markedly increased.

For paper, there was not an evident effect on the yield of combustible fuel gas when adding small amounts of NaCl to paper. However, the yield of combustible fuel gas increased when the addition of NaCl reached 5%, which reflects that the yield of H_2 increased from 8.28% to 11.22%. The findings indicated that NaCl addition can play a catalytic role only when it exceeds a certain value. Adding NaCl could promote the reaction of carbon with steam to produce CO_2 and H_2 according to Eq. 4. Because of CO_2 yield increasing, the carbon conversion efficiency also increases. In this process, the yield of H_2 increases obviously; thus, the gasification efficiency increases. Relevant reactions can be listed as follows:

Tar cracking reaction

$$Tar \rightarrow CO_2 + CO + H_2 + CH_4 + C_nH_m \quad (1)$$

Hydrocarbon cracking reaction

$$C_nH_m + 2nH_2O \rightarrow (2n+m/2)\ H_2 + nCO_2 \quad (2)$$

Water gas shift reaction

$$CO + H_2O \rightarrow CO_2 + H_2 \qquad \Delta H = -\ 41.2\ \text{kJ/mol} \quad (3)$$

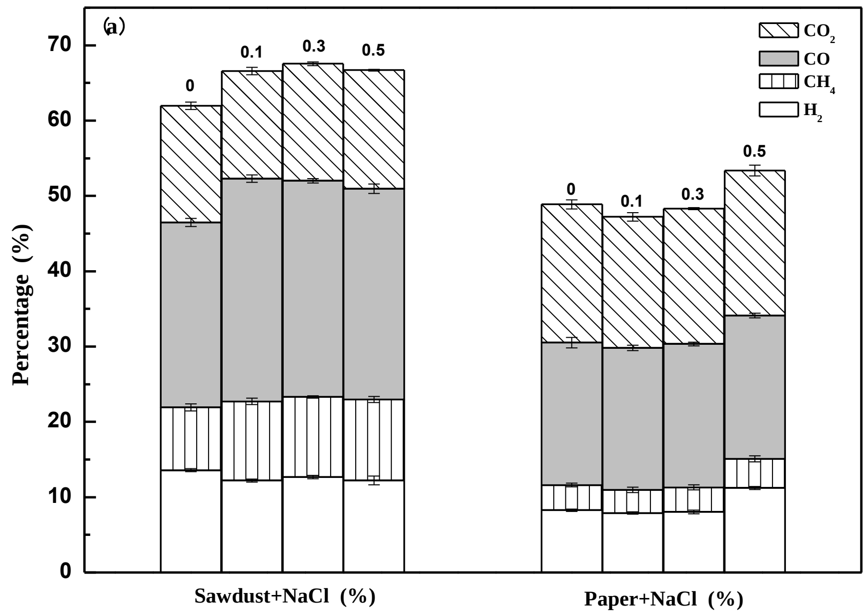

Fig. 2a. Effect of NaCl on various material gasification characteristics; percentages of gases yield of adding various percentages NaCl on sawdust and paper gasification

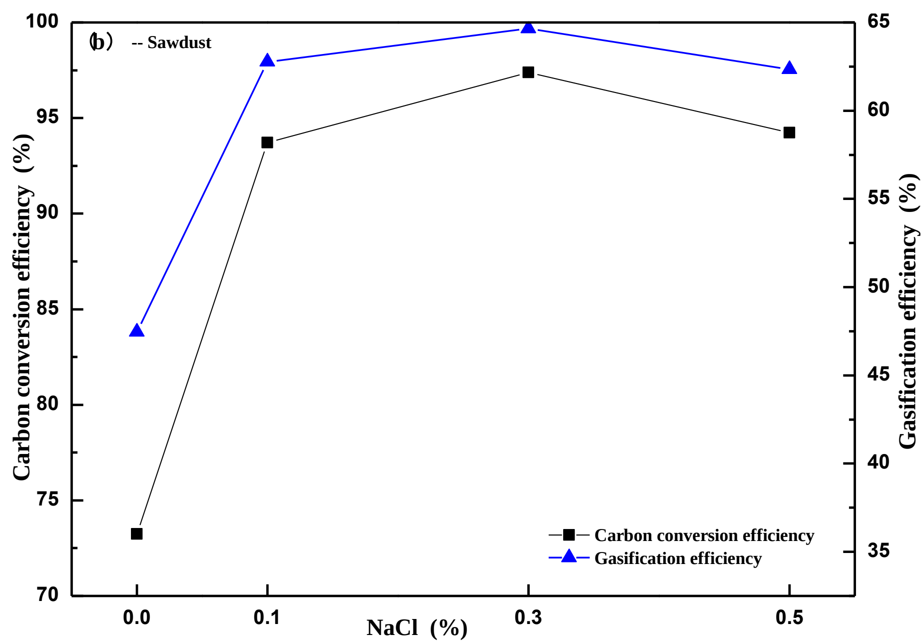

Fig. 2b. Effect of NaCl on various material gasification characteristics; carbon conversion efficiency and gasification efficiency of adding various percentages NaCl on sawdust gasification

