



Full Length Article

An advanced biomass gasification technology with integrated catalytic hot gas cleaning. Part III: Effects of inorganic species in char on the reforming of tars from wood and agricultural wastes



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HIGHLIGHTS

- The raw and H-form char were used to reform tar in a pilot scale gasifier.
- The effects of inorganics in the char catalyst on tar reforming were obvious.
- The catalyst also captured volatilised inorganics from raw gasification gas.

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ABSTRACT

Char is used directly as a catalyst for the catalytic reforming of tar during gasification. Experiments have been carried out to examine the effects of inorganics in char as a catalyst for the catalytic reforming of tar during the gasification of mallee wood, corn stalk and wheat straw in a pilot plant. The char catalyst was prepared from the pyrolysis of mallee wood at a fast heating rate. The catalytic activities of char and acid-washed char for tar reforming were compared under otherwise identical gasification conditions. For all biomass feedstocks tested for gasification, the tar contents in product gas could be drastically reduced by the catalyst, reaching a tar concentration level well below 100 mg/N m³. The acid-washed char also showed profound activity for tar reforming although its catalytic activity was definitely lower than the raw char. Both catalysts could effectively reform the aromatic ring systems (especially large aromatic ring systems with three or more fused benzene rings) in tars as is revealed using UV-fluorescence spectroscopy. The char itself was also partially gasified. After being used as a catalyst, the condensation of the aromatic rings and the accumulation of inorganic species led to drastic changes in char reactivity with O₂ at 400 °C. The inorganic species in char tended to enhance the formation of H₂ and CO during the reforming reactions in the catalytic reactor.

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1. Introduction

Biomass, as one of the main renewable energy resources, is abundantly available worldwide, especially in remote areas where electricity grid network may not necessarily cover. The gasification of local biomass feedstock combined with a gas engine could be an economically viable and environmentally friendly option for distributed electricity generation. Due to its high reactivity, biomass will immediately decompose into volatiles and char once it is fed

into a hot reactor. The contact between the highly reactive volatiles and char could considerably inhibit the reaction rate of char and gasifying agents inside a gasifier [1–6]. Furthermore, the volatiles would consume the gasifying agents (e.g. oxygen and steam) at a much higher rate than char, which again is undesirable in terms of char conversion and gasification efficiency. It is therefore highly desirable to minimise the volatile-char interactions and to optimise the volatile-oxygen reactions inside the gasifier, which could be potentially realised by our recently proposed gasification technology [3,4,7].

Tar reduction is a well-recognised roadblock in the commercialisation of advanced biomass gasification technologies. A variety of catalysts such as dolomite, olivine and Ni–Al₂O₃ catalysts have

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been tried for tar removal [8–11]. These catalysts have high activities to reform tar, but they are expensive and easily lose their activities due to the coke deposition. Our studies [12–18] have shown that char and char-supported catalysts could be an ideal candidate to substantially reform the tarry material. Based on these studies, our technology will use char or char-supported catalysts to reform tar [3,7]. The feasibility of tar removal using char or char-supported catalysts has been demonstrated in our pilot plant [3,4].

The active sites in the char catalysts for tar reforming were mainly attributed to the carbon structure as well as the inorganic species in char. Alkali and alkaline earth metallic (AAEM) species can be abundant in biomass and become important catalysts in char for tar reforming. Unfortunately, AAEM species [19–24] undergo drastic transformations during pyrolysis and gasification. Their concentrations and chemical forms would vary significantly with the pyrolysis and gasification conditions under which the char catalyst is prepared. The studies using small amounts (<a few grams) of char [5,12–18] would provide fundamental understanding on the reactions taking place during the catalytic reforming of tar using char catalysts. The inherently-existing and externally-loaded AAEM in char may not only play key roles in tar reduction but also affect compositions of light gases (H_2 , CO , CO_2 and CH_4). Therefore, trials in a pilot plant are essential to answer these fundamentally important questions.

This study aims to investigate the effects of inorganic species on the catalytic reforming of tar in a pilot plant. The char (catalyst) was prepared from the pyrolysis of mallee wood at fast heating rates. The char was also washed with acid to remove inorganic species. Our results indicate that the AAEM-laden char can have higher catalytic reactivities than the corresponding AAEM-lean char. AAEM species also affect the product gas compositions.

