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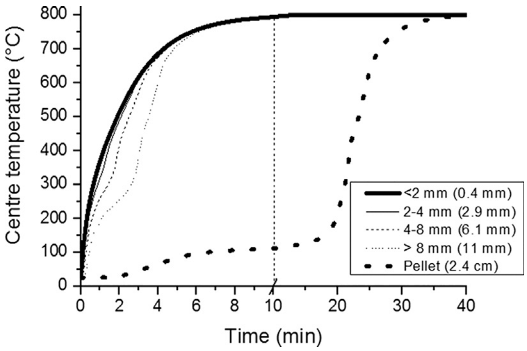
Pyrolysis of a lignite briquette – Experimental investigation and 1-dimensional modelling approach

Anthony De Girolamo[a], Vincent Tan[a], Zhenyu Liu[b], Lian Zhang[a,*]

[a] *Monash University, Wellington Road, Clayton, Victoria 3800, Australia*
[b] *State Key Laboratory of Chemical Resource Engineering, Beijing University of Chemical Technology, Beijing 10029, PR China*

GRAPHICAL ABSTRACT



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ABSTRACT

A laboratory-scale shaft furnace was used for the pyrolysis of the lignite briquette together with thermogravimetric analysis to study the intrinsic pyrolysis kinetics under two different heating rates, 10 °C/min and 100 °C/min. Apart from coal conversion rate, the tar yield, quality and radical concentration in char were also measured to explore the difference between lignite briquette and the respective pulverised powder. Additionally, a 1-D model coupled with the chemical percolation devolatilisation (CPD) code was developed to quantitatively understand heat transfer using temperature dependent parameters; product distribution and yields, and the pyrolysis mechanism. It was discovered that heat transfer was the limiting factor for pyrolysis of the lignite briquette, which subsequently lowered the heating rate and led to increased cross-linking and decreased tar production compared to a coal particle. Simultaneously, the primary tar also underwent internal cracking and even deoxygenation to decompose into light aromatics and gases inside the briquette char matrix. Providing a hot gas environment was found to facilitate the cracking of tar species compared to a slow heating rate where tar is released at a lower temperature. The changes in radical concentration in the solid material were linked to the structural changes predicted by the CPD model including bridge-breaking, tar release and cross-linking phenomena.

1. Introduction

Being the single largest source in Victoria, Australia, Victorian brown coal is a resource with vast reserves and can be extracted at a low cost [1]. Upgrading it to higher value products through pyrolysis is one of the promising ways in the carbon-constrained future. The

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[*] Corresponding author.
E-mail address: lian.zhang@monash.edu (L. Zhang).

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resultant solid char can be used in a number of applications such as its use as a low-volatile pulverised coal injection (PCI) coal replacement in the metallurgy industry, which has a market price of USD $100–150/t in 2015 [2].

The pyrolysis process induces both physical and chemical changes during the conversion to char, liquid tar and gas [3]. To date, plenty of knowledge has been achieved for the pyrolysis of VBC and other lignites, as documented in the two major monographs [1,4]. However, past research on the pyrolysis has focused on the behaviour of pulverised particles that are generally smaller than 200 μm. From the practical perspective, increasing the transportability (e.g. mechanical strength and particle size) of the pyrolysis char is crucial in the Australian context, since the produced char will be primarily exported overseas to feed the global energy and metallurgy markets. For such a purpose, a prior pelletization of fine coal particles is essential. While the previous studies have provided an understanding of the reactions within a small particle [5], little is known about the complicated inter-influences between pyrolysis reactions and physical processes (heat and mass transfer) within a coal briquette. Some research has also been carried out on the pyrolysis of Collie coal briquette in Western Australia, with a focus on the production of metallurgical reductant requiring a mechanical strength of 6.9–30 MPa and a reactivity index of 2.6–28% towards CO_2 [6,7]. However, the results are not directly applicable to Victorian brown coal which has a much higher volatile yield and lower strength. Research has also been conducted on the pyrolysis of low-rank coal – biomass/municipal solid waste plastic (MSW) blends and the briquettes (using commercial humates and molasses as the binders) [8,9]. However, efforts have yet to be made to examine the pyrolysis behaviour of the briquetted brown coal.