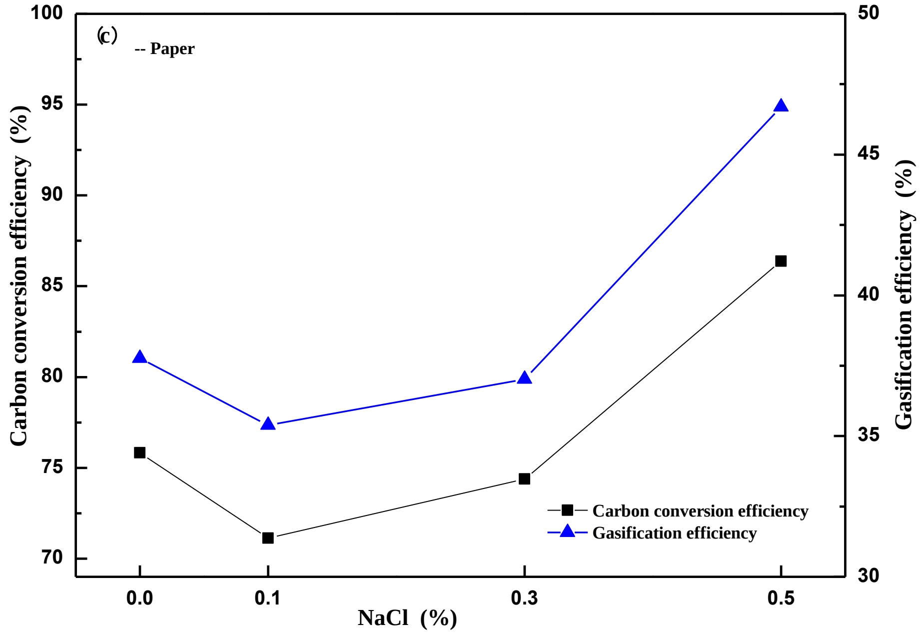

Fig. 2c. Effect of NaCl on various material gasification characteristics; carbon conversion efficiency and gasification efficiency of adding various percentages NaCl on paper gasification

Carbon reaction with steam

$$C + 2H_2O \rightarrow CO_2 + 2H_2 \qquad \Delta H = +\ 90.1\ \text{kJ/mol} \tag{4}$$

$$C + H_2O \rightarrow CO + H_2 \qquad \Delta H = +\ 131.3\ \text{kJ/mol} \tag{5}$$

Carbon reaction with carbon dioxide

$$C + CO_2 \rightarrow 2CO \qquad \Delta H = +\ 172.5\ \text{kJ/mol} \tag{6}$$

Methane reforming reaction with steam and carbon dioxide

$$CH_4 + H_2O \rightarrow CO + 3H_2 \qquad \Delta H = +\ 205.8\ \text{kJ/mol} \tag{7}$$

$$CH_4 + 2H_2O \rightarrow CO_2 + 4H_2 \qquad \Delta H = +\ 164.6\ \text{kJ/mol} \tag{8}$$

$$CH_4 + CO_2 \rightarrow 2CO + H_2 \qquad \Delta H = +\ 247.0\ \text{kJ/mol} \tag{9}$$

Methane decomposition reaction

$$CH_4 \rightarrow 2H_2 + C \qquad \Delta H = +\ 752.400\ \text{kJ/mol} \tag{10}$$

Effect of K_2CO_3 on Sawdust and Paper Gasification Characteristics

Figures 3 (a) and (b) show the effect of K_2CO_3 on sawdust and paper gasification characteristics when the air equivalence ratio (ER) was 0.4 and the steam to material ratio (S/B) was 0.8 at gasification temperature of 850 °C. The addition ratio of K_2CO_3 was 0%, 1%, 3%, and 5%. As can be seen from Fig. 3, adding K_2CO_3 can noticeably improve the yield of H_2 in the combustible fuel gas of sawdust, which increased from 13.58% to 22.00%. Firstly, K_2CO_3 is not in a particle state, but rather almost a molten condition when temperature is greater than 700 °C. Most of the K_2CO_3 can react with the coke, and generate in the intermediate reaction process. And then, the potassium (K) migrates to the surface of sawdust. All these conditions increase the carbon active sites and promote wood lignin pyrolysis to produce tar and oil. The phenols and hydrocarbons generated in gasification further react to produce CO, CO_2, H_2, and CH_4 (Fanmin *et al*. 2013). However, with increasing amounts of K_2CO_3, the reaction rate is accelerated and the water gas shift reaction is promoted toward positive direction according to Eq. 3. Then, the yield of combustible fuel gas increases. This is in agreement with the experimental result of Xiang *et al*. (2009). Adding K_2CO_3 promotes the decomposition of CH_4 and promotes the methane decomposition reaction toward positive direction according to Eq. 10 (Jie *et al*. 2013). Thus, the yields of H_2 and CO_2 increased, and the yield of H_2 increased from 8.28% to 19.20% for paper after adding K_2CO_3. The yields of CO and CH_4, the carbon conversion efficiency, and the gasification efficiency all increased at the beginning. However, with increasing K_2CO_3 addition, these values decreased.