2. Experimental

2.1. Biomass samples

Three different biomass samples (mallee wood, wheat straw and corn stalk) were chosen as feedstock for the gasification experiments. Mallee wood and wheat straw were grown in Western Australia. Corn stalk was obtained from Shanxi Province in China. All the biomasses were sized to the range of 0–6 mm and further dried at 105 °C for 10 h in an oven. The dried biomass samples containing 3–5 wt% moisture due to the transfer from oven to biomass hopper were then ready for use. The proximate and ultimate analyses of biomass are listed in Table 1 and the AAEM contents (Na was negligible) in the biomass are shown in Table 2.

2.2. Gasification experiments

A lab-scale gasification pilot plant that has been described in detail in previous publications [3,4] was used for conducting the gasification experiments. All experiments were operated at slightly

Table 1
Property of biomass feedstock.

Biomass	Ash	Volatile	C^b , %	H^b , %	O^c , %	N^b , %	S^b , %
	Yield ^a , %	Matter ^b , %					
Corn stalk	14.2	82.5	49.3	6.0	43.3	1.0	0.4
Wheat straw	6.5	79.8	48.6	6.5	43.2	1.5	0.2
Mallee wood	0.9	81.6	48.2	6.1	45.5	0.2	0.0

^a Dry-basis.

^b Dry-ash-free basis.

^c By difference.

Table 2
AAEM contents (dry basis) of biomass feedstock.

Biomass	K, %	Mg, %	Ca, %
Corn stalk	1.51	0.24	0.33
Wheat straw	1.16	0.06	0.13
Mallee wood	0.06	0.03	0.15

above the atmospheric pressure to maintain the required gas flows. Briefly, 3 pairs of cones as internal structure were purposely built inside the reactor (H1.50 m × Φ 0.44 m) to increase the residence time of biomass particles in the reaction zone as well as to improve the heat transfer to the biomass particles. A catalytic reactor (H0.5 m × Φ 0.16 m) was integrated with the top of the gasifier. The gaseous products including tarry compounds from gasification had to travel through the catalytic reactor where the condensable hydrocarbons would be considerably reformed into light and clean product gases. To ensure identical GHSV when the gas products went through the char catalysts bed, the char catalysts were always over loaded (~1.5 kg) to ensure that the outlet of catalytic reactor was fully covered during the experiments. The outlet of catalytic reactor was located at the side of cylindrical catalyst reactor while the catalysts could be loaded into the reactor from its top. The configuration of the catalytic reactor could be easily found in Part I of this series of study. The temperature distribution inside the gasifier from the top to the bottom has been plotted and shown in Part I of this series of study. The average temperatures in the main gasifier reactor and catalyst chamber are ~880 °C and 800 °C respectively. The ratios of steam to biomass and oxygen to biomass were kept the same as previous studies, namely 0.59 kg/kg and 45 L/kg respectively. The flow rates of O_2 and N_2 were accurately controlled by a mass flow controller and a rotary flow meter respectively. The mixture of O_2 and N_2 entered into the reaction zone from the bottom of gasifier while the steam was supplied from the side bottom of gasifier by injecting a prescribed flow of water through a high precision peristaltic pump.

To determine the tarry materials in gas products, two sampling tubers were installed just before and after the catalysts bed, enabling that a stream of gas could be collected from the hot region (>330 °C) before and after the catalysts bed respectively at 2 L/min for 10 min for each sample. The hot gas would pass through a series of bubblers (impingers) filled with a mixture of chloroform and methanol (4:1 by vol.) which were placed in a dry ice bath (−78 °C) for condensing the tar out of the product gas. After the condensing unit, the permanent gases flew into a rotary flow meter and then went into an on-line gas analyser (ABB). There was a pump integrated inside the gas analyser, which facilitated the control of gas flow rate.

After experiments, the spent catalysts from the reaction zone rather than those above the reactor outlet were collected for further analysis.

2.3. Catalyst preparation

The bio-char catalysts used in this study were prepared from the pyrolysis of mallee wood at fast heating rates in the gasification plant itself. 30 kg of mallee chips (6–10 mm) were fed into the hot reactor at a feeding rate of 20 kg/h at a temperature of 600–950 °C. Due to the specially-designed internal structure, the biomass particles would immediately drop on the hot surface of the first cone and move down in an “S” shape. The contact with the hot stainless steel greatly enhanced the fast heating rate experienced by the biomass particles. 3 L/min nitrogen was continuously supplied from the bottom of the reactor to ensure an inert atmosphere for the biomass decomposition and the growing char (catalysts) bed. At

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