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Entrained flow gasification behaviour of Victorian brown coal char at low temperature

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ABSTRACT

This paper presents gasification data for two Victorian brown coal chars (Yallourn and Morwell) in CO_2 at temperatures between 700 and 1000 °C. The effects of temperature, gas-particle contact mode (entrained mode vs drop-tube mode), reactant gas concentration and residence time on carbon conversion, and syngas yield and composition was investigated. The results indicate that at a higher temperature and CO_2 concentration, carbon conversion increases as expected, along with syngas yield and composition. It is found that gasification under entrained mode achieves higher carbon conversion and gas yield than that under the drop-tube mode, even at a shorter residence time. The finding suggests that the gasification experiments should be conducted under entrained flow condition (gas velocity > particle velocity) to truly reflect gasification performance. At 1000 °C, with an increase in residence time, the carbon conversion and syngas yield gradually increases to nearly 100% and an estimated equilibrium value, respectively. The experimental data establishes that for a complete char conversion, around 17 s and 21 s residence times are required for Yallourn and Morwell, respectively, if gasification is conducted at 1000 °C in an environment of 20% CO_2. This finding indicates that Victorian brown coals need to be gasified at a temperature above 1000 °C to shorten the residence time considering that the typical residence time of industrial entrained-flow gasifier is 6–10 s.

1. Introduction

Coal is still the most important fuel for primary energy production in the world, and it will continue to dominate the global power sector in the foreseeable future [1]. The latest BP statistical report indicates that current coal resources (1140 billion tons) can meet the world's coal demand until 2170 at current consumption rates [2]. Low rank coals, including sub-bituminous and brown coals, make up approximately 30 of the coal reserves worldwide. In Australia, Victorian brown coals represent a significant, low cost energy resource with reserves of 430 billion tons [3]. However, its utilization is limited to mine-mouth power generation using conventional pulverized coal-fired combustion units at relatively low efficiencies. Therefore, it is necessary to assess and develop alternative utilization techniques for this vast resource, such as the production of high-value chemicals.

Gasification is an alternative mode of coal utilization. Coal gasification is a thermochemical conversion method that produces syngas, consisting of carbon monoxide, hydrogen, and methane, by reacting coal with gasification mediums like steam, carbon dioxide, and oxygen. A variety of chemicals and liquid fuels can be obtained from the syngas produced from coal gasification [4,5].

The major gasifier types used in large scale include fixed bed, fluidized bed, and entrained flow gasifiers [6]. Of these, the entrained flow gasifier is attractive for commercial plants due to its flexibility of using different feedstocks [7]. For syngas production, entrained flow gasifiers dominate the world gasification market [8], but specific technological information for gasification of brown coals is limited.

Several studies on entrained flow gasification of coal have been reported in the literature. Numerical simulation of coal gasification performance using an entrained flow gasifier was developed by Labbafan et al. [9] and Kong et al. [10]. The effect of temperature on the carbon conversion and gas quality of a German lignite during gasification at high temperature (1000–1400 °C) under oxygen atmosphere was investigated by Tremel et al. [11]. While the gasification characteristics, including gas compositions and carbon conversion, of coke and bituminous coal under oxygen atmosphere was studied by Lee et al. [12], investigations on gasification behaviour of Victorian brown coal are limited. There are small scale studies assessing alkali and alkaline earth metal (AAEM) emissions and their catalytic effects during pyrolysis and gasification of Victorian brown coals and other lignites [13–15]. There has been one study reporting on pressurized fluidized bed gasification of Victorian brown coals at a pilot scale [16]. This

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study concluded that fluidized bed gasification might not be suitable for Victorian brown coals and recommended investigations using entrained flow gasification at low to high temperature ranges. The reason for the unsuitability of fluidized bed for the gasification of Victorian brown coal was stated to be its friable nature and elutriation of the carbon rich fine char particles from the gasification zone.

The majority of the reported entrained flow gasification work are on bituminous coals or other lignites. Only one study [17] is reported in the literature on entrained flow pyrolysis and gasification of one Victorian brown coal (Morwell coal) at low temperature. This study was conducted at temperatures up to 1000 °C and concluded that 6 s residence time at 1000 °C in an atmosphere of 20% CO_2 and 80% N_2 resulted in only 56% carbon conversion. It was not known from this study [17] what residence time would be required for ~100% carbon conversion. Also, this study (a) is on only one variety of Victorian brown coal, (b) does not deal with the difference in performance between drop-tube mode and entrained flow mode, (c) does not assess the effects of residence time, and (d) does not deal with char morphology change.

The above are important scientific information necessary for technology development, but not available in the literature. This paper fills the scientific gap, and also extends the database of performance results by using two other varieties of Victorian brown coal with very different mineral composition.

In this study, the gasification behaviour of two different Victorian brown coal chars, Yallourn and Morwell is reported including the char properties and reactivity, carbon conversion, and gas quality during CO_2 gasification in a large bench scale entrained flow reactor. As part of this study, the relation between 100% carbon conversion and residence time was examined based on regasifying the chars after every run of 5–6 s residence time. This study generates fundamental information that will be useful for the development of commercial entrained-flow gasifiers using Victorian brown coals. This information is not available in any reported literature.

