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Steam/oxygen gasification of dried sewage sludge in a two-stage gasifier: Effects of the steam to fuel ratio and ash of the activated carbon on the production of hydrogen and tar removal

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ABSTRACT

Steam/oxygen gasification of dried sewage sludge was performed in a two-stage gasifier to produce an H_2-rich and tar-free syngas. The experiment mainly investigated the effects of activated carbon, ash of activated carbon, steam to fuel ratio and the combination of additives on syngas quality. In the results, all the syngases obtained with activated carbon did not contain any tar. Activated carbon increased the H_2 production and decreased the NH_3 content in syngas. Acid-treated activated carbon, which has less ash content than the original activated carbon, was less active in tar cracking and H_2 production. The steam to fuel ratio had a strong influence on syngas quality, causing a significant rise in the H_2 and NH_3 contents in syngas at a high steam to fuel ratio. The extra addition of CaO and activated carbon to the base additive (activated carbon) led to increased H_2 production and active tar cracking. The maximum H_2 content in syngas (52.2 vol%) was obtained with 2.5 kg of activated carbon at a steam to fuel ratio of 0.52. The minimum NH_3 content in syngas was 20 ppm.

1. Introduction

Gasification, one of the thermo-chemical conversion processes, is defined as the thermal decomposition of a material in the presence of limited oxygen to produce a gas called syngas or producer gas. Syngas (or producer gas) can be used as a fuel for heat and electricity generation, or as a feedstock for chemical and fuel synthesis. It is also considered a promising route to produce H_2 that is an alternative energy source to fossil fuels. During the past few decades, many studies on the production of H_2 from gasification have been conducted [1–5]. In particular, biomass gasification is regarded as an attractive route for H_2 production, because biomass-derived H_2 contributes to renewable energy production while leaving a minimal environmental footprint [6]. Sewage sludge is a biomass resource that can be steadily supplied from waste water treatment plants. Not only does the gasification of sewage sludge generate energy but it also reduces the sludge volume and generation of toxic compounds [7]. The gasification of biomass including sewage sludge can be conducted using different gasifying agents. Among them, steam has proved to be the most effective and efficient agent for H_2 production [8–10]. However, steam biomass gasification for H_2 production still cannot cross the threshold of full commercialization. There seem to be two reasons for the delay of its development: 1) it is difficult to find a proper route for providing heat for the endothermic steam gasification reactions without any deterioration of syngas quality and 2) biomass gasification is essentially associated with the tar problem, which can prevent the operation of a full gasification system and cause problems in the process equipment, such as gas engines and turbines. Meanwhile, the use of a mixture of steam/oxygen as a gasifying agent has been a representative way to produce H_2 from various feedstocks. Oxygen in the mixture plays a significant role, providing heat by burning fuels for endothermic reactions associated with steam. In steam/oxygen gasification, the STF (steam to fuel ratio), which is usually defined as the ratio of the total mass (or mole) of steam to the total mass (or mole) of fuel, is a crucial operation parameter. Many papers have announced the significance of the STF on the syngas properties: 1) Turn et al. performed the gasification of sawdust from lumber in the STF range of 1.1–4.7 [11], 2) Chang et al. studied the effect of steam to α-cellulose ratio at 800 °C [12], and 3) Shen et al. investigated the gas composition trends under the STF (0.4–1.0) [13]. Their research has shown that the increase in the STF

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favored H_2 production to an extent. However, a rise in the STF also had negative side-effects, giving rise to a low heating value of syngas and the need for extra equipment for water separation. Meanwhile, steam/oxygen gasification cannot be free from the tar problem. One representative approach to overcome the tar problem in biomass gasification is the use of tar-cracking additives. Elena et al. investigated the effect of dolomite on tar removal and concluded that dolomite not only led to the decrease of tar but also to the increase of H_2 [14]. Corella et al. also observed a decrease of total tar with dolomite as the bed material [15]. Rapagna et al. also reported the positive effect of dolomite on the syngas yield [16]. CaO has also been widely tested in biomass gasification both for CO_2 absorption and tar cracking. Liu et al. showed that CaO played an important role as an additive and conditioner in steam gasification to enhance syngas production [17]. Udomsirichakorn et al. reported the importance of the reproduction of CaO from $CaCO_3$ after capturing CO_2 [18]. Meanwhile, Fukuyama et al. proved that activated carbon had the ability to absorb tar in syngas [19]. Studies conducted using a two-stage gasifier (the University of Seoul (UOS) gasifier) composed of a fluidized bed gasifier and fixed bed reactor confirmed the tar removal ability of activated carbon [20–23].

