



## Reducing tar yield in gasification of paper-reject sludge by using a hot-gas cleaning system

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### ARTICLE INFO

#### Article history:

Received 4 April 2012

Received in revised form

10 December 2012

Accepted 12 December 2012

Available online 10 January 2013

#### Keywords:

Catalytic gasification

Zeolite

Tar

Synthesis gas

### ABSTRACT

A new tar destruction technology has been developed by integrating in-situ catalytic gasification and a hot-gas cleaning system in the catalytic gasification of paper-reject sludge. Experimental results indicated that the tar yield in syngas decreased significantly from 13.64 to 10.76 g/m<sup>3</sup>, when increasing the zeolite addition during in-situ catalytic gasification. When the syngas passed through the hot-gas cleaning system, the tar concentration measured from downstream was approximately 0.22 g/m<sup>3</sup>. The hot-gas cleaning system used in this research seems to have played a significant role in reducing tar concentration of the syngas. The major tar speciation identified after hot-gas cleaning system was 1-ring or 2-ring hydrocarbons, which included benzene, toluene, xylene, styrene and naphthalene. In summary, the energy yield efficiency of syngas will be enhanced due to the tar reduction and transformation. The syngas seems to be appropriate to use as a fuel in gas engines for electricity generation.

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### 1. Introduction

Biomass gasification is a promising technology that can produce the green, high efficiency, and eco-friendly gas fuels, such as H<sub>2</sub>, CO, CH<sub>4</sub> and other hydrocarbons. However, tar derived from biomass gasification is a critical problem for the synthesis gas application and further energy utilization [1–3]. Generally speaking, the tar derived from biomass gasification may cause to decrease the utilization of combustible gas, increase the problems of operation and maintenance due to the clogging in the pipeline. The tar contains large amounts of hydrocarbon compounds may increase the possibility of carcinogenic risks. Besides, the tar contains the potential energy content that can be transformed to synthesis gas containing relatively high energy value by good gasification practice. Environmental concerns about gasification technology application have become greater due to increasing amounts of tar yield, and consequently, decreasing energy conversion efficiency. Therefore, enhancing the energy conversion efficiency, reducing the tar yield and reducing the carcinogenic risks are important research topics. Recently, there are many techniques that have been used to reduce and/or remove the tar yield, including the self-modification methods and in-situ furnace reduction techniques. These

methods include gasifier design change, temperature control, gasifying agent adjustment, and catalysts addition, etc. [3–18]. Accordingly, the tar removal efficiency can reach between 35% and 90%, respectively. In addition, traditional air pollution control techniques, such as bag filter, wet scrubber, (electrostatic precipitator) ESP, and packed adsorption tower, were also applied widely to solve the tar problems in the synthesis gas applications. The tar removal efficiency could reach significantly 90% and above [3,6,13,19]. When introducing the traditional air pollution control methods, it lost the sensible heat of synthesis gas and produced a large amount of wastewater, resulting in the energy utilization efficiency to decrease and wastewater to be treated. Although above technologies that are effective at among tar cracking, hydrocarbon reforming and tar removal would improve synthesis gas quality, the synthesis gas clean-up system also must still meet the requirements for gas quality on particulates, ammonia, hydrogen sulfides, and other trace pollutants. In this research, to achieve the purposes of enhancing the tar removal efficiency and decreasing the loss of sensible energy in synthesis gas, we developed a new technology integrated with in-situ tar reduction and high-temperature cleanup technique for improving the quality of synthesis gas and enhancing the energy utilization efficiency. Therefore, the main objectives of this research are: (1) to assess the tar elimination efficiency by developed a new hot-gas cleaning system; (2) to understand the tar speciation and transformation in a catalytic gasification.

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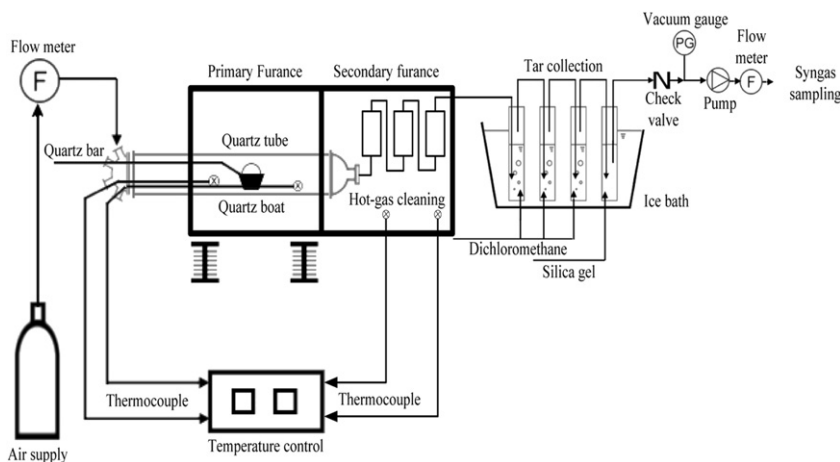


Fig. 1. Schematic diagram of two-stage gasifier.

