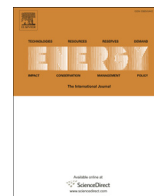




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Two-stage air gasification of mixed plastic waste: Olivine as the bed material and effects of various additives and a nickel-plated distributor on the tar removal

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ABSTRACT

Air gasification of mixed plastic waste was conducted in a two-stage gasifier. The effects of the combination of olivine as the fluidized bed material and activated carbon with or without other additives for tar cracking, as well as a Ni-plated distributor, the use of steam as a gasifying agent, and the calcination of olivine on the producer gas compositions and tar production, were also investigated. The maximum H₂ concentration (27.3 vol%) was obtained with 900 g of activated carbon in the tar-cracking zone, and through the use of calcined olivine as the bed material. In the experiments, the maximum tar removal efficiency calculated using a base case reached 98.2%. The LHV_s of the producer gases were in the range of 6.1–9.0 MJ/Nm³. The increase in the activated carbon amount led to an enhanced H₂ production, as well as a decrease in tar production. The Ni-plated distributor was found to be effective for tar removal. In the application of dolomite in the tar-cracking zone and the use of steam as a fluidizing medium resulted in a high rate of HCl removal. The minimum HCl concentration in the producer gases was under 1 ppm.

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1. Introduction

Producer gas from gasification has a wide variety of applications, as it can be integrated into several industrial processes, or can be used for power generation [1]. However, the removal of contaminants in producer gas should be considered for many applications of producer gas (synthesis gas). In particular, the so-called tar in producer gas has been one of the main constrictions in the application of producer gas [2,3]. Tar is defined as hydrocarbons with a molecular weight higher than benzene (>78 kg/kmol) [4]. A producer gas with a tar content of less than 100 mg/Nm³ is typically suitable for use in internal combustion engines, and one less than 5 mg/Nm³ is suitable for use in gas turbines [5]. In order to reduce the amount of tar generated, two tar removal methods, a direct reduction of tar in a gasifier (primary method) and a gas cleaning method after a gasifier (secondary method), have been widely studied [6]. The primary method may be fundamentally more ideal, and is mainly related to the design an effective gasifier to yield producer gas of low-tar content [7]. When using catalysts for tar

removal, natural catalysts and Ni-based catalysts have mainly been applied [8–10]. Among these catalysts, an inexpensive and non-toxic olivine, a mineral containing magnesium, iron and silicon, has been considered as a suitable catalyst for the tar removal in the fluidized bed gasification [11–15]. Meanwhile, Di Gregorio et al. investigated the fluidized bed gasification of a packaging derived fuel from the energetic, environmental and economic viewpoints [16].

As reported in earlier papers, olivine showed a slightly lower catalytic activity towards tar reduction in the mixed plastic waste gasification, but a higher attrition resistance than dolomite [17,18]. In this study, again olivine as the fluidized bed material and activated carbon as the tar-cracking additive were applied in a two-stage gasifier. Main difference from the previous work [18] is the use of additional additives (calcined dolomite and calcined zeolite) along with activated carbon in the upper reactor to see the effects of the combination of the additives on the reduction of tar and HCl in producer gas. Meanwhile, instead of a stainless distributor, a Ni-plated distributor was newly applied in this study to reduce tar content in producer gas. Along with the Ni-plated distributor, this work will also show the effects the addition of steam as a gasifying agent (together with air) and the calcination of olivine, on producer gas composition and tar production.

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2. Experimental

2.1. Feed material and additives

A pellet-type fraction of mixed plastic waste was used as fuel in the gasification process. The mixed plastic waste was supplied by a recycling company, and was comprised of various types of plastics, such as polyethylene, polypropylene, polystyrene, polyvinyl chloride, PMMA (polymethylmethacrylate) and PET (polyethylene terephthalate). The mixed plastic waste was first sieved to obtain a material with a diameter between 2 and 3.35 mm, which was then applied to the various gasification processes. Table 1 shows results of the proximate, ultimate, and calorific value analyses of feed samples.

As shown in Table 1, the mixed plastic waste was mainly composed of volatile matter. The nitrogen and chlorine contents in the feed material were low, with values of 0.18 and 1.13 wt%, respectively. The LHV (Lower Heating Value) of the mixed plastic waste, which was calculated by Dulong's formula, was determined to be 43.7 MJ/kg.

Natural olivine with a diameter of 150–300 μm was used as the fluidized bed material. The composition of the olivine was 39.6 wt% SiO_2 , 26.5 wt% Fe_2O_3 , and 27.9 wt% MgO , along with other minor components such as Al_2O_3 and CaO . In some experiments, natural olivine was calcined in air flowing at 10 NL/min at a temperature of 900 $^\circ\text{C}$ for 10 h. A commercially available pellet-type coal-based activated carbon of about 3.35–10 mm was applied as a tar-cracking additive. The specific surface area of the activated carbon was 1020 m^2/g . Natural dolomite and zeolite ($\text{M}_2/\text{n} \cdot \text{Al}_2\text{O}_3 \cdot \text{xSiO}_2 \cdot \text{yH}_2\text{O}$, M: metal, n: valence of the metal cation) were also applied, as tar-cracking additives, to the experiments. The natural dolomite used in this study was mainly composed of 29 wt% CaO , 20 wt% MgO and 48 wt% CO_2 . In the experiments, natural dolomite and zeolite were also calcined at the temperature of 900 $^\circ\text{C}$ for 10 h in a furnace. The diameters of calcined dolomite and zeolite were in the range of 2–3.35 mm.

