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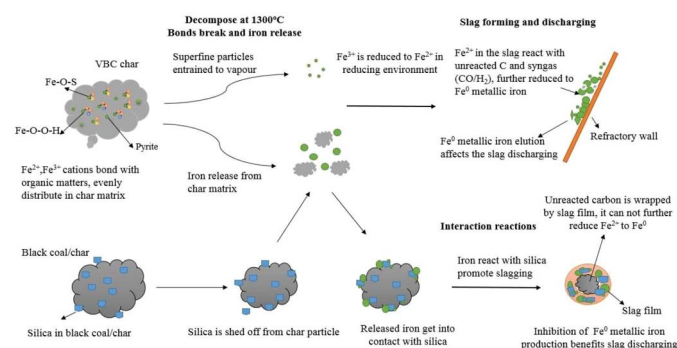
Characteristics of high temperature co-gasification and ash slagging for Victorian brown coal char and bituminous coal blends

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GRAPHICAL ABSTRACT



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ABSTRACT

The low ash and high iron content of Victorian brown coal (VBC) makes it difficult to achieve desirable slagging for entrained-flow gasification. This study investigates the blending of VBC char with a high-ash bituminous coal (BCC), and VBC char with BCC blends in three different blending ratios (20%BCC, 40%BCC and 80%BCC) are selected for high temperature gasification investigation in a lab-scale drop tube furnace (DTF) for temperatures ranging from 1000 to 1300 °C with different CO₂ volume fractions. Scanning electron microscopy (SEM) coupled with elemental mapping, advanced synchrotron XANES and Mössbauer spectroscopy (MS) are employed to clarify the slagging propensity and iron transition behaviours during the blend co-gasification. It is found that VBC char, even in large sized particles, achieves a high char conversion and more than 50% CO fraction in syngas at 1300 °C, while BCC shows a lower char conversion and only 15% CO fraction in the same condition, due to only small particles being consumed. The blending of BCC into VBC lowers the char conversion compared to that of VBC alone due to the ash interactions. However, the element (Fe/Ca/Mg) loss during gasification reduces remarkably upon the addition of BCC, showing that the silica from BCC captures these elements. In addition, VBC generates more than 20% metallic iron out of total Fe-species during gasification, and the amount increases when temperature increases, which is undesirable for slag discharge. In contrast, blending 20% BCC with VBC char reduces α-Fe from 15% to about 7%, and reduces Fe₃O₄ from 12% to less than 6%. This is because the silica in BCC can preferentially capture the iron-bearing compounds before they are reduced to metallic iron. Additionally, the unreacted carbon is covered by the slag, stopping it from reducing the Fe²⁺ to metallic iron

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Fe⁰. The reduction of metallic iron is closely linked to the increase on the ash slagging extent, which is beneficial to slag discharge in entrained-flow gasifiers.

1. Introduction

Victorian brown coal (VBC) is the single largest source of solid fuel in Victoria, Australia. With abundant resources, it dominates the supply of electricity in the state [1]. However, the high moisture content restricts VBC to be utilized domestically. The moisture content up to 70 wt% means that the transportation and storage of VBC uneconomic, and also lowers the thermal efficiency in the combustion of these brown coals [2,3]. Gasification has attracted a growing attention as a way of utilising coal because of its fuel flexibility [4]. Since current gasification technologies can adequately handle various coal ranks [5], there has recently been a renewed interest in the use of low rank coal as a fuel for gasification processes, particularly for areas with significant domestic resources. Thus, VBC as a low cost resource shows a great application potential for gasification.

Numerous studies have investigated VBC gasification in lab-scale facilities to clarify the effects of volatilisation and catalysis from alkali and alkaline earth metallic species during pyrolysis and gasification [6–9], the changes in char reactivity and structure during the gasification [10], as well as the kinetics of VBC char steam gasification [11]. However, the experiments in these studies were conducted at temperatures below 1000 °C. Compared with bituminous coal, the volatile content in VBC is higher on a dried basis, which can generate a certain amount of hydrocarbon gases and tarry vapour during low temperature gasification [12]. It not only lowers the quality of the syngas, but also affects the syngas cleaning and ash discharge once the tarry oil condenses in gasifiers with bed material [5]. From this perspective, the low temperature gasification of VBC is not desirable. Moreover, VBC is also not suited to moving bed and fluidised bed gasifiers due to its fine and soft structure. These gasifiers prefer to operate with coal lumps. By contrast, entrained-flow gasification can achieve higher cold gas efficiency, and close to 100% carbon conversion extent. This form of gasification offers a great potential for VBC utilization.