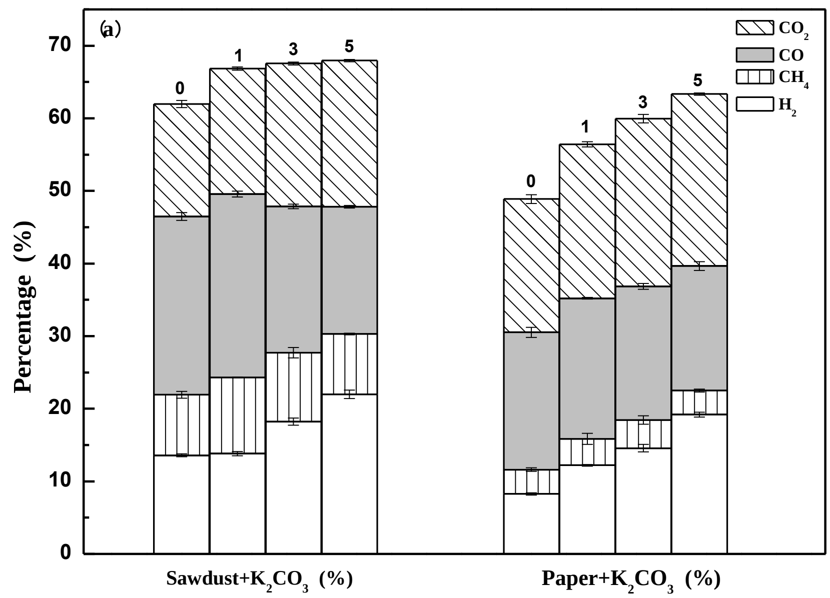

Fig. 3a. Effect of K_2CO_3 on various material characteristics; percentages of gases yield of adding various percentages K_2CO_3 on sawdust and paper gasification

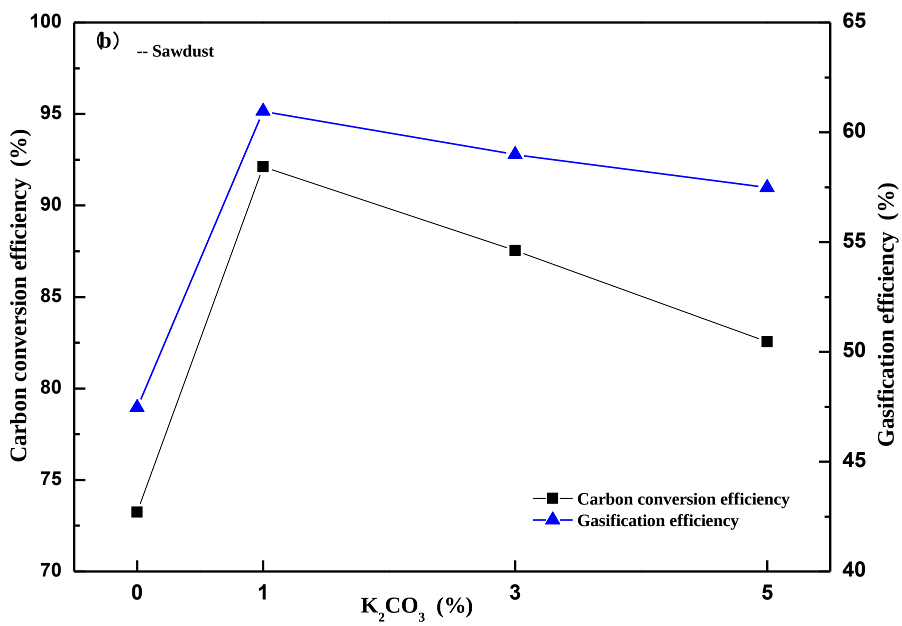

Fig. 3b. Effect of K_2CO_3 on various material characteristics; carbon conversion efficiency and gasification efficiency of adding various percentages K_2CO_3 on sawdust gasification

