



Rapid pyrolysis of brown coal in a drop-tube reactor with co-feeding of char as a promoter of *in situ* tar reforming



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ABSTRACT

A Victorian brown coal (Loy Yang, LY) was co-fed with char prepared from the same coal in an atmospheric drop-tube reactor (DTR) at 900 and 950 °C in the presence of 50% steam to study *in situ* reforming of tar derived from rapid pyrolysis of brown coal over a char surface. Two different chars were prepared, including devolatilised LY at 800 °C under nitrogen flow (LYC, surface area 524 m²/g) and LYC gasified with steam at 900 °C (GLYC, surface area 734 m²/g). The concentrations of chars in the blended samples varied from 50% to 85% on a carbon basis. The yield of tar derived from LY (no char was blended) was 4.2 wt.% at 900 °C and particle residence time around 3–4 s. The yield decreased with increasing char concentration, and was 0.5 wt.% at a GLYC concentration of 85% at 900 °C. Tar conversion over GLYC was more extensive than that over LYC and occurred more significantly at a higher temperature. Increasing the feeding rate of sample particles enhanced tar reforming, suggesting that solid hold-up was an important factor determining tar conversion characteristics. Liquid chromatography (LC) showed that the major components of heavy tar included typical polycyclic aromatic hydrocarbons (PAHs) ranging from three-membered rings (acenaphthylene) to seven-membered rings (coronene).

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1. Introduction

Gasification is an efficient and environmentally friendly process of coal utilization. Low-temperature gasification has been developed to improve cold-gas efficiency [1–3]. However, low temperatures generate refractory aromatic compounds, namely tar, which not only inhibit char gasification [4,5] but also make it difficult to operate downstream facilities. Research has focused on developing technologies/methods for reducing or eliminating tar [6,7], such as liquid scrubbing and catalytic reforming. In the former process, the product gas is cooled to a target temperature to convert tar from the gas to liquid phase, resulting in an undesirable overall thermal efficiency loss. Although catalytic reforming effectively decomposes tar, it suffers from catalyst poisoning by Cl/S-containing species and deactivation by carbon deposition. Thus, it is challenging to develop a practical tar reforming system with high thermal efficiency and without catalyst poisoning and deactivation.

Because the effective decomposition of tar over biomass/coal-derived chars has been reported [8–12], a triple-bed combined circulating fluidised-bed reactor system (Fig. 1), consisting of a

downer (pyrolyser), a bubbling fluidised bed (BFB, gasifier), and a riser (combustor), was proposed [13–16]. This reactor system was expected to decompose tar at the downer and avoid the detrimental effects of tar on the following char gasification and system operation. The details of the reactor system have been reported elsewhere [8,13–16]. In brief, the downer was added to a dual-bed circulating fluidised reactor system, after the riser (combustor) and before the BFB gasifier. The fresh coal was fed, mixed with hot circulating solids, including recycled char and circulating medium, and rapidly pyrolysed in the downer at around 900 °C. The resultant tar was simultaneously and rapidly decomposed into syngas and/or coke over the nascent and recycled chars. Then, the tar-free char was separated from syngas, sent to the BFB gasifier, and gasified without the negative effects of the volatiles–char interaction [4,17–19]. The volatile reforming and char gasification are thus expected to be decoupled under this reactor system. The endothermic and exothermic processes were separated in this system, incorporating the concept of low-temperature gasification with high cold-gas efficiency [2].

The decomposition of naphthalene over chars from different biomass/coal has been investigated in a fixed bed [11]. More than 90% of the naphthalene, one of the most refractory compounds among tar constituents, was converted over every char within a gas residence time of 0.25 s. We studied the elimination of nascent tar via pyrolysis of biomass over nano-porous alumina in the

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presence of steam [20]. We found that tar constituents, except benzene and naphthalene, were completely decomposed over the coke accumulated in the pores of the alumina within a gas residence time of 0.03 s. These studies demonstrate the potential of utilization of char for tar elimination.

In our most recent study [8], a two-stage reactor was used to investigate *in situ* reforming of nascent tar from the rapid pyrolysis of a brown coal over char from the same coal. The tar produced from coal pyrolysis almost completely decomposed over the char at 900 °C, indicating the feasibility of the proposed reactor system. The excellent performance of the two-stage reactor for tar elimination was due in part to its specific structure, which forced the nascent tar to pass through the char bed, causing a tar–char interaction. These results demonstrated the potential for rapid and complete decomposition of tar over char due to an intensified tar–char interaction.

It is not possible to use a fixed char bed for tar decomposition in a circulating fluidised reactor system, even though it showed good performance for tar decomposition. This is the first study examining tar reforming over a char surface under reaction conditions approximated in a downer reactor where coal, char particles, and gas are concurrently flowing. In this work, a drop-tube reactor (DTR) was chosen to simulate the reaction environment approximated in a downer reactor. Coal was co-fed with char into the drop-tube reactor, and the effects of the char properties, char concentration in the blended sample, temperature, and feeding rate related to solid hold-up on tar decomposition were investigated.