## 2. Materials and methods

### 2.1. Materials

Representative paper-reject sludge samples were collected from a recycled paper manufacturing plant in Tai-Chung City, Taiwan. The sludge samples were applied as the pelletized (sludge-derived-fuel) SDF manufactured by drying and pelletizing. The dimensions of SDF were approximately 50 mm in height and 20 mm in diameter. To assess the energy yield and tar removal efficiency of tested biomass during in-situ gasification, we chose the natural zeolite to use as a mineral catalyst due to its higher syngas production rate, lower purchasing cost, and high availability. The amount of catalyst addition was 10% and 20% (by weight) compared to the feedstock. On the other hand, the calcined dolomite and activated carbon were also used as adsorbents for applying the hot synthesis gas clean-up system. To maintain the appropriate porosity for providing gas passage, the particle size of tested adsorbents ranged between 0.59 mm and 0.84 mm, respectively.

### 2.2. Experimental apparatus and procedure

The schematic diagram of the experimental apparatus is shown in Fig. 1. The experimental facility used in this study was a two-stage gasifier, which consisted of an electric-heated fixed bed gasifier and a hot-gas cleaning unit. In order to meet all requirements for synthesis gas quality including possible limitations on particulates, alkali, tar, sulfur, chlorine, and other trace components, the hot-gas cleaning system was a multi-packed tower with a series of three columns to capture particles with a high concentration of tar. The columns were filled with zeolite, dolomite and activated carbon, respectively. The hot-gas cleaning system was heated to 250 °C using an electrical heating furnace, to reduce sensible heat loss in the synthesis gas. After the two-stage gasifier, the condensed tar was captured in a series of sampling train of impingers. For each batch, the gasification temperature was set at 900 °C. The gasification air was supplied with an air compressor, which corresponded to 30% of the amount of theoretical air supply. That is, the (equivalence ratio) ER was controlled at 0.30 by introduced into gasifier with an air flow rate 1 l/min. A detailed description of the experiments is presented in our previous paper [20].

### 2.3. Characterization of materials and tars

The moisture content of the paper-reject sludge samples was determined by heating for 24 h at 105 °C. The combustible fraction

and ash content of targeted samples were measured in triplicate using the standard procedure of the ASTM (American Standard Testing Materials). The analysis results indicated that the average moisture content and combustible fractions of the tested biomass sample were  $3.79 \pm 0.49\%$  and  $91.82 \pm 1.68\%$ , respectively. The average (lower heating value) LHV of paper-reject sludge was found to be  $7010 \pm 300$  kcal/kg by a laboratory bomb calorimeter (Parr 1341 calorimeter). The elemental analysis of combustible matter in the paper-reject sludge was analyzed in triplicate by the elemental analyzer (Elementar Vario EIII). The analysis results indicated that the carbon, hydrogen, nitrogen, sulfur, and chlorine content of tested biomass were  $59.17 \pm 4.63\%$ ,  $9.73 \pm 0.76\%$ ,  $0.23 \pm 0.10\%$ ,  $0.23 \pm 0.11\%$ , and  $0.34 \pm 0.01\%$ , respectively. The bulk density, specific surface area, pore volume, and chemical composition of the tested catalysts and/or adsorbents were also determined by surface area analyzer (Micromeritics, ASAP 2020) and inductive coupled plasma optical emission spectroscopy (ICP-OES, Perkin Elmer, Optima 2000DV). Acidity of zeolite was determined by volumetric titration [21]. The amounts of zeolite sample were 0.5 g which is prepared at 120 °C for 6 h and then was taken in a conical flask to which 15 ml of 0.1 N NaOH was added. After stirring the flask for 10 min, the zeolite was titrated with 0.1 N H<sub>2</sub>SO<sub>4</sub> with phenolphthalein indicator. The analysis results of characteristics of tested catalysts were indicated in Table 1. The surface area and acidity of zeolite were approximately 99.65 m<sup>2</sup>/g and 163 meq/100 g, respectively. To compare with results of other literature, it can act as a tested catalyst in gasification of paper-reject sludge. Tar produced in this research is commonly

Table 1

The properties of the catalysts and adsorbents.

	Zeolite	Calcined dolomite	Activated carbon
Density (g/cm <sup>3</sup> )	2.26 ± 0.01	2.59 ± 0.08	1.87 ± 0.02
Surface area (m <sup>2</sup> /g)	99.65	13.03	1011
Pore volume (cm <sup>3</sup> /g)	0.07689	0.04167	0.5012
Pore diameter (Å)	30.86	127.9	19.83
Particle size (mm)	0.59–0.84	0.59–0.84	0.59–0.84
Acidity (meq/100 g)	163	N.A.	N.A.
Chemical composition (%)			
K <sub>2</sub> O	1.36 ± 0.07	0.06 ± 0.01	N.A.
Na <sub>2</sub> O	1.35 ± 0.04	0.07 ± 0.01	N.A.
SiO <sub>2</sub>	86.14 ± 9.93	17.30 ± 2.95	N.A.
Al <sub>2</sub> O <sub>3</sub>	8.35 ± 0.24	0.07 ± 0.00	N.A.
Fe <sub>2</sub> O <sub>3</sub>	0.97 ± 0.14	0.10 ± 0.00	N.A.
MnO	0.01 ± 0.00	0.00 ± 0.00	N.A.
MgO	0.11 ± 0.00	28.87 ± 0.39	N.A.
CaO	1.64 ± 0.06	53.46 ± 0.62	N.A.

N.A.: not available.

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