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Co-gasification of municipal solid waste with high alkali coal char in a three-stage gasifier

Binhang Hu[a], Qunxing Huang[a,*], Alfons Buekens[a,b], Yong Chi[a], Jianhua Yan[a]

[a] State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou 310027, People's Republic of China
[b] Chemical Engineering Department, Vrije Universiteit Brussel, Brussels, Belgium

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ABSTRACT

The commercial scale up of municipal solid waste (MSW) gasification system is restricted by the low quality of the syngas, especially due to the low heating value and high contents of tar and HCl. In this study, an innovative three-stage system for co-gasification of MSW with high alkali coal char was developed. The modeled MSW was pyrolyzed in the first stage and the raw syngas was partially oxidized in the second stage and then reduced with high alkali coal char in the final stage to produce high quality syngas. The effects of temperatures and equivalence ratios (ER) have been evaluated experimentally and the concentration of condensable tar species and HCl was examined carefully. In general, the two key pollutants in produced gas could be controlled as low as 11.3 mg/Nm^3 (tar content) and 17.6 mg/Nm^3 (HCl content). Meanwhile, the level of H_2, CO, CH_4 in synthesis gas reach a stable high level of 41.9 vol%, 29.3 vol% and 7.49 vol%, respectively, while the lower heating value (LHV) attains 12.2 MJ/Nm^3, meeting the intake-gas conditions for internal combustion engines. The experimental results confirm that the highest pyrolysis temperature leads to the maximum gas yield from oxidation stage (i.e., 0.913 Nm^3/kg at 650 °C), to be compared with 0.898 Nm^3/kg (550 °C) and 0.747 Nm^3/kg (450 °C). The lowest gasification temperature (800 °C) is indicated as most favorable for HCl removal from syngas, linked with the advancement of reversible reactions between HCl and Ca-based compounds. H_2, tar and LHV all decrease with rising equivalence ratio. In summary, the high-quality syngas can be produced at a steady yield rate of 1.57 Nm^3/kg from three-stage gasifier, due to dichlorination and catalytic tar cracking action of high alkali coal char at a low cost.

1. Introduction

The amounts of municipal solid wastes (MSW) are endlessly increasing with economy growth, raising the sustainable management solutions issues [1–10]. Gasification is another thermochemical process involving a partial oxidation of organic compounds between 500 and 1800 °C to realize high-quality and clean utilization of MSW, producing a syngas which can be used as a fuel for efficiently yielding heat and power, or as a feedstock for the production of organic chemicals and ammonia after some reforming reactions [11–13]. Various studies have also pointed out the gasification possesses better performance in higher electrical and overall energy efficiency, lower emissions and lower investment costs than direct combustion [14–19].

As a consequence, lots of studies have focused on the performance of MSW gasification, as well as co-gasification process. A compact design of "UNIQUE" gasification concept integrating gasification, gas cleaning and conditioning in one single reactor unit was developed after collaborative R & D efforts [20]. The activity of catalysts and sorbents sited in the gasifier is improved through the integral layout, while keeping the thermal efficiency high [21]. A specially designed three-stage gasification process of "FLETGAS" was proposed by Nilsson et al. [22], which was consist of a first, fluidized bed devolatilization (700–750 °C), a subsequent steam reforming of fresh tar at 1200 °C and a final moving bed downdraft gasifier [23]. Nevertheless, the actual industrial implementation of gasification processes of MSW faces several challenges when brought to the market. In particular, the gas produced contains a high level of condensable organic compounds (usually referred to as tars) which cause blockage and corrosion in gasifiers and also reduces overall efficiency [24–27]. Secondly, MSW typically contains significant amounts of chlorine, mainly associated with the food waste stream and plastic waste, forming acid gas (HCl) during gasification and causing severe corrosion of down-stream equipment, as well as poisoning the catalysts used for tar cracking [28–30].