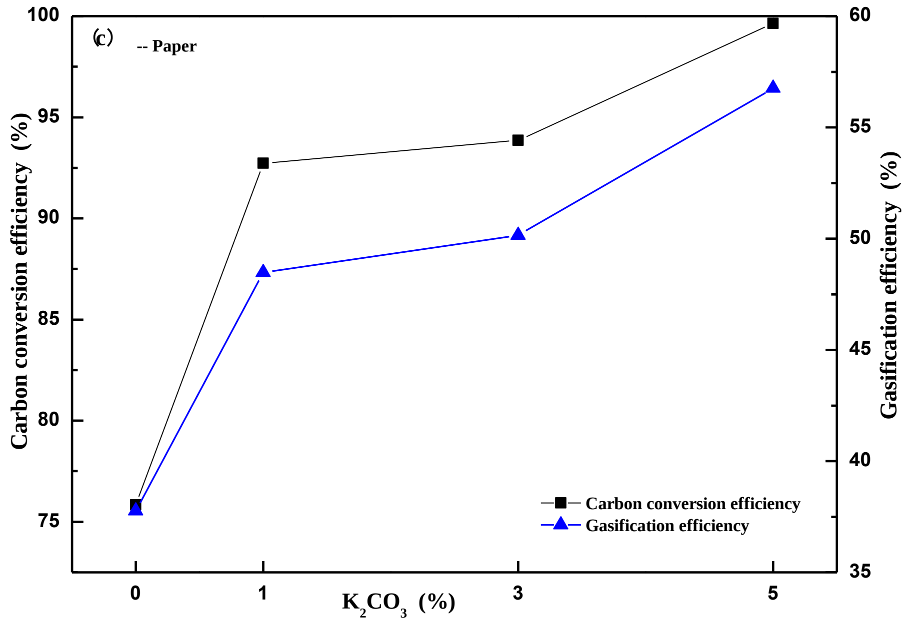

Fig. 3c. Effect of K_2CO_3 on various material characteristics; carbon conversion efficiency and gasification efficiency of adding various percentages K_2CO_3 on paper gasification

Because the main components of paper are cellulose and hemicellulose, adding K_2CO_3 could facilitate the chemical bond length fracture of cellulose and hemicellulose and promote the water gas reaction to produce a large amount of CO_2 and H_2 according to Eq. 3. Thus, the carbon conversion efficiency and gasification efficiency increased noticeably for paper. Meanwhile, the addition of K_2CO_3 for sawdust was found to be better than that for paper, which might be because there is no K_2CO_3 in paper but some potassium in sawdust.

Effect of LAS on Sawdust and Paper Gasification Characteristics

Figures 4 (a), (b), and (c) showed the effect of LAS on sawdust and paper gasification characteristics when the air equivalence ratio (ER) was 0.4 and the steam to material ratio (S/B) was 0.8 at gasification temperature of 850 °C. The addition ratio of sodium dodecyl benzene sulfonate (LAS) was 0%, 1%, 3%, and 5%. LAS is a surface-active agent that can noticeably increase the reaction rate of gasification and reduce the activation energy of the gasification reactor. The addition and dispersion of LAS catalyst can increase the carbon active sites and the gasification rate (Chen *et al*. 2013). As shown in Fig. 4, adding LAS was conducive to the production of H_2 while noticeably inhibiting the production of CO in sawdust gasification. First, adding LAS accelerates the tar cracking reaction according to Eq. 1 and the hydrocarbon cracking reaction according to Eq. 2, subsequently promoting the water gas shift reaction according to Eq. 3. And the methane reforming reaction with steam to generate a large amount of CO_2 and H_2 is shown by Eqs. 7 and 8. With increasing LAS addition, the yield of H_2 increased obviously for sawdust, increasing from 13.58% to 17.14%. The carbon conversion efficiency improved when the yields of carbon gases was increased. The gasification efficiency also increased because the yield of H_2 increased.

For gasification of paper, because paper consists of large hydrocarbons, adding LAS can accelerate the hydrocarbon cracking reaction according to Eq. 2 to generate the products of aromatic structures, and then the volatility of the materials gradually leads to the product of large amounts of H_2. At the same time, the short-chain hydrocarbons further become cleaved into small-molecular weight compounds. The yield of H_2 increased noticeably with the addition of LAS for paper, increasing from 8.28% to 18.00%. In consequence, the catalytic efficiency of LAS was enhanced with increasing LAS addition to paper. The carbon conversion efficiency and the gasification efficiency also increased.

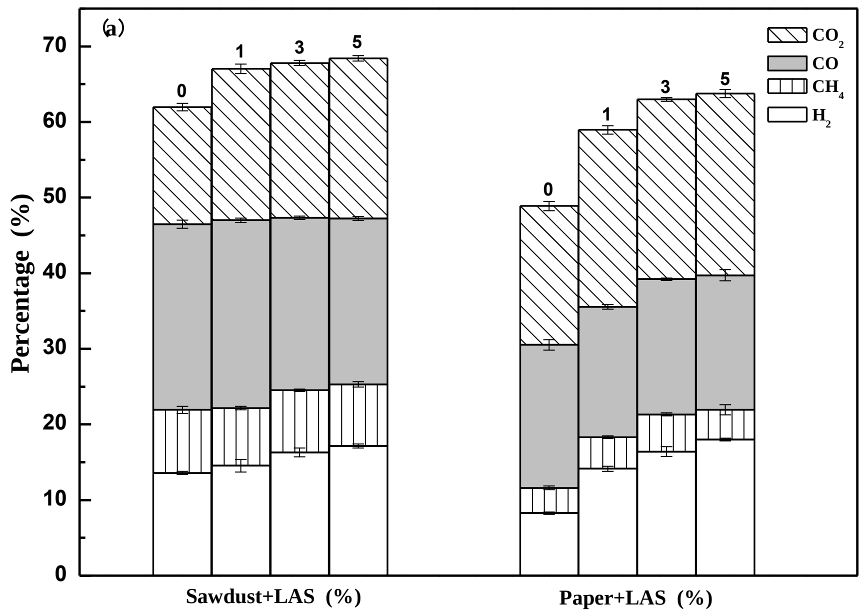

Fig. 4a. Effect of LAS on various material characteristics; percentages of gases yield of adding various percentages LAS on sawdust and paper gasification