2.2. Gasifier and experimental procedure

A schematic of the gasification process is shown in Fig. 1.

The gasification plant consists of four main sections: feeding system, two-stage gasifier, char removal system and quenching system. The two-stage gasifier (denoted as the UOS gasifier) consists of a bubbling fluidized bed (lower reactor) and a tar-cracking zone (upper reactor) separated by a distributor. The two reactors are made of a 310 SS tube and are indirectly heated with electricity. The fluidized bed reactor has an inner diameter of 110 mm and a height of 390 mm. The upper reactor, which is operated as a fixed bed reactor, has an inner diameter of 160 mm and a height of 340 mm. To determine the reaction temperature in the experiments, four thermocouples were installed in the lower reactor and

two in the upper reactor. The reaction temperature was defined as the average value of the thermocouples, showing a maximum difference among readings from four thermocouples of about 10 $^\circ\text{C}$ during operation. Unlike typical one-stage gasifiers, the two-stage gasifier has the advantage that various additives can be introduced at the same time for the tar removal process: the lower reactor contains the fluidized bed material, which can be used for the tar cracking catalysts, and the upper reactor contains tar-cracking agents that can further remove tar components flowing from the lower reactor. The feeding system is comprised of two screw feeders, which work to uniformly convey the feed material. The upper screw feeder controls the feeding rate, while the lower feeder, equipped with a water jacket, rotates rapidly to uniformly unload the feed material.

In the experiments, the fluidizing gas (air) was heated to 650 $^\circ\text{C}$ using a pre-heater, and entered into the lower reactor through a distributor plate made of steel. After leaving the two-stage gasifier, the producer gas passed a cyclone and hot filter made of Al_2O_3 to capture particles larger than 10 and 2 μm , respectively. After particle removal, condensable components in the producer gas were removed at the quenching system, which consists of three steel condensers cooled to 10 $^\circ\text{C}$ with water. Steam entered into the lower reactor from below the distributor with the help of a steam generator. Some of non-condensable producer gas was sampled by means of teflon gas bags (1L) at intervals of 5–10 min to analyze its composition, and the remaining gas stream was burned in a flare stack after its gas volume and temperature were measured. All experiments were conducted without an electrostatic precipitator, which is commonly applied to clean producer gases in gasification systems.

2.3. Reaction conditions

In all the experiments, the reaction temperatures of the lower and upper reactors were maintained at about 800 $^\circ\text{C}$, and ER (equivalence ratio), defined as the air to fuel weight ratio used in the gasification process divided by the air to fuel weight ratio used for stoichiometric combustion, was approximately 0.3. Flow rates of preheated air were approximately 15 NL/min, which corresponded to gas velocity of approximately 3.7 U_{mf} (the calculated minimum fluidization velocity). The calculated gas residence time in the fluidized bed was approximately 1 s. The amount of feed material in each experiment was 0.3 kg. The feed rate was in the range of 4.3–5.0 g/min. The operation time of each run was 60–70 min. The reaction conditions are summarized in Table 2.

Gasification experiments (Runs 1–3) were conducted to investigate the influence of the combination of calcined dolomite and activated carbon in the upper reactor on the composition of producer gas and the tar reduction. When activated carbon was added, dolomite was first put on the distributor plate located between two reactors, with activated carbon placed on top. The sum of activated carbon and calcined dolomite in the upper reactor was fixed to 1500 g, while the relative amount of each additive was varied between 0 and 900 g for activated carbon and between 600 and 1500 g for calcined dolomite. The contact time between the producer gas and the additives in the upper reactor was between 0.63 and 0.9 s (calculated as the voidage volume of the activated carbon bed (0.55) and dolomite (0.41) divided by the total volumetric flow of the producer gas at the reaction temperature). In Run 4, calcined zeolite was used in the upper reactor instead of calcined dolomite. The results of Run 4 were compared with those of Run 3. Run 5 was carried out using a Ni-plated distributor between the lower and upper reactor instead of the stainless steel distributor to reduce tar in the producer gas. In Run 6, steam as an additional gasifying agent together with air, was introduced into the lower reactor to examine

Table 1
Properties of the mixed plastic waste.

Proximate analysis ^a	Mean \pm standard deviation (wt%)	Elemental analysis	Mean \pm standard deviation (wt%)
Moisture	0.38 \pm 0.01	Carbon	82.41 \pm 0.02
Volatile matter	94.71 \pm 0.06	Hydrogen	13.42 \pm 0.01
Fixed carbon ^b	4.37 \pm 0.03	Oxygen	2.86 \pm 0.12
Ash	0.54 \pm 0.03	Nitrogen	0.18 \pm 0.01
		Chlorine	1.13 \pm 0.00
		LHV (MJ/kg)	43.7

^a ASTM D3172.

^b By difference.

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