2. Experimental

2.1. Sample preparation

The coal samples used in this study are Yallourn (YL) and Morwell (MW) coals, two brown coals from the Latrobe Valley, Victoria, Australia. Proximate analysis was carried out in a Thermo-Gravimetric Analyser (Model STA 449 F3 Jupiter®, NETZSCH-Gerätebau GmbH, Germany), according to Australian standards -AS 2434.2, 2434.7-2434.8. The ultimate analysis was conducted by an analyser (Model 2400, Perkin-Elmer, USA). The proximate and ultimate analysis of these coals is listed in Table 1.

For the preparation of char, coal samples were first ground, air-dried, and sieved to 90–106 μm. This chosen particle size is in the range as commonly used in industrial entrained flow gasifiers. After that, the coal chars were generated by using the entrained flow reactor which was also used for char gasification in this study. The reactor was first heated to 1000 °C and maintained isothermally for 3 h. Coal samples were then fed and pyrolysed in nitrogen with a residence time of ~6 s to generate coal chars. The char properties are listed in Table 1. As seen, the residual volatile matter was 8–10%, much lower than the 47–49% volatile matter in the coal. The average particle size of the char particle is around 100 μm. The generated chars were dried at 105 °C for around 2 h in the oven immediately before the char gasification experiments.

Table 1
Proximate and ultimate analysis of coal and char samples.

Items	YL coal	YL char	MW coal	MW char
Proximate analysis (wt%)				
Moisture (as received)	5.82	0.59	14.92	3.01
Volatile matter (d.b.)	47.06	7.61	49.31	9.94
Fixed carbon (d.b.)	50.62	79.71	48.65	82.91
Ash (d.b.)	2.32	12.68	2.04	7.13
Ultimate analysis (d.b., wt%)				
Carbon	66.35	86.46	60.42	88.12
Hydrogen	4.92	0.45	4.59	0.82
Nitrogen	0.39	0.2	1.54	0.2
Sulfur	0.3	0.21	0.86	0
Ash	2.32	12.68	2.04	7.13
Oxygen (by difference)	28.77	0	30.55	3.73

d.b. = dry basis.

2.2. Apparatus and procedure

An electrically heated entrained flow reactor was employed for the gasification experiments, as described elsewhere [17]. The furnace is of 2 m length and consists of six controllable electrically heated zones. Each heating zone is separately controlled by one thermostat. During the experiments, the six heating zones were programed using the same heating rate and the system was kept isothermal at operating temperature. The quartz reactor is designed to have two layers with an inner diameter of 50 and 80 mm, respectively. The pyrolysed coal char was fed continuously from the top of the reactor at an average rate of 600 mg/min by a piezoelectric feeder and introduced into the inner chamber through a water-cooled injector. For each run, primary gas N_2 was fed from the top of the reactor at a rate of 0.5 L/min entraining the coal char into the reactor. The majority of the secondary gas used for gasification was introduced from the bottom of the reactor through the annulus at a total rate of 4.5 L/min into the external chamber. Therefore, it was heated up to the furnace temperature before entering into the inner reaction chamber along with the mixture of the coal and primary gas at the top of the reactor.

In the downstream piping, a glass beaker connecting the quartz reactor was used to collect solid products – ash and char residue. A water-ice box was used to condense the water vapour in the produced gas. To remove the small particles mixed in the produced gas, a thimble filter was installed before the gas analyser.

The reactor was heated up to the desired temperature before feeding the sample. Inert gas N_2 was fed for at least 30 min until the micro-GC showed that there was only N_2 in the outlet gas. Then, reactant gas CO_2 at a controlled flow rate was introduced into the reactor. After stable CO_2 composition had been measured by micro-GC, the coal char was fed at the top of the reactor. Throughout the experiment, the solid products were collected.

In this study, the experiments were conducted at varying temperatures (700–1000 °C), gas flow rate (2–5 L/min), CO_2 concentrations (5–20%) in N_2 and residence time (6–24 s). The effect of gas-particle contact mode, CO_2 concentration, and residence time on gasification behaviour of Victorian brown coals was investigated regarding gas yield and carbon conversion.

2.3. Data analysis

2.3.1. Solid and liquid phase analysis

Both proximate and ultimate analysis were performed on the solid residues – chars before and after gasification. The morphology change of the solid products is observed by a scanning electron microscope (FEI Nova NanoSEM 450). According to our previous study [17], little or no tar is generated from entrained flow gasification, so it is negligible and not discussed in this study.

To investigate the reactivity of the gasified chars with different residence time, the thermo-gravimetric analyzer (Model STA 449 F3 Jupiter®, NETZSCH-Gerätebau GmbH, Germany) was used for isothermal gasification at 800 °C. A carefully weighed sample of 10 mg was used for each reactivity test. Under a N_2 atmosphere, the temperature was raised to 110 °C at a heating rate of 5 K/min and then to 800 °C at a heating rate of 10 K/min. CO_2 was then introduced to the furnace and