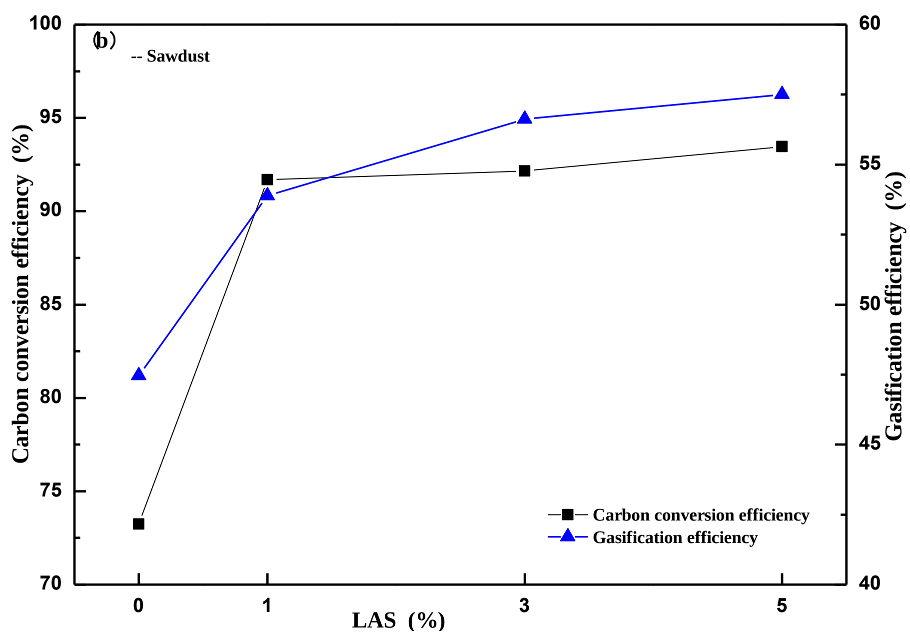

Fig. 4b. Effect of LAS on various material characteristics; carbon conversion efficiency and gasification efficiency of adding various percentages LAS on sawdust gasification

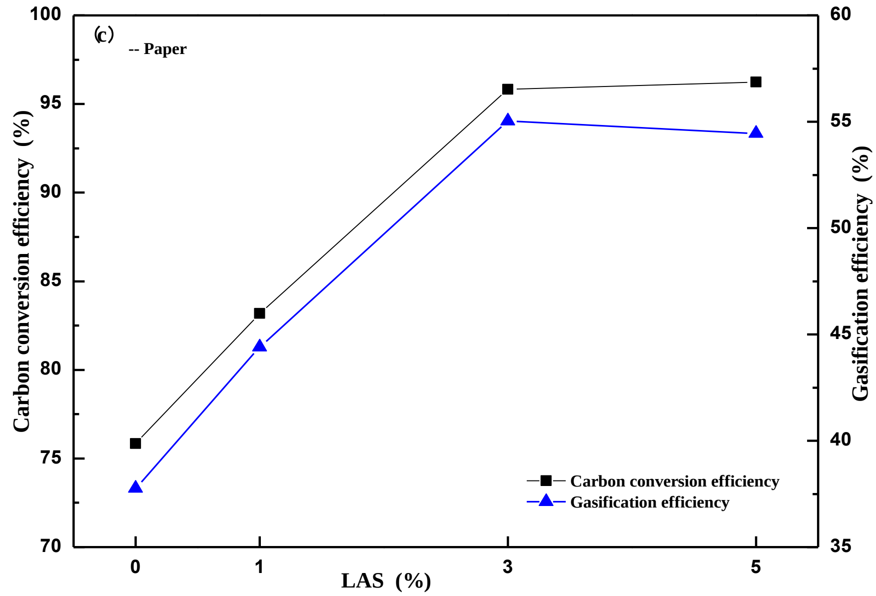

Fig. 4c. Effect of LAS on various material characteristics; carbon conversion efficiency and gasification efficiency of adding various percentages LAS on paper gasification

Catalytic Properties Comparison for K_2CO_3 and LAS

Figures 5 (a) and (b) compare K_2CO_3 and LAS catalytic properties to gasification characteristics when the air equivalence ratio (ER) was 0.4 and the steam to material ratio (S/B) was 0.8 at gasification temperature of 850 °C. The 5% K_2CO_3 and 5% LAS were added to sawdust and paper, respectively. As shown in Fig. 5, the improvement of the gasification gas yield with the addition of K_2CO_3 to sawdust and paper was greater than that of LAS when the catalyst addition amount was identical, as determined by the increase in H_2 yield. For sawdust, adding K_2CO_3 increased the yield of H_2 by 8.42%, while adding LAS increased by 3.56%, making 4.86% percentage points difference compared with K_2CO_3. This may occur because the better flow ability of K_2CO_3 is more conducive to covering the feedstock surface than that of LAS.