There have been some studies on steam/oxygen (or air) gasification of DSS (dried sewage sludge) which reported the influence of operating conditions on the syngas quality [14,24]. In contrast to those studies, the present study reports experimental results on the steam/oxygen gasification of DSS using a two stage gasifier (the UOS gasifier) mainly with activated carbon for the first time. Till now, our group has been conducting a series of research on the gasification of DSS (dried sewage sludge) using the UOS gasifier and air as a gasifying agent [21,23,25]. The present work differs from earlier ones in that it aims to produce an H_2-rich and tar-free syngas from the steam/oxygen gasification DSS. This research provides the effects of the STF in combination with additives on H_2 production and tar removal. In addition, the effects of the amount of activated carbon and its ash content on syngas quality are examined. Finally, the present study provides the effects of such operational variables on the NH_3 content of syngas.

2. Material and methods

2.1. Feed material and additives

DSS obtained after anaerobic digestion and thermal drying was supplied from a sewage disposal plant in Seoul. As pretreatment for feeding, it was first crushed and sieved to obtain a material with a diameter of 0.6–3.35 mm. The fraction obtained after sieving was sufficiently mixed to obtain a homogeneous distribution of composition and size. Table 1 shows results of the proximate, ultimate, and caloric value analyses of the DSS.

Table 1
Properties of dried sewage sludge.

Proximate analysis[a] (wt%)		Ultimate analysis[b] (wt%)	
Moisture	5.29 ± 0.1	Carbon	40.31 ± 0.2
Volatile matter	61.56 ± 0.5	Hydrogen	5.29 ± 0.08
Fixed carbon	7.01 ± 0.3	Nitrogen	5.73 ± 0.02
Ash	26.14 ± 0.1	Oxygen[c]	21.21
		Sulfur	1.32 ± 0.1
Lower heating value (MJ/kg)		17.1	

[a] ASTM E871-82, ASTM E872-82M, ASTM D1102-84.
[b] Dry and ash free basis.
[c] Calculated by difference.

The feed material was mainly composed of volatile matter (61.6 wt%) and ash (26.1 wt%). It had a nitrogen content of 5.7 wt%. The above results indicated a high potential for the tar and NH_3 generation during gasification.

In the experiment, natural olivine (SiO_2, Fe_2O_3 and MgO), the size of which was 150–300 μm, was used as the fluidized bed material. Pellet-type coal-based AC (activated carbon) with a size range of 3.35–10 mm and a surface area of 959 m^2/g was applied as a base tar-cracking additive. In the study, pretreatment of AC with an acidic solution was also performed to reduce its ash content. The procedure was as followings: At first, the original AC was sufficiently soaked in hydrochloric acid solution (1.0 N), followed by 5 h stirring at room temperature, and then the wet AC was washed by distilled water until the color change was not observed on the litmus paper. Finally, the acid-treated AC was dried. A gravel-typed CaO with a diameter of 30–40 mm was used as an additional tar-cracking additive. Before putting in the gasifier, it was calcined at 900 °C for 10 h in an electric laboratory furnace.

2.2. Gasification process and experimental procedure

A diagram of the gasification process used for the present study is shown in Fig. 1. Details of the process and experimental procedure have been published elsewhere [21,22].

The key part of the process is the two-stage gasifier composed of a bubbling fluidized bed (lower reactor) and a fixed bed reactor (upper reactor) in series, which are divided by a distributor with holes measuring 1 mm in diameter. Functionally, the two-stage gasifier removes tar in stages: First, tar produced in the lower reactor can be removed by the catalytic action of additives used as the fluidized bed material, which is natural olivine in this study. The gas escaping from the lower reactor still has a considerable amount of tar, most of which can then be destructed by additives like activated carbon and CaO in the upper reactor. The syngas leaving the two-stage gasifier could contain some amount of tar, which would be condensed on the tube, condenser and EP (electrostatic precipitator) of the process. The amount of condensed tar is defined as “total tar” in this study.

For steam/O_2 gasification, steam was generated in this study via a steam generator using distilled water and supplied through a pipeline connected with the lower reactor. Oxygen was separately supplied via a preheater. Temperatures of preheated steam and oxygen were about 450 °C.

2.3. Reaction conditions

The main experimental variables were the STF, the amount and kind of tar-cracking additive and the amount of ash in activated carbon. STF in this work was defined as the ratio of the sum of water in DSS and water for fluidization (kg) to the mass (kg) of fuel on a dry and ash free basis. ERs (equivalence ratios) were kept constant during gasification at about 0.25. The ER was defined as the actual oxygen/sludge ratio divided by the stoichiometric oxygen/sludge ratio. The flow rates of preheated oxygen were ~7 Nl/min. Temperatures of the lower and upper reactors were almost constant around 800 °C. The feed amount in each run was 1 kg, with the feeding time slightly exceeding 30 min. A further longer operation could not be performed because of ash accumulation in the fluidized bed reactor with increasing operation time. The resultant feed rates were 29–31 g/min. The amounts of natural olivine used as the fluidizing bed material and AC used in the upper reactor were 2.5 kg and 1.5–2.5 kg, respectively. The amount of CaO added to activated carbon was 1 kg. When CaO was added, it was first put on the distributor plate located between two reactors, with activated