2. Experimental

2.1. Coal and char samples

Dried Loy Yang brown coal (LY) with particle sizes ranging from 0.5 to 1.0 mm was used as a coal sample. Two different char samples were prepared from the LY. The devolatilised char, hereafter referred to as LYC, was prepared in the same way as in our previous study [8]. In short, the LY was heated in a fluidised-bed reactor under an atmospheric flow of N₂ as a fluidising gas without fluidising medium at 800 °C for 30 min. Gasified LYC (GLYC) was also prepared by gasifying LYC at 900 °C for 20 min with steam using a fluidised-bed reactor. Both the LYC and GLYC were sieved to collect a fraction with particle sizes of 0.35–0.50 mm, which was used to prepare the blended samples. Table 1 lists the elemental compositions of LY, LYC, and GLYC, and the specific surface areas of the chars measured by N₂ adsorption at –196 °C (Quantachrome, Autosorb-1) using the BET method.

2.2. *In situ* tar reforming in a drop-tube reactor

A schematic diagram of the experimental set up is illustrated in Fig. 2. The DTR included a transparent quartz tube, 2550 mm long with a 15-mm internal diameter, which was heated externally with electric furnaces. An isothermal zone, ~2100 mm long, was confirmed by measuring the temperature distribution inside the reactor using a thermocouple. The sample particles were continuously fed into the DTR from a screw feeder at a feeding rate of 0.15 and 0.5 g/min under nitrogen flow as the primary carrier gas. The nitrogen–steam mixed gas was preheated at 200 °C and supplied to the DTR. The partial pressure of steam was 0.05 MPa. The gas residence time in the reactor was about 6 s, and that of the particles was estimated to be 3–4 s.

Blended samples were prepared by mixing LY and LYC/GLYC at different ratios, and subjected to *in situ* tar reforming at 900 and 950 °C. The products were collected in a collection train consisting of a char trap heated at 200 °C with a flexible heater, a silica-fibre-made thimble filter (200 °C), two cold traps kept at –73 °C, and a

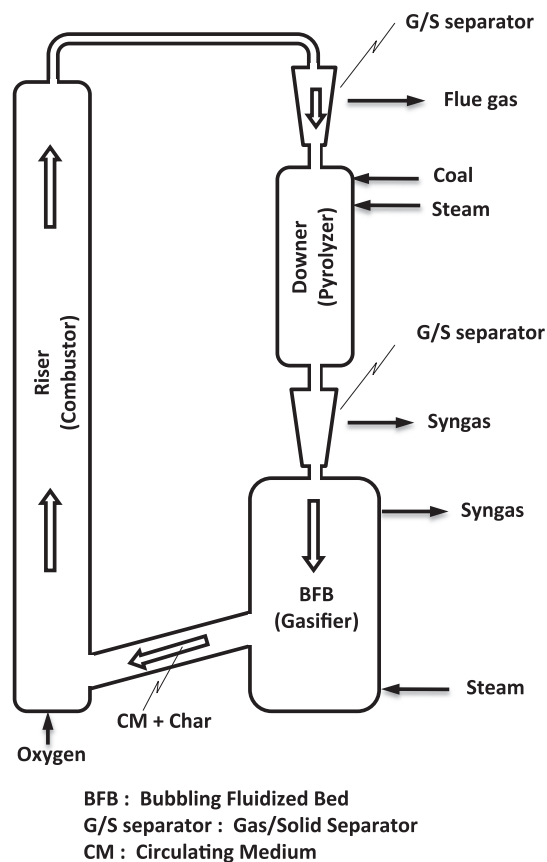


Fig. 1. Schematic diagram of the triple-bed combined circulating fluidised-bed reactor system.

Table 1

Properties of Loy Yang coal (LY), devolatilised LY (LYC), and steam-gasified LYC (GLYC).

	Ultimate analysis (daf ^a wt%)				Specific surface area ^c (m ² /g)
	C	H	N	O ^b	
LY	66.90	4.45	0.55	28.10	–
LYC	91.27	1.48	0.73	6.52	524
GLYC	96.00	0.70	0.32	2.98	734

^a Dry ash-free.

^b By difference.

^c Determined by N₂ adsorption at –196 °C using the BET method.

gas bag. The downstream one of these two cold traps was filled with glass beads to enhance its capability of recovering condensable compounds by intensifying heat transfer. Char was separated from gaseous products in the char trap. A portion of condensable compounds in the gaseous products, i.e., tetra-aromatics and greater polycyclic aromatic hydrocarbons (PAHs), were deposited on the thimble filter, while the others, such as mono- to tri-aromatics and steam, were condensed in the cold traps. The non-condensable gases (carbon oxides and gaseous hydrocarbons, CO_x and GHCs) passed through the cold traps and were collected in the gas bag.

The cold traps were washed with methanol. We defined this methanol-soluble portion of the organic compounds condensed in the cold traps as light tar. Then, the two cold traps, the thimble filter, and the empty char trap after recovering char were washed with dichloromethane. The resultant slurry was filtered using a membrane filter with a pore size of 0.45 μm to separate soot and solution. The soot remaining on the membrane filter and in/on the thimble filter after washing was quantified based on the mass

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