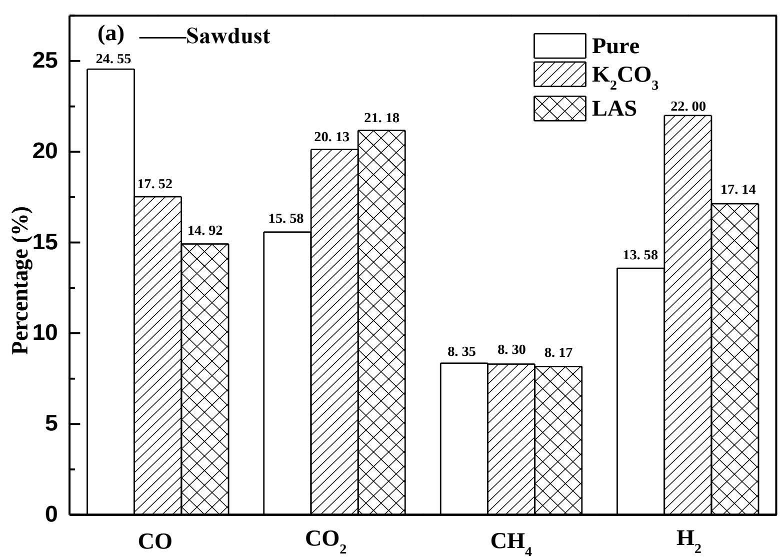

Fig. 5a. Effect of additives on various materials; percentages of gases yield of adding various additives on sawdust gasification

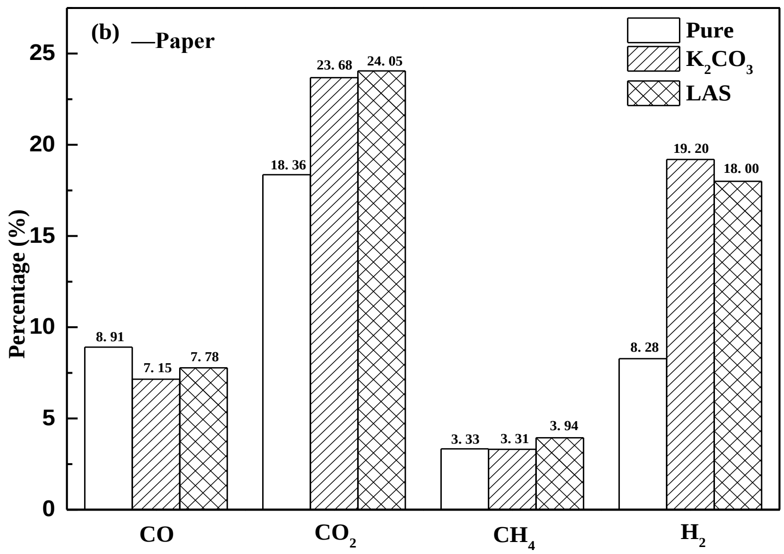

Fig. 5b. Effect of additives on various materials; percentages of gases yield of adding various additives on paper gasification

For paper, the influence of K_2CO_3 and LAS on paper gasification characteristics was the same as sawdust. However, the catalytic effect on paper was not as obvious as it was for sawdust. When K_2CO_3 was added to the paper, the yield of H_2 increased by 1.20% compared with the addition of LAS. On the whole, the catalytic properties of K_2CO_3 are better than those of LAS.

Effect of Temperature on Catalytic Properties of Metal Salt

Figures 6 (a), (b), and (c) show the influence of temperature on the catalytic properties of metal salts when the air equivalence ratio (ER) was 0.4 and the steam to material ratio (S/B) was 0.8 at various temperatures; 0.3% NaCl and 3% K_2CO_3 was added to sawdust, respectively. From Fig. 6, the yield of H_2 increased noticeably, while the yield of CO showed an inverse trend with the increase in temperature. With temperature increasing, NaCl catalytic activity was enhanced and accelerated the water gas shift reaction according to Eq. 3 and the methane decomposition reaction according to Eq. 10 to produce a large sum of CO_2 and H_2, while the yields of CO and CH_4 were evidently inhibited. However, the positive function of CO and CH_4 to combustible fuel gas calorific value was more than the negative function of H_2, so the calorific value of fuel gas declined sharply. At the same time, the carbon conversion efficiency and the gasification efficiency showed a decreasing tendency with fuel gas yield decreasing. As shown in Fig. 6, when the gasification temperature rises, the potassium-based catalyst can show better dispersion in the reactants. The fluidity of K_2CO_3 is enhanced with increasing gasification temperature, and the catalytic activity is also stronger to promote the generation of H_2 from volatile materials.

