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Low temperature entrained flow pyrolysis and gasification of a Victorian brown coal

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HIGHLIGHTS

- The entrained flow pyrolysis and gasification of a Victorian brown coal was studied.
- Entrained flow processing of this fuel was proven to be technically viable.
- Pyrolysis up to 1000 °C resulted in high surface area char suitable for gasification.
- Tars and syngas contaminants decreased with increasing pyrolysis temperature.
- Char conversion and syngas yield increased with increasing gasification temperature.

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ABSTRACT

The entrained flow gasification of Victorian brown coals is of interest for its potential use as a syngas feedstock in established commercial processes. To determine the applicability of entrained flow gasification to a Victorian brown coal, and to establish the scope for further investigation, coal pyrolysis and char gasification trials were conducted using a vertical entrained flow reactor. Pyrolysis between 800 and 1000 °C resulted in high surface area char, and tar production decreased with increasing temperature. Char conversion and syngas yield increased with increasing gasification temperature. Pyrolysis tars and contaminants in the syngas were also evaluated to determine their potential impact on the industrial process. The concept of entrained flow gasification of Victorian brown coals was shown to be operationally sound, with minimal gas and liquid phase contaminants. However, the temperature and residence times chosen for this study were insufficient to achieve complete conversion of the fuel.

1. Introduction

Entrained flow (EF) technologies are currently state of the art for coal-to-products processes, as they can achieve higher carbon conversion and a cleaner syngas product than the previously more prolific moving and fluidised bed technologies [1]. In an EF gasifier, pulverised coal is entrained in a stream of gas, and a series of pyrolysis, gasification and gas-phase reactions result in a syngas product composed predominantly of H_2 and CO with fuel-dependent concentrations of CO_2, CH_4, H_2O, and H_2S. The syngas can be cleaned and utilised directly for power generation, as in the IGCC process [2], or further converted into liquid fuels or chemical products by various commercial processes [3]. Some by-products of the coal gasification process include tars and gas- and solid-phase inorganic species. Any evolved tar species represent a reduction in process efficiency in terms of overall conversion to syngas as carbon and hydrogen are bound in the tars and therefore unavailable for conversion to the desirable products – H_2 and CO.

The behaviour, efficiency and by-products of Victorian brown coals (VBC) treated under entrained flow gasification conditions is of interest to determine the potential to convert this abundant, low quality resource into high quality, value added products for domestic and international markets. Data for various gasification processes is available for similar European [4] and US coals [5]; however it is difficult to apply data obtained using these fuels to predict the behaviour of VBC due to the differences in coal properties such as coal rank and composition, which affect the complex suite of reactions which occur [6]. Additionally, while a considerable amount of work has been done assessing fluidised bed gasification of Victorian brown coals at both ambient and high pressure [7,8], data for process design and optimisation concerning the entrained flow gasification of these fuels is not available.

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The purpose of this study was therefore to conduct a proof-of-concept investigation to determine the potential applicability of entrained flow gasification to a Victorian brown coal. By undertaking low temperature (800–1000 °C) coal pyrolysis and char gasification trials using a drop tube furnace (DTF) operated under entrained flow conditions, a baseline and scope for further investigation has been established. Numerous studies have been performed for other coals using DTF apparatus as entrained flow reactors (EFR) to simulate various aspects of industrial gasification processes [6,9,10] as they can be operated with sufficiently short residence times under isothermal conditions. In order to gain a clear understanding of the potential effect of various parameters on the process operability and products from the entrained flow gasification of Victorian brown coal, and to provide a baseline for further investigations, the two main process steps – coal pyrolysis and subsequent char gasification – were studied separately, and the implications of the results with respect to the industrial process considered.

2. Materials and methods

2.1. Raw materials

Morwell brown coal was used in this study. The sample was air-dried, then pulverised and sieved to obtain particles within the size range 90–106 μm, representative of those commonly used in industrial EF gasifiers. The coal properties are presented in Table 1.

2.2. Coal pyrolysis and gasification

Pyrolysis and gasification experiments were conducted using an electrically heated vertical entrained flow reactor (EFR), as described in [11], at furnace temperatures of 800 °C, 900 °C and 1000 °C. The samples were oven-dried at 60 °C for over 1 h (final moisture content ~2–3%) prior to feeding to the reactor. The particles were entrained under a total of 5.0 L/min of gas with 10% of the flow being diverted to purge the vibratory feed system, and the balance delivered directly to the reactor. A schematic of the apparatus is presented in Fig. 1.

