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Promoting gas production by controlling the interaction of volatiles with char during coal gasification in a circulating fluidized bed gasification reactor ☆

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ABSTRACT

A novel circulating fluidized bed (CFB) gasifier, which consists of a downer and two bubbling fluidized beds, was developed. Coal is continuously pyrolyzed in the downer and the resultant char is sent to the bubbling bed gasifier for steam gasification while evacuation of the volatiles avoids the char–volatile interactions. Any ungasified char is transferred from the gasifier to the bubbling bed combustor, where it is either completely combusted or partially combusted and then recycled to the downer. Steam gasification was successfully performed in the absence of pyrolysis-derived volatiles, which strongly inhibit gasification. The recycling of the partially combusted char greatly increased its concentration in the downer thereby enhancing the reforming of the volatiles, in particular that of tar over that of char, and the resultant gas formation. The total yield of gases from the downer and bubbling bed gasifier was 45% higher than that obtained under direct feeding of the coal into the bubbling bed gasifier and full combustion of the char, i.e., with steam gasification of char in the presence of volatiles and subsequent reforming over char at a much lower concentration.

1. Introduction

The development of highly efficient coal gasification technology has attracted significant attention. The product gas can be used as fuel for power generation or further processed and concentrated into chemical or liquid fuel. For power generation, an integrated coal-gasification combined-cycle (IGCC) power plant, which represents the most environmentally friendly coal-fired power generation technology, has been developed and its efficiency is higher than that of conventional coal-fired power plants. More recently, an advanced-type integrated coal-gasification combined-cycle system (A-IGCC) was proposed by Tsutsumi et al. [1,2]. The A-IGCC system is based on an exergy recuperation concept that involves recycling the exhaust heat of the gas turbine (GT) via an endothermic reaction during steam gasification. The A-IGCC system can drastically improve the thermal efficiency; as a result, its overall efficiency is theoretically higher than that of a conventional IGCC system, which uses an entrained-bed-type gasifier. In the A-IGCC system, coal is gasified at a relatively low temperature (<1173 K) using only the steam generated by the exhaust heat of the GT. The design of the reactor for low-temperature steam gasification of coal is essential for development of the A-IGCC system. It is well known that the rate of coal gasification is quite low at low temperatures; therefore, the entrained-bed-type gasifier employed in the conventional IGCC system cannot be used. A fluidized-bed-type gasification reactor is a potential replacement because the residence time (i.e., reaction time) of the coal in the gasifier can be adjusted. If the essential conditions for a new type of coal-gasification process based on fluidized-bed technology can be elucidated and satisfied, the process can be applied to further optimize the A-IGCC system. In addition, the new process can also be applied to highly efficient systems for the production of chemical or liquid fuel via gasification.

To realize low-temperature gasification using a fluidized-bed reactor for the A-IGCC system and/or highly efficient gasification system for chemical production, common problems, such as the large amount of tar formed, low gasification rate of char, etc., should be solved from the chemical reaction control perspective. It is known that char gasification is inhibited by the presence of co-existing gasses [3–12]; minimizing this inhibition is a key to promoting char gasification. In our previous study [13], a novel circulating fluidized-bed (CFB) gasification reactor was developed to promote coal char gasification at low temperatures. In the reactor, the pyrolysis unit (i.e., pyrolyzer) is physically separated from the char gasification unit (i.e., gasifier). The pyrolyzer plays a role in the separation of the volatiles and char, and, in the absence of volatiles, char gasification could proceed in the gasifier. Thus, the inhibition of char gasification inhibition was successfully

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minimized resulting in promotion of gasification. However, to demonstrate the basic concept for a highly efficient coal gasification process, further detailed study is needed. In particular, the theoretical optimal operating conditions for the fluidized bed gasifier should be elucidated and the technical feasibility should be experimentally demonstrated.

In the present study, we first theoretically demonstrate the process efficiency and optimal operating conditions to maximize the process efficiency using circulating fluidized bed technology. Second, solutions to the barriers to achieving highly efficient coal gasification, i.e., improvement of char gasification rate and suppression of tar emission, are experimentally demonstrated using a new pyrolyzer-separated circulating fluidized bed gasification reactor on the laboratory scale. Finally, the feasibility of the theoretical operating conditions was verified by comparison with experimental data.

2. Experimental

2.1. Sample

Indonesian low-rank coal (i.e., Adaro coal) was used as the sample, and its properties are listed in Table 1. The sizes of the raw coal were in the range of 0.5 to 1.0 mm. The samples were dried at 353 K *in vacuo* for 8 h prior to use.