The operating temperature of entrained-flow gasification is usually up to 1600 °C, and pressures of up to 3 MPa [13]. The key concern of high temperature gasification is ash-related slagging. Coals with low ash content are usually recommended for entrained-flow gasifiers for both economic and technical reasons. An increase in coal ash content will lead to a decrease in gasification efficiency and an increase in slag production and disposal. The ash composition is also important as some components can cause accelerated deterioration of the refractory lining of the gasifier. Current refractory linings have a service life of no more than two years and their replacement is costly [14]. Components present in the slag such as SiO₂, CaO and Fe₂O₃ can penetrate deeply into the high chrome refractory and cause cracks [15]. In slagging entrained flow gasifiers, the ash flows down the gasifier wall and drains from the gasifier as molten species. Coal or coal blends selected for slagging gasifiers should thus have ash fusion temperatures below the operating temperature of the gasifier (1400–1600 °C) [5].

A typical feature of VBC ash is the high iron content, that is up to 40%, which affects not only the gasification process, but also determines the slagging propensity in some cases [16,17]. It is essential to understand the iron species transition behaviours for brown coal high temperature gasification. Iron is normally found as an inherent element in VBC [2,18]. It is well known that the iron can promote gasification reactivity, and it has been studied as a catalyst for char gasification or hydrogasification over the decades [19–21]. Iron may appear as ferrous and ferric slag, when slag is tapped or drained from an entrained flow gasifier. Under strong reducing conditions, it can even be fully reduced to metallic iron, which causes blockage for slag tapping [22]. An earlier study illustrated the iron transition during gasification at 800 °C by using Mössbauer spectroscopy. It was reported that the iron species changed drastically during gasification and was affected by reaction time as well [19]. However, these findings were from experiments conducted at a relatively low temperature, which does not represent the conditions in entrained flow gasifiers. Iron transition behaviour in ash/slag at high temperature gasification conditions has been studied from the perspective of chemical equilibrium [23,24]. A recent study clarified the behaviour of iron species in molten slag from iron-rich coal in high temperature gasification conditions between 1300 and 1550 °C [25], but this study used ash instead of coal, which ignored the role of carbon, and that may significantly affects the iron transition behaviour.

As mentioned above, it is neither efficient nor ideal to use VBC in low temperature moving bed and fluidised bed gasifiers. Moreover, the low ash content and the high ash fusion temperature make the VBC entrained-flow gasification impractical. On the other hand, some bituminous coal with high ash content cannot be used in an entrained-flow gasifier either, due to economic and efficiency concerns. To solve this problem, an integrated pyrolysis and gasification process has been developed for blending VBC with bituminous coal. A high efficient process and high gasification reactivity were achieved based on our previous research [26–28]. As a continuation of our previous works, the current paper aims to clarify the interaction between VBC and BCC during blending gasification. It is hypothesized that the ash-forming elements (Fe, Ca, Mg) in VBC will react with silica in bituminous coal to promote the ash slagging for the blends. The characteristics of VBC char and a BCC, as well as their blends (ranging from 20 to 80% blending ratios on the mass basis), have been investigated in a high temperature drop tube furnace (DTF) and a horizontal furnace (HF). The iron speciation from the samples has been characterised extensively by using scanning electron microscopy (SEM) coupled with energy dispersive X-ray (EDX), Mössbauer spectroscopy (MS) and advanced K-edge synchrotron X-ray absorption near edge structure spectroscopy (XANES). The transition route of the iron at temperature and gas atmospheres typical of entrained flow gasification has been fully investigated. The results are expected to contribute directly to the prediction of the slagging behaviour for brown coal entrained flow gasification, which is essential for the design and optimisation of industrial gasification applications for

Table 1
Proximate and ultimate analysis for coal/char samples.

	Proximate analysis				Ultimate analysis (% db)				
	Moist, %ar	FC, %db	VM, %db	Ash, %db	C	H	N	S	O ^a
VBC char	10	76.8	5.8	7.4	87.1	1.2	0.66	0.33	3.31
Bituminous coal	5.75	60.05	17.13	22.82	67.86	3.6	1.32	0.36	4.04
Bituminous coal char (BCC)	1.43	66.53	8.46	25.01	/	/	/	/	/

(ar:as received basis, db:dry basis, ^afrom differential).

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