To determine the gas flowrate required to achieve entrainment of the coal or char particles, the terminal velocity for a particle of the maximum size employed for a given experiment was compared with the total gas velocity inside the reactor under the relevant conditions. The gas flowrate was selected such that the gas velocity was equal to or greater than the terminal velocity of the maximum sized particle [12,13], therefore minimising the slip velocity and corresponding to the flow conditions in industrial gasifiers. Samples were further considered to be entrained for the entirety of each experiment based on the decrease in density and particle size which occurs as either pyrolysis or gasification proceed. The residence time for a particle of average size was 6 s.

Table 1
Properties of prepared Morwell coal.

Chemical analysis	
Moisture (% a.s.)	14.92
Ash (% d.b.)	2.04
C (% d.a.f.)	61.68
H (% d.a.f.)	4.69
N (% d.a.f.)	1.57
S (% d.a.f.)	0.87
O (by difference)	31.19
Physical analysis	
Surface area (m^2/g)	196.66

a.s. = as sampled; d.b. = dry basis; d.a.f. = dry, ash free basis.

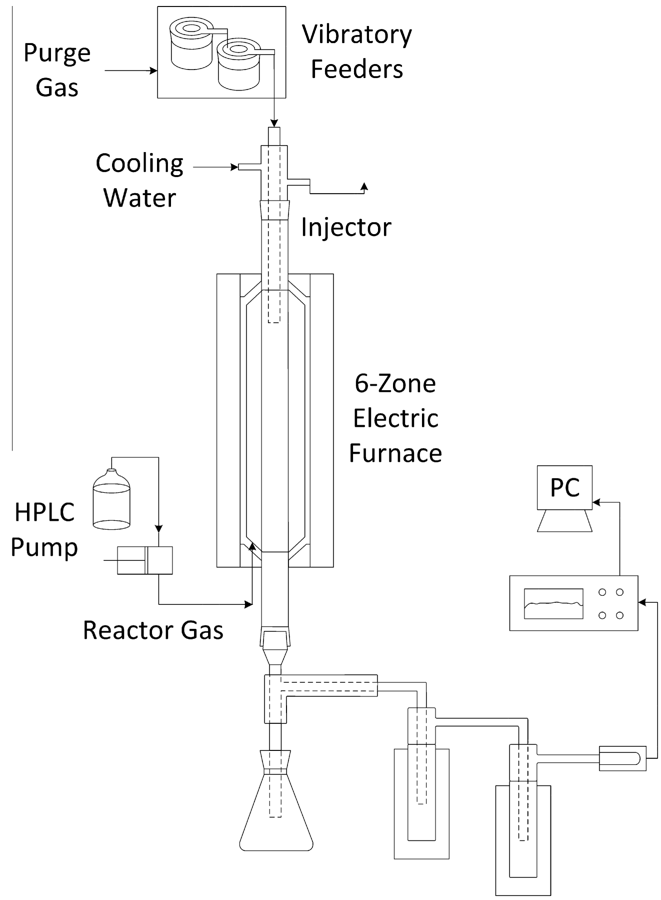

Fig. 1. Entrained flow reactor schematic.

Pyrolysis experiments were conducted under an atmosphere of 100% N_2. The atmosphere was varied from 5 vol% to 20 vol% CO_2 in N_2 for the char-CO_2 gasification experiments, all of which were performed at 1000 °C.

The solid residue from each experiment was collected and the final char conversion determined by the ash tracer method by comparison of the char ash content before and after gasification experiments [4,14]. The char samples from pyrolysis experiments were stored under dry conditions at less than 4 °C to retard surface oxidation prior to gasification and further analysis. The solid residues from gasification experiments were also preserved in this manner. The surface area of the solid samples was determined by CO_2 gas adsorption at 0 °C using a Micromeretics Accelerated Surface Area and Porosimetry (ASAP) analyser 2020 instrument.

Tar species were collected after the reactor using a series of dry impingers submerged in ice-water at 0 °C. The traps were washed out with dichloromethane (DCM) and tar analysis was performed offline using a Perkin Elmer Clarus 600 Gas Chromatograph coupled with a Clarus 600S Mass Spectrometer (GC–MS). The gas evolved from the pyrolysis and gasification experiments was analysed online using a Varian 490-GC Micro-GC equipped with Molsieve-5A and PoraPlot Q columns and a thermal conductivity detector. The gas volume generated was determined by normalisation with the known reactant gas flowrate.

3. Results and discussion

Pyrolysis and gasification of Morwell coal and char samples were carried out to validate the process of gasification of Victorian brown coals under entrained flow conditions at temperatures up to 1000 °C. Representative subsamples of the coal were