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Fuel

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Full Length Article

Gasification of dried sewage sludge using an innovative three-stage gasifier: Clean and H₂-rich gas production using condensers as the only secondary tar removal apparatus

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ARTICLE INFO	A B S T R A C T
<i>Keywords</i> : Dried sewage sludge Gasification Tar Producer gas Active carbon	Air gasification of dried sewage sludge was performed in a new-type three-stage gasifier consisting of an auger, a fluidized bed, and a tar-cracking reactors. The study aimed to evaluate the possibility of gasification using only condensers for the secondary tar removal method. The effects of Ni- and Fe-impregnated active carbons on the quality of producer gas were also investigated. Finally, to investigate the deactivation behavior of active carbon, a spent active carbon obtained after about 4.3 h of gasification, was used again for approximately 3.7 h of gasification. The active carbons used effectively reduced the contents of condensed and gaseous tars. Ni-impregnated active carbon produced a gas with a high H ₂ content (26 vol%) and a low NH ₃ content (198 ppmv), while Fe-impregnated active carbon produced a gas with a low H ₂ S content (96 ppmv). The compositions of the producer gases obtained solely using condensers were similar to those obtained using electrostatic precipitator.

total pore volume slowly decreased with time.

1. Introduction

Due to a rise in global energy demand, attention is focused worldwide on producing renewable energy from various biomass sources. Gasification, a thermo-chemical conversion process, is a technique that has been in use since 1839, when the wood gasifier was built, and has recently been identified as a solution for producing renewable energy from biomass [1,2]. The gas produced from biomass gasification, called producer gas or syngas, can be used as a fuel for heat or electricity generation, or as a feedstock for chemical and fuel synthesis [3]. Biomass gasification is a promising route for H₂ production, because biomass-derived H₂ contributes to renewable energy production [4]. Impurities that evolve during biomass gasification can cause issues that need to be resolved to allow commercialization of gasification processes. For example, tar can result in mechanical problems, such as blocking the pipelines of gasification processing or fouling processing equipment, but it can also deactivate catalysts used during gasification [5,6]. Additionally, tar in producer gas prevents its direct use in gas engines or turbines. Milne and Evans proposed acceptable guideline values for tar content in producer gas for devices, as follows: less than 5 mg/Nm³ for direct-fired industrial gas turbines, and 50–100 mg/Nm³ for internal combustion engines [7]. Tar removal approaches are often divided into two methods: primary and secondary. The primary method relies on tar removal inside gasifiers, using tar-cracking additives and/ or optimal tar-cracking parameters. The secondary method uses apparatus after gasification, such as a scrubber or an electrostatic precipitator (EP). In addition to tar removal, the secondary method is also effective in removing other impurities (H₂S, NH₃, and HCl) [8]. Currently, most biomass gasification processes that are running or being tested are equipped with a scrubber and (or) an EP. This method, however, poses significant problems when commercialization of a biomass gasification process is considered, because they limit the economic efficiency of the process [9]. Another secondary method is to use catalysts in tar-cracking or reforming reactors. Wang et al. used Ni/char catalysts, and found that the catalysts led to the removal of over 97% of tar at 800 °C and increased H₂ in syngas [10]. These catalysts were also tested for removing tar inside gasifiers [11-13]. Garcia et al. investigated the effect of a Ni-Al catalyst as a fluidized bed material, and concluded that the Ni-Al catalyst enhanced H₂ production and decreased methane content in producer gas [14]. Virginie et al. examined the effect of Fe/olivine, which is inexpensive and non-toxic, on tar removal in a dual fluidized bed biomass gasification process, and concluded that it acted as a catalyst for reforming tar and hydrocarbons [15]. Ko et al. reported that Fe is effective as a sorbent of H₂S between

During the total ~ 8 h of gasification, active carbon could efficiently remove tar; however, its surface area and

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https://doi.org/10.1016/j.fuel.2017.12.068

Received 9 October 2017; Received in revised form 4 December 2017; Accepted 17 December 2017 0016-2361/ © 2017 Elsevier Ltd. All rights reserved.





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Nomenclature		EP	Electrostatic precipitator	
			ER	Equivalence ratio
	List of abbreviations		GC-TCD	Gas chromatography-thermal conductivity detector
			GC-FID	Gas chromatography-flame ionization detector
	BET	Brunauer–Emmett–Teller	ICP-AES	Inductively coupled plasma atomic emission spectrometry
	CCE	Carbon conversion efficiency	TRE	Tar removal efficiency
	CGE	Cold gas efficiency	UOS	University of Seoul
	DSS	Dried sewage sludge		

the temperatures of 500–700 °C [16]. Most catalysts used as primary and secondary methods, however, suffered from deactivation by coke, sulfur, and chlorine [17,18]. Using modified gasifiers as a primary method is very promising, because this does not rely on expensive equipment, like EPs and catalysts. Some approaches reported that modified gasifiers could help overcome the problems associated with tar contaminants, resulting in enhanced H₂-rich syngas yield and process efficiency [19,20].

