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Research article

Pyrolysis of low-rank coal with heat-carrying particles in a downer reactor

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ABSTRACT

A novel co-current downer pyrolyzer was developed that can feed heat-carrying particles (silica sand, particle size, $d_{p,\text{sand}} = 0.116$ mm) preheated in a fluidized bed separately with coal from the top of the reactor. This allows the high-temperature particles to directly contact coal particles. The influence of the pre-heated heat-carrying particles on the pyrolysis of dried Loy Yang coal particles (particle size, $d_{p,\text{coal}} = 0.350$–$0.500$ mm) in a quartz glass downer reactor (1.0 m in length and 20 mm in inner diameter) was investigated at 1173 K in the absence and presence of the steam (up to 30 vol%) under different silica/coal ratios. Increase in sand feed rate increased the amount of carbon surface deposits and the yields of CO_2 and CO. TOF-MS and FT-IR spectra of the produced heavy tar and deposited carbon on silica sand indicated that the heavy tar very rapidly deposited on the surfaces of sand and forms cokes by polymerization. No significant influence of the steam on pyrolysis reaction was observed within the estimated residence time of coal particles (~0.54–0.61 s) in this pyrolyzer.

1. Introduction

The integrated coal gasification combined cycle (IGCC) has received much attention because of its higher thermal efficiency than conventional coal-fired power generation systems [1–5]. To increase thermal efficiency of the IGCC systems, advanced IGCC and advanced integrated gasification fuel cell combined cycle systems have been proposed [6–16]. In these systems, steam recovers the exhaust heat of a gas turbine or solid oxide fuel cell. The steam is then utilized as a heat source for endothermic gasification. The exhaust heat is much cooler than is suitable for entrained flow bed gasifiers [10,15]. Therefore, the authors proposed a novel triple-bed combined circulating fluidized bed (TBCFB) gasifier, consisting of a downer pyrolyzer, a bubbling fluidized bed char gasifier and a riser unreacted char combustor [10,15,16]. The authors analyzed hydrodynamic behaviors of solids in the TBCFB with high mass flux at ambient temperature and pressure [15–22].

Matsuoka et al. investigated reactivity of Adaro coal (particle size, $d_p = 0.5$–1.0 mm) char in steam gasification at 973–1173 K [23,24]. They used a lab-scale TBCFB gasifier, which physically isolates volatiles, especially hydrogen, with char soon after pyrolysis to avoid the strong volatile–char interaction during gasification. They reported char gasification was significantly promoted at 1073–1173 K by the physical isolation of the pyrolyzer from the gasifier. However, detailed characteristics of pyrolysis in the TBCFB remain unknown.

Downer pyrolyzer performance is considered to have a substantial influence on subsequent steam gasification in the TBCFB reactor because of the strong volatile–char interaction and reduction in tar emissions [11,23,24]. Reactivity of coal in pyrolysis has been examined using a drop-tube reactor (DTR) (free-fall reactor) [25–31] or a downer reactor [32–34]. Yan et al. investigated devolatilization from coal in slow pyrolysis (10^{-1} to 10^{1} K/s) and fast pyrolysis (10^{2} to 10^{4} K/s) using a drop-tube reactor [31]. They reported that increasing the reaction temperature and heating rate of coal greatly increased the yield of light gases owing to enhancement of secondary tar cracking reactions. Hayashi et al. investigated composition of volatiles produced from pulverized Australian Yallourn brown coal during pyrolysis using a DTR and Curie-point reactor in the presence of steam [26]. They reported that tar yield decreased due to dealkylation forming gaseous hydrocarbons and deoxygenation forming carbon monoxide. They also reported that the addition of steam increased the total conversion of carbon into volatiles at 1173 K within an estimated residence time of coal/char particles (<2 s). Zhang et al. prepared Loy Yang coal chars and blended these with original Loy Yang coal at ratios of 50%–85%. They investigated the reactivity of the blended sample in a DTR (2.55 m in length and 15 mm in inner diameter) at 1173 and 1223 K [29]. In a follow-up study using the same DTR at 1123–1173 K, they reported that Na and Ca in the raw coal are highly active catalysts for char gasification and

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Nomenclature

A	surface area of particles [m^2]
C_D	drag coefficient [−]
C_p	specific heat [kJ/(kg K)]
d_p	average particle size [m]
F	view factor [−]
F_p	feed rate of particles [g/min]
g	gravitational acceleration [m/s^2]
h	heat transfer coefficient [W/(m^2 K)]
Nu	Nusselt number [−]
Pr	Prandtl number [−]
Q	heat transfer rate [W]
Re_p	particle Reynolds number [−]
t	time [s]
T	Temperature [K]
v_g	gas velocity [m/s]
v_p	particle velocity [m/s]
V_{reac}	reactor volume of the downer [m^3]
z_{coal}	vertical displacement of coal [m]
z_{sand}	vertical displacement of sand [m]
	Greek alphabets
ε	emissivity of particles [−]
ε_s	solid holdups [−]
κ	thermal conductivity of gas [W/(m K)]
μ	viscosity of gas [Pa s]
ρ_g	gas density [kg/m^3]
ρ_p	particle density [kg/m^3]
σ	Stephan-Boltzmann Constant [W/(m^2 K^4)]
τ:	estimated residence time of particles [s]

nascent tar reforming in the presence of steam [30]. Co-pyrolysis of coal and biomass in a free-fall reactor have also been conducted [35–39].