These two obstacles result in a lower calorific value of syngas, with impurities that impose restrictions on a wider market penetration of

* Corresponding author.
E-mail address: hqx@zju.edu.cn (Q. Huang).

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commercial advanced gasification technologies. The inferior coal at a low-cost contains significant amounts of alkali metals which are important catalysts for tar cracking, and it can be co-gasified with MSW to improve the syngas quality. Co-gasification of waste with coal is very promising method to convert the solid waste into syngas which can be burned or used as chemical feedstock. It has received intensive interests in the past few years. Pinto et al. [31] tested the co-gasification of coal mixed with waste and compared tar cracking catalysts. Five mixtures of coals, plastics and wood have been pelletized and fed into a bubbling fluidized bed gasifier by Zaccariello et al. [32] to compare the overall performance efficiency of the co-gasification process. Filomena et al. [33] performed the co-gasification experiment of different grade coals mixed with different types of biomass waste. Whereas, some problems exist in these direct blending co-gasification as: (1) the waste must be dried and shredded to achieve well blending and the syngas contains high concentration of particulate matters; (2) the syngas contains high content of tar due to the lack of reduction stage; (3) the HCl problem during the gasification of waste with high chlorine content is ignored [31–37]. In this study, an innovate three-stage system was developed for co-gasification of MSW and coal char. The innovation points lie in the usage of high alkali coal char as catalyst for tar cracking, absorbent for HCl removal and gasification reactant, and the two feedstocks are separated into two reactors in the integrated gasification system. The findings of this paper are expected to provide a deeper understanding on the characteristic of the multi-stage co-gasification process and suggest a feasible gasification way for the disposal of municipal solid waste with high chlorine content, which can be driven to the market.

2. Materials and experimental methods

2.1. Materials

The major raw materials adopted were wood chips from Shanghai Yourui Wood Industry Co., Ltd, simulating a first key component (wood waste) of MSW [1]. The wood material was sieved to a size between 0.1 and 0.84 mm (average: 0.4 mm) [38]. The chlorine content of the feed was less than 0.01% by weight; no polymer resin or glue was present. Polyvinyl chloride (PVC) powder was purchased from Sigma-Aldrich Co. (USA) and used as an additive, simulating a second key component (plastic waste) of MSW [1], the particle size of which was less than 0.125 mm. A mixture of wood chips and PVC powder (chlorine content 1%) was employed as simulated waste feedstock. The proximate and ultimate analysis of raw materials was shown in Table. 1. The “Zhundong” coal char added during the third stage serves as reactant and catalyst for tar cracking. It derives from the giant Eastern Junggar coalfield (Sinkiang Uygur Autonomous Region, China) and is rich in both alkali and alkaline-earth metals. Table. 2 shows its ash composition; the Na_2O and K_2O content attains 4.7% and 0.5%, respectively, and the CaO and MgO content 35.1% and 12.5%, respectively.

2.2. Experimental set-up

A schematic representation of the three-stage laboratory-scale co-gasification of MSW and coal char apparatus is shown in Fig. 1. The apparatus mainly consists of a reverse L tubular reactor made of steel (length: 1150 mm, height: 650 mm, outer diameter: 30 mm, inner diameter: 20 mm). The reactor is subdivided into three stages, signifying pyrolysis, oxidation and reduction. The three process stages are heated by three electric furnaces (SKF-2-13 Hangzhou blue sky Instrument Co., ltd, China) and temperature is controlled for pyrolysis at 450, 550, and 650 °C, for oxidation at 800 °C, and for reduction at 800, 900, and 1000 °C.