In the high-temperature phase, adding K_2CO_3 accelerates the secondary cracking reactions of tar and the water-gas reaction and promotes the cracking of macromolecular materials to produce a large amount of H_2. Meanwhile, when the gasification temperature increases, the endothermic reforming reaction of hydrocarbons was enhanced, the methane decomposition reaction was also enhanced according to Eq. 10, and methane reforming reaction with carbon dioxide accelerates according to Eq. 9. Methane cleavage generates C and H_2, which will help improve the yield of H_2 and reduce the contents of

CH_4 and C_2 hydrocarbon gases. As a result, the yield of CH_4 and CO rapidly decreased, while the yield of H_2 showed a clear upward tendency. The carbon conversion efficiency and the gasification efficiency decreased at the beginning. However, with increasing gasification temperature, the carbon conversion efficiency and the gasification efficiency showed a slight increase.

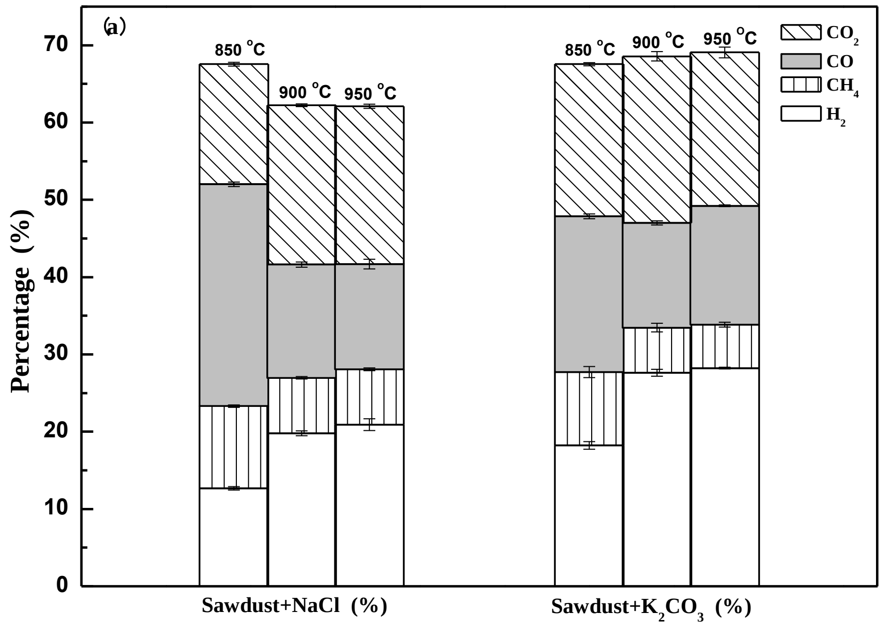

Fig. 6a. Effect of temperature on catalytic properties of metal salts; percentages of gases yield of adding same percentage NaCl and K_2CO_3 on sawdust gasification at different temperature

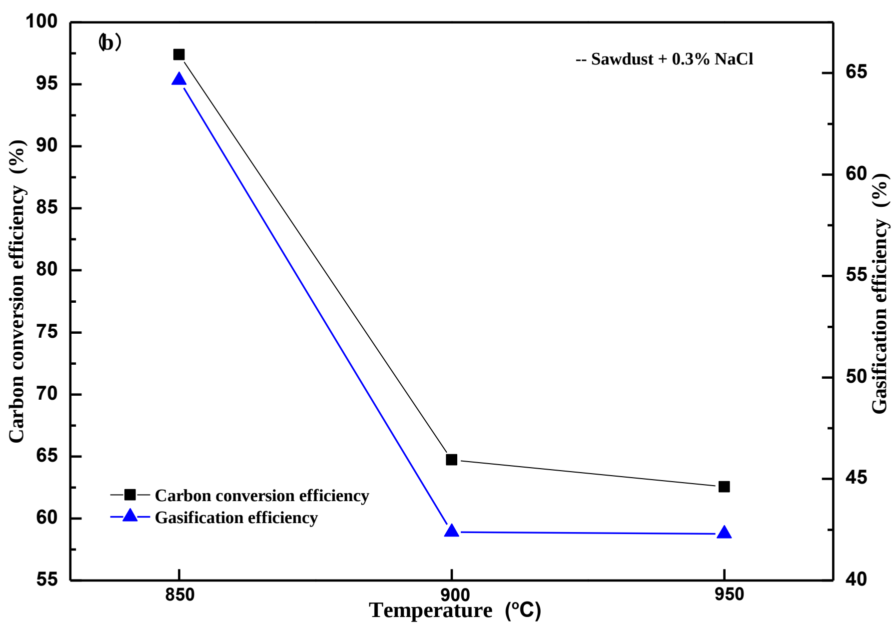

Fig. 6b. Effect of temperature on catalytic properties of metal salts; carbon conversion efficiency and gasification efficiency of adding 0.3% NaCl on sawdust gasification at different temperature