2.2. Apparatus

A new pyrolyzer-separated circulating fluidized-bed gasification reactor was developed. Fig. 1 shows a schematic diagram of the reactor, which comprises an interconnected drop-tube furnace (DTF) pyrolyzer, bubbling-bed gasifier, and bubbling-bed combustor. Bed material was dropped from the pyrolyzer into the dense bed of the gasifier, and the outlet gas from the pyrolyzer could not flow into the dense bed because of the configuration of the gasifier. The bed material was circulated from the gasifier to the combustor through a screw feeder between the gasifier and combustor. The bed material was circulated back from the combustor to the pyrolyzer through the H-shaped loop seal valve. Although the screw feeder was not suitable for industrial application for controlling the circulating rate of the bed material, the screw feeder can accurately control the circulating rate of the bed material in laboratory scale reactor. Therefore, the screw feeder was here applied for controlling the circulating rate. The temperature of pyrolyzer, gasifier and combustor was stable during the circulation of the bed material in preliminary experiments, indicating that stable circulation of the bed material in the reactor was achieved. The mean residence time of the bed material of each reactor was consequently determined by the circulating rate of the bed material and the size of the reactor. The mean residence time in the gasifier and combustor was ca. 20 and 10 min, respectively. In other words, the size of gasifier and combustor was designed to obtain the above mean residence time.

2.3. Bed material

In the present study, porous γ-alumina with a pore volume of 0.8 cm^3/g and specific surface area of 200 m^2/g was used as the bed material. The sizes of the porous γ-alumina particles were in the range of 75–150 μm. Some researchers have reported the activity of alumina towards the cracking/reforming of tarry materials formed from coal or woody biomass [14–19]. According to the reports, alumina can rapidly capture tarry materials and deposit and crack them to form light gas and coke.

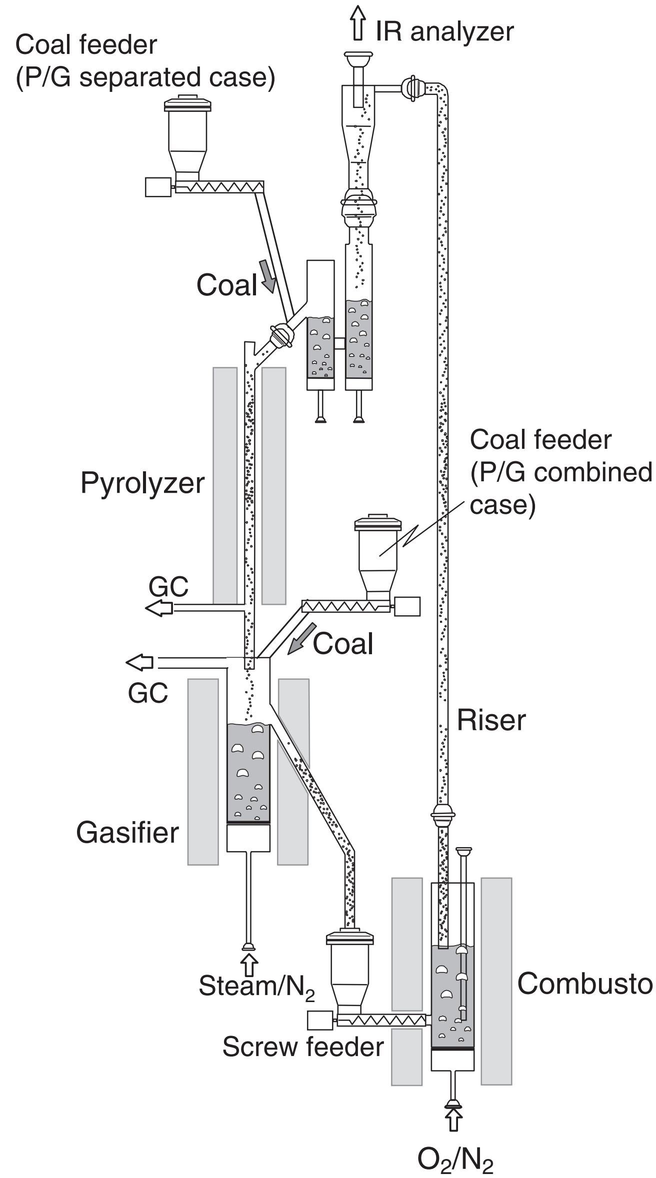

Fig. 1. A schematic diagram of the pyrolyzer-separated CFB gasification reactor.

Table 1
Properties of Indonesian Adaro coal.

	C	H	N	O(diff)	Ash
	[wt%, daf]				[wt%, dry]
Adaro	70.4	5.3	0.8	23.5	0.6

2.4. Procedure

Raw coal was fed into the pyrolyzer to evacuate the volatiles. As mentioned above, most of the tarry materials are deposited as coke on the alumina bed material in the pyrolyzer. The volatiles derived from pyrolysis could not flow into the dense bed of the gasifier while the resultant char was dropped into the dense bed with the bed material. Thus, the pyrolyzer played a role in separating the char and volatiles. The char and coke deposited on the alumina were simultaneously gasified with steam in the absence of volatiles. The molar ratio of steam to carbon (S/C) in the raw coal ranged from 1.0 to 2.0. The temperatures of the pyrolyzer and combustor were fixed at 1173 and 1223 K, respectively, while the gasifier temperature ranged from 1073 to 1173 K. The product gas, which comprised H_2, CO, CO_2, CH_4, C_2H_4,