In a TBCFB reactor, coal pyrolysis occurs in a downer pyrolyzer with high-temperature heat-carrying particles that recover the combustion heat from the riser combustor. Because pyrolysis is a rapid reaction, the interactions and heat transfer between the heat-carrying particles and the coal are considered to be very important. Bi et al. have investigated pyrolysis of coal [40] and coal/biomass [41] in a moving bed pyrolyzer with hot ash (i.e., solid heat carrier). Recently, several research groups have published computational fluid dynamics data that used lump-sum or global-devolatilization pyrolysis models of downer pyrolyzers [42–44]. However, to the best of our knowledge, no experimental study has been conducted on the pyrolysis of coal co-feeding with pre-heated heat-carrying particles in a downer reactor. Detailed knowledge of coal pyrolysis behavior in a downer pyrolyzer is important for the design and operation of TBCFB reactors, to sufficiently reduce tar emission from the pyrolyzer and the following gasifier. In the present study, we developed a novel concurrent downer pyrolyzer that can directly bring coal particles into contact with heat-carrying particles (silica sand) pre-heated at approximately 1173 K using a fluidized bed preheater. We investigated the effect of the heat-carrying particles on the pyrolysis behavior of pulverized Loy Yang brown coal in the downer reactor by changing the feed rates of silica sand particles.

Table 1
Proximate and ultimate analyses of Loy Yang coal.

Proximate analysis [wt%]				Ultimate analysis [wt%, d.a.f.]			
Moisture	Ash	Volatiles	Fixed carbon	H	C	N	O (by diff.)
15.3	0.8	45.0	38.9	4.7	62.8	0.6	31.9

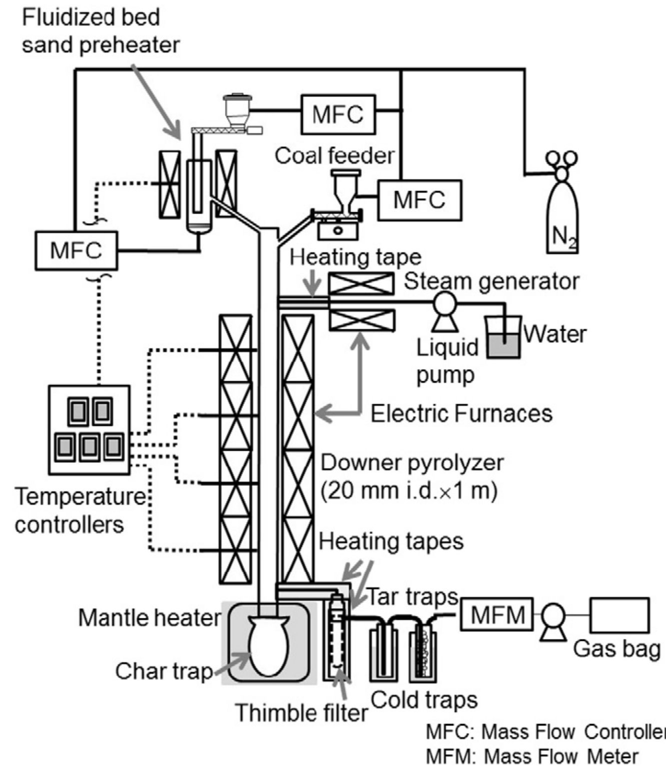

Fig. 1. Schematic view of experimental apparatus.

2. Experimental

2.1. Samples

Australian Victorian low-rank coal, Loy Yang brown coal, with particle size (d_p) ranging from 0.350 to 0.500 mm was used as the coal sample. The properties of the coal sample are listed in Table 1. The sample was dried at 363 K under vacuum for 4 h prior to experiments. Silica sand particles were sieved (<0.18 mm) and then used as heat-carrying particles.

2.2. Experimental apparatus and procedure

Fig. 1 shows a schematic diagram of a downer pyrolyzer. A reactor tube was made of quartz glass with inner diameter 0.020 m and length 1.0 m. First, the silica sand particles were fed to a fluidized bed preheater

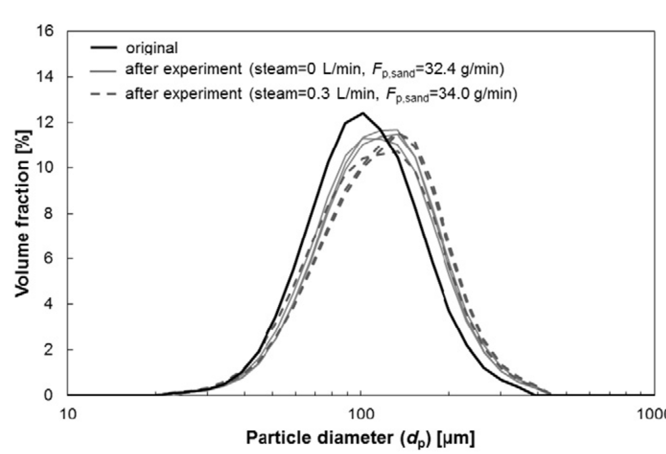

Fig. 2. Particle size distribution of silica sand. Solid black line indicates original sand; solid gray lines indicate steam feed rate of 0 L/min and sand feed rate of 32.4 g/min; and broken gray lines indicate steam feed rate of 0.3 L/min and sand feed rate of 34.0 g/min.