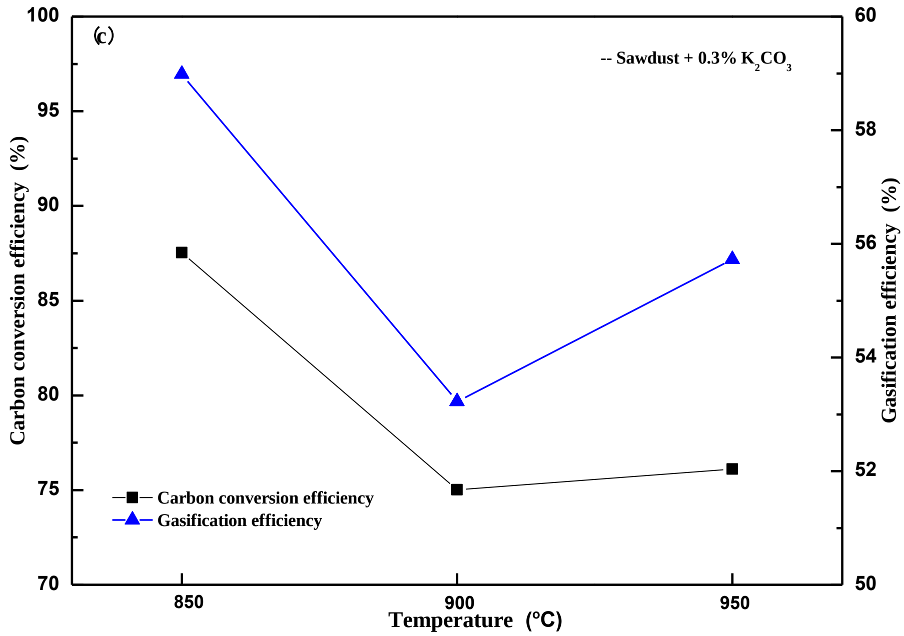

Fig. 6c. Effect of temperature on catalytic properties of metal salts; carbon conversion efficiency and gasification efficiency of adding 0.3% K_2CO_3 on sawdust gasification at different temperature

In the temperature range of 850 to 900 °C, the yield range of each fuel gas generated by sawdust and paper was extremely obvious. However, the yield range of each fuel gas was not obvious in the temperature range of 900 to 950 °C. That is to say, the higher gasification temperature is not the best for waste gasification. As the temperature rises, the metal ions show a better dispersity and catalysis is also enhanced. However, the catalytic activity of metal ions may be inhibited if the temperature is too high.

CONCLUSIONS

1. For sawdust in fluidized bed gasification, adding NaCl can improve the yield of combustible fuel gas CO. The carbon conversion efficiency and the gasification efficiency increased noticeably after adding NaCl to sawdust. For paper in fluidized bed gasification, adding NaCl can raise the yield of the combustible fuel gas H_2, while NaCl can play a catalytic role only when the added amount of NaCl reaches a certain value. The carbon conversion efficiency and the gasification efficiency also increased noticeably when NaCl was added to paper.
2. Adding K_2CO_3 can influence the gasification characteristics of sawdust and paper markedly. Adding K_2CO_3 can promote the cracking reaction of tar and hydrocarbon at first. Subsequently, with increasing K_2CO_3 addition, the water gas shift reaction and the methane decomposition reaction can be promoted to produce a large amount of H_2. Meanwhile the catalytic properties of K_2CO_3 on fluidized bed gasification with paper are greater than they are with sawdust. The carbon conversion efficiency and the gasification efficiency increased at the beginning. Adding K_2CO_3, the carbon conversion efficiency and the gasification efficiency decreased. However, the carbon conversion efficiency and gasification efficiency of paper showed an increasing trend

with increasing K_2CO_3 addition. There were similar results for LAS to produce the combustible fuel gas H_2. When adding LAS to sawdust and paper, the carbon conversion efficiency and the gasification efficiency of sawdust and paper all increased. The catalytic effect of K_2CO_3 is superior to that of LAS.

3. Gasification temperature is one of the most important reaction parameters for waste gasification. With increasing gasification temperature, metal salt catalysis can be enhanced within a certain temperature range. The catalysis properties of metal salt catalyst to gasification were extremely remarkable in the gasification temperature range of 800 to 900 °C. However, when the gasification temperature was in the range of 900 to 950 °C, the catalysis properties of metal salts were reduced slightly in this experiment. With increasing gasification temperature, the yield of H_2 increased and the yield of CH_4 showed an inverse trend.

ACKNOWLEDGEMENTS

This research was funded by the National Basic Research Program of China (973 Program, No. 2011CB201506).

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Article submitted: July 29, 2016; Peer review completed: October 2, 2016; Revised version received and accepted: October 15, 2016; Published: October 21, 2016.
DOI: 10.15376/biores.11.4.10314-10328