In this paper, we will report the pyrolysis of a Victorian brown coal briquette under the varying conditions in a fixed-bed reactor, including two particle heating rates (10 °C/min and 100 °C/min), five terminal temperatures (600–1000 °C) and hold-time (10–60 min) at 800 °C. The slow heating regime (10 °C/min) is expected to occur in a rotary kiln pyrolyser or the middle zone of an industry-scale fixed-bed pyrolyser, under which the briquette temperature is expected to be even in the radial direction, whereas for the fast heating (100 °C/min), which is expected to occur close to the furnace wall in an industry-scale pyrolyser, the pyrolysis extent could be limited by the internal heat transfer within the coarse particles or briquette. Apart from the briquette, the parent raw coal was screened to four different sizes and each size was examined for comparison. To quantify the temperature distribution and reaction controlled rates for the solid char production, a 1-D model was further established and coupled with the existing Chemical Percolation Devolatilisation (CPD) model [10] to integrate structural, temperature and pressure variations within the cylindrical coal briquette. To the best knowledge of the authors, such an effort has yet to be made in the past research where the CPD model has only been applied to smaller particles. The incorporation of the 1-D model will provide information on temperature and pressure changes within the pellet to the CPD model which is suitable for non-isothermal heating rates and incorporates a pressure term within the tar vaporisation submodel. Additionally, the resultant char was characterised for the concentration of radicals within it, which are expected to be related to the extent of the cleavage of the covalent bonds within the briquette matrix [11], as well as tar yield and quality from the briquette pyrolysis. The radical concentration has proven to be one of the effective parameters to correlate the properties of char and tar and the pyrolysis conditions [12,13].

2. Experimental methods

2.1. Properties of coal particles and briquette

The feedstock materials for pyrolysis are coal particles of varying size and a coal briquette sampled from the Latrobe Valley, Australia. The coal particles were grouped into four size bands, < 2 mm, 2–4 mm, 4–8 mm and > 8 mm with the average size in each group approximately 400 μm, 2.9 mm, 6.1 mm and 11.2 mm, respectively. The upper limit of the > 8 mm size bin is ∼16 mm and the < 2 mm size bin contains particles as small as 40 μm. The respective briquette possesses a dimension of 4.0 cm in height and 4.8 cm in diameter. The manufacturing of coal briquette is the same as that has been detailed previously [1]. In brief, the coal fine particles with a moisture content of around 10 wt% were roller pressed at a pressure larger than 100 MPa. The proximate and ultimate analysis of these samples is shown in Table 1. The different sizes of the raw coal show slight variation, in particular in the content of ash. The properties of coal briquettes are however very similar with the four coal sizes, except for ash and sulphur that are slightly higher in the briquette. This could be due to the preferential use of coal fine particles that are slightly rich in ash-forming elements for coal briquetting, as evident in the size of < 2 mm in Table 1. The coal briquette also contains a lower moisture content than the air-dried pulverised coal, possibly due to the release of the physically trapped moisture during attritioning and extrusion.

Table 1
Proximate and ultimate analysis for feed materials.

Size range	Air-dried pulverised coal particles				Briquette
	< 2 mm	2–4 mm	4–8 mm	> 8 mm	
Proximate analysis, wt%					
Moisture (ar)	13.67	14.20	14.85	12.37	9.68
Volatile (db)	54.56	55.51	52.43	56.57	55.83
Fixed carbon (db)	42.88	42.76	45.74	41.83	40.56
Ash (db)	2.55	1.72	1.83	1.60	3.61
Ultimate analysis (db), wt%					
C	62.22	62.12	61.69	62.43	62.44
H	4.51	4.69	4.81	4.68	4.87
O (by difference)	29.97	30.67	30.91	30.61	28.24
N	0.58	0.62	0.60	0.55	0.61
S	0.16	0.17	0.16	0.14	0.23

2.2. Pyrolysis conditions

Pyrolysis was conducted in a fixed-bed shaft furnace as shown in Fig. 1. A quartz reactor of 1 m length and 55 mm inner diameter was used. Argon at a flow rate of 2 L/min is used to purge the reactor for 20 min prior to heating and during the pyrolysis process to sweep both the light gasses and condensable tar gasses to the collection system. The collection system is made up of three impingers surrounded by an acetone-dry ice cooling bath, and kaowool was also placed close to the outlet of the impingers to trap liquid entrained in the gas flow. Note that, the tar deposited on the reactor wall was also weighted and collected by acetone. It was then mixed with the tar collected from the impinger train, and the mixture was analysed hereafter. Water in the crude tar collected in impingers was quantified using the Karl-Fischer titration method. The remaining fraction was identified as tar. Gasses exiting the impinger system enter a gas detector which can measure O_2, CH_4, CO, CO_2, H_2S, and SO_2 real-time.

Coal particles and briquette were pyrolyzed in two different processes which are defined as slow heating and fast heating hereafter. For slow heating, coal particles or briquette were heated with the reactor together at 10 °C/min up to a terminal temperature from 600 to 1000 °C, with increments of 100 °C. For fast heating conditions, the furnace was heated up to 800 °C first, and subsequently, the particle/briquette-laden quartz reactor was inserted into the furnace and held for a time that is varied between 10 and 60 min, with increments of 10 min. The fast heating rate was measured to be an average of approximately 100 °C/min for the environment surrounding the coal particle/briquette, and the terminal 800 °C was reached within approximately 8 min once the coal-laden reactor was injected inside the furnace. After both the heating modes, the quartz tube was removed