Our group has developed a staged gasifier (the UOS two-stage gasifier) consisting of a fluidized bed gasifier and fixed bed reactor serially connected, and we conducted a series of gasification of dried sewage sludge (DSS) [21-24]. The two-stage gasifier used active carbon as a tar removal additive, and showed excellent tar removal efficiency with high H₂ production. Recently, a new type of staged gasifier, called the UOS three-stage gasifier, consisting of three serially connected reactors (auger, fluidized bed, and tar cracking reactors), was developed to remove tar more efficiently than the UOS two-stage gasifier. This paper reports for the first time the experimental results of the three-stage DSS gasification, solely using condensers as the primary tar removal method without using any typical expensive tar removal apparatus, such as an EP or a scrubber [25]. This paper also reports the effects of Fe and Niimpregnated active carbons in the three-stage gasification on the impurity removal (tar, NH₃, and H₂S), and on the H₂ production. In addition, Fe-impregnated active carbon was firstly tested in this paper to reduce the H₂S content in producer gas, and its effectiveness was also provided in detail. Finally, this paper provides results on a long-term gasification (total ~8 h) without any EP and scrubber to observe the deactivation behavior of active carbon and producer gas development, which was the longest operation with DSS in the UOS staged gasifier.

2. Experimental

2.1. Feed material and active carbon

In the experiments, DSS obtained from anaerobic digestion supplied by a wastewater treatment facility in Korea was used. The DSS was first crushed by a disintegrator and sieved to obtain homogenous fractions with diameters ranging between 0.6 and 3.35 mm. The main characteristics of the feed material are presented in Table 1.

Table 1 shows that the DSS contained high nitrogen ($\sim 5 \text{ wt\%}$) and sulfur (~2 wt%) contents. Nitrogen and sulfur in feed material are the precursors for NH₃ and H₂S liberated during gasification. The DSS had a high ash content (\sim 40 wt%), with Fe, Ca, Mg, and Al as the main metal components. The lower heating value of DSS determined using a bomb calorimeter (Model 6100, Parr) was about 13 MJ/kg. Silica sand was used as the bed material in the fluidized bed gasifier, which had a diameter between 150 and 300 µm. A coal-based virgin active carbon with a size of 1-2 mm and a surface area greater than $1030 \text{ m}^2/\text{g}$ was used as a tar-cracking additive in the three-stage gasification process. Along with this, metal-impregnated (Ni and Fe) active carbons were also used. The surface areas of the Ni- and Fe-impregnated active carbons, with metal content amounting to $\sim 9 \text{ wt\%}$, were approximately 960–980 m²/g. Metal impregnation was conducted as follows: first, a metal (either Ni or Fe) ion solution was mixed with a coal-based active carbon and stirred. During stirring, the mixture was heated for $\sim 4 \text{ h}$ to remove any water content. After heating, the remainder was dried overnight at 90 $^{\circ}$ C, and then calcined at 400 $^{\circ}$ C to activate it and remove impurities.

2.2. The UOS three-stage gasification process

A diagram of the three-stage gasification process is presented in Fig. 1.

The three-stage gasifier consists of an auger, a fluidized bed, and tar-cracking reactors, connected serially. The fluidized bed and tarcracking reactors are separated by a perforated distributor with 1 mm pores. The auger reactor, which is absent from the UOS two-stage gasifier, has a length of 850 mm and an inner diameter of 32 mm and is located in front of the fluidized bed reactor. The auger reactor plays an important role to reduce the tar content in producer gas: it generates a huge amount of tar in advance of the main gasification in the fluidized bed reactor. A portion of the tar generated in the auger reactor can be destroyed through thermal and oxidative thermal decomposition inside the auger reactor. The undestroyed tar leaving the auger reactor can have a long time to react with air used as the gasifying agent inside the fluidized bed reactor, compared to tar directly generated inside a gasifier. The fluidized bed reactor of the three-stage gasifiers, which is the main gasifier, has a height of 390 mm and an inner diameter of 110 mm. Tar can also be produced inside the fluidized bed gasifier by the gasification of rest feed material from the auger reactor and it then enters the tar cracking reactor, which has a height of 410 mm and an inner diameter of 160 mm. The active carbon in the tar cracking reactor absorbs and cracks tar. After gasification in the three-stage gasification process, producer gas can be cleaned by condensers or an EP. Details of the process elements and the operational procedure for the three-stage gasification process have been previously published [26,27].

2.3. Experimental conditions

In all the experiments, the feed rate was 1 kg DSS/h, and gasification time ranged from ~1 to 4.3 h. The amount of silica sand used as the fluidizing bed material was 2.5 kg. Preheated air (~550 °C) was used as the gasifying agent, and the air flow rates for all of the experiments were ~17 Nl/min. The amount of active carbon used, whether it was the virgin or metal impregnated active carbons, was 1 kg. The contact

Table	1			
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Dried sewage sludge						
Proximate analysis ^a (wt%)		Ultimate analysis ^b (wt%)				
Moisture Volatile matter Fixed carbon Ash	$\begin{array}{l} 7.07 \ \pm \ 0.31 \\ 51.53 \ \pm \ 1.39 \\ 3.99 \ \pm \ 0.28 \\ 37.41 \ \pm \ 1.02 \end{array}$	Carbon Hydrogen Nitrogen Oxygen ^c Sulfur	$\begin{array}{r} 31.32 \ \pm \ 0.11 \\ 4.56 \ \pm \ 0.08 \\ 4.72 \ \pm \ 0.02 \\ 20.72 \ \pm \ 0.31 \\ 1.27 \ \pm \ 0.01 \end{array}$			
Lower heating value ^c (MJ/kg)		13.2				

^a ASTM E871-82, ASTM E872-82, ASTM D1102-84.

^b dry basis.

c calculated by difference.

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