The first treatment stage is a pyrolysis process [16]. Simulated MSW feedstocks are delivered at 1.33 g/min by a screw feeder compactly installed in a steel tube to seal the entrance. Initial trials demonstrated that the 1.33 g/min of the screw feeder could reach the complete reactions. The second stage is oxidation of pyrolysis tar and gas with a mix of nitrogen and oxygen. Two mass flowmeters (D08-1F, Beijing seven-star Huachuang Electronics Co., Ltd, China) are used to set the gas flow of N_2 and O_2, at three different equivalence ratios (ER), to define an appropriate amount of oxidizing agent. The third stage is an up-draft fixed bed gasifier with coal char of high alkali contents placed above an air distributor using oxidation gas from the burning chamber as a reacting agent. A stack of 250 mm high alkali coal char provides enough residence time (about 3 s) to complete the Boudouard, water-gas reaction and methanation reaction [39]. Since the last chemical reaction is strongly endothermic and the upward flue gas could transfer a portion of heat to downward feed in up-draft gasification furnace, the synthetic gas cools down to below 300 °C. Therefore, this “chemical quenching” enhances the calorific value.

Gas is sampled at the entrance and exit of this third stage, with two flowmeters to measure the gas yield rates, respectively. The gas bubbles through two 50 ml scrubbing bottles containing 30 ml of 0.1 mol/L sodium hydroxide solutions for capturing HCl gas before and after the third stage in the flue gas, respectively. The chloride ion concentration in the mixed solutions, which accounts for the HCl gas concentration can be directly detected through high-pressure ion chromatography (Integrion Thermo scientific, USA) with maximum operating pressure up to 6000 psi. When detecting the tar content in the gas, the sodium hydroxide solution in the scrubbing bottles is replaced by aqueous solutions and the tar in the tar trap is collected by washing with about 50 mL of dichloromethane for three times. The mixture of water and organic solvent (mixture of tar and dichloromethane) was separated by a tap funnel. Then, the tar was separated from the mixed organic solvent by a rotary evaporator [40]. Eventually, the gas lines are connected to gas sampling bags and gas is collected for 6 min for every bag. The entire experimental procedure lasts for 60 min. Thus, 20 gas sampling bags (2 L for each one) in each experiment are analyzed by gas chromatography and the key components of the produced gas (H_2, CO, CH_4, C_2H_4, C_2H_2, C_2H_6, CO_2, O_2) are analyzed using an Agilent 490 micro GC (type of chromatographic columns, MS5A 10 m BF and PPU 10 m BF; and temperature of the columns, 80 °C, Agilent Technologies Inc., USA). After the co-gasification experiments, the coal char after reaction in Run 2 was analyzed with an Empyrean 200895 X-ray diffractometer system with Cu Kα as radiation source (λ = 1.5406 Å) made by Panalytical, Netherlands to characterize the crystalline phases. All detail experimental conditions were displayed in Table. 3. Each run was performed for three times, ensuring the experimental repeatability.

3. Results and discussion

The experimental results featuring three-stage co-gasification

Table 1
Proximate and ultimate analysis of raw materials.

Sample	Proximate analysis/%					Ultimate analysis/%					$Q_{net,ad}$ (kJ/kg)
	M_{ad}	A_{ad}	V_{ad}	FC_{ad}	C_{ad}	H_{ad}	N_{ad}	$S_{t,ad}$	O_{ad}	Cl_{ad}	
Wood	11.9 ± 0.3	9.2 ± 0.3	59.8 ± 1.2	19.1 ± 0.5	48.0 ± 0.6	4.9 ± 0.1	0.2 ± 0.0	0.9 ± 0.1	24.9 ± 0.4	0.0 ± 0.0	15980.2 ± 33.6
PVC	1.4 ± 0.0	0.0 ± 0.0	93.6 ± 2.1	5.0 ± 0.1	39.1 ± 0.3	5.0 ± 0.1	0.4 ± 0.0	1.2 ± 0.1	0.0 ± 0.0	52.9 ± 0.2	20710.3 ± 42.7
Coal char	0.9 ± 0.0	8.9 ± 1.7	6.9 ± 0.2	83.3 ± 1.1	78.2 ± 1.5	1.1 ± 0.1	1.1 ± 0.0	0.4 ± 0.0	9.4 ± 0.6	0.0 ± 0.0	31210.9 ± 53.1