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Research article

Effect of charcoal addition on the properties of a coke subjected to simulated blast furnace conditions

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ABSTRACT

A pilot oven produced coke with the addition of 7.5 wt% charcoal (bio-coke) and the similarly produced base blend coke were subjected to gasification and annealing under the simulated blast furnace conditions. The effect of charcoal addition on the coke properties after gasification and annealing was characterised using Raman spectroscopy, ultra-micro indentation and tensile testing. The addition of charcoal significantly increased the coke reactivity with CO_2 as a result of its increased surface area by charcoal. The addition of charcoal made no further contribution to the devolatilisation or mineral reactions of coke during annealing. The charcoal particles were well preserved after annealing at 2273 K, but preferentially consumed in the gasification. Annealing temperature had less effect on the graphitisation of the charcoal component compared to the coexisting inert maceral derived component (IMDC) and reactive maceral derived component (RMDC). The charcoal addition had no effect on the microstructure and microstrength of coexisting microtextural types during annealing and gasification. The charcoal presented a higher resistance to fracture than IMDC and RMDC under annealing conditions. The addition of charcoal did not deteriorate the coke resistance to strength degradation under the thermal load of blast furnace. However, charcoal addition caused more severe degradation by gasification as demonstrated by more reduction in the tensile strength of the bio-coke. This was attributed to the preferential solution loss reaction of the charcoal particles leaving voids which coalesced the pores in coke, thereby increasing the defects that cause stress concentration under loading.

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1. Introduction

Global warming has been a great concern as the average atmospheric CO_2 level increased by 6% in the last 10 years [1]. The steelmaking industry contributes approximately 7% of the global CO_2 emission [2]; typically, 1.8 t CO_2 is generated per tonne of liquid steel produced by the integrated blast furnace (BF)-basic oxygen furnace process, which is the major steelmaking route [2]. The production and consumption of coke contribute over 80% of the total CO_2 emissions from the ironmaking process [3,4]. A very promising approach to reduce the total CO_2 emissions from the industry is the use of biomass (charcoal) from renewable sources [5]. The partial substitution of fossil-derived metallurgical coking coals by renewable biomass can result in a net decrease in CO_2 emissions [6].

The bio-coke is defined as the coke produced with addition of biomass which incorporates plant and animal matter on the earth surface [7]. In cokemaking, biomass acts as an inert material. It does not soften or melt upon heating. Therefore, the addition of biomass to a coking coal blend will significantly change its rheological and caking properties, typically degrading the quality of produced coke [8–10]. Previous studies [11–15] found that the extent of strength degradation of bio-coke depends on the type, quantity, particle size and pre-treatment of added charcoal. Diaz et al. [11] studied the effect of biomass with size range of 80–200 μm on the fluidity development of coking blends using high temperature small-amplitude oscillatory-shear (SAOS) rheometre. They suggested that the coke strength degrades with more than 5 wt% biomass addition. Montiano et al. [13] reported that the Gieseler maximum fluidity of the industrial coal blend decreased by 50% with 3 wt% biomass addition and Coke Strength after Reaction (CSR) [16] decreased proportionately to the quantity of biomass. According to Ng et al. [14], the substitution of coal by fine charcoal had a minor effect on the cold strength of the bio-coke produced. However,

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MacPhee et al. [12] claimed that the addition of fine-sized charcoal had a stronger adverse influence on CSR than that of coarse charcoal; carefully sized fractions of coarse charcoal (−3/8 + 1/4 in.) produced much higher quality coke.

The integrity of coke is critical to BF ironmaking operations, since coke is subjected to various degradation mechanisms, including reactions with gases (CO_2 and H_2O) and heating to high temperature up to 2273 K (close to the typical raceway flame temperature). The coke is required to maintain adequate strength upon heating and reaction, so as to minimize fines generation and secure the required furnace permeability and distribution of multiphase flows (liquids, powder and gas). The property changes of metallurgical coke under simulated blast furnace operational conditions have been studied at the microstructure level [17–19]. Hilding et al. [20] reported a strong relationship between the crystallite size of BF coke carbon and measured BF temperature. Peters et al. [21] observed that the coke extracted from the BF at the tuyere level showed a clear proportional increase of IMDC compared to its feed coke, which can be explained only by more preferential consumption of the RMDC microtexture in BF. Gill and Coin [22] found that the coke extracted from the blast furnace tuyere level showed significant difference in the coke degradation depending on the coke microtextural type. Degradation of Port Kembla coke microtexture was preferential, RMDC reacted in preference to IMDC; no loss of IMDC was detected until all the RMDC was reacted; with preferential attack of the RMDC, which is the binder of IMDC, the coke structure loses its structure integrity leading to early break-up and generation of IMDC rich fines. However, the degradation of the coke used in Newcastle blast furnaces was non-preferential with respect to RMDC or IMDC. Xing et al. [17,19] studied the coke degradation under simulated BF conditions using Raman Spectroscopy and found that, with increasing temperature, the carbon structure in coke microtextures transformed from non-graphitic to graphitic microstructures, accompanied by decrease in fracture toughness; the IMDC had a stronger resistance than RMDC to deterioration. The behaviours of microtextures upon BF reactions and thermal load ultimately affect the integrity of coke. The degradation of bio-coke and the behaviour of the charcoal microtexture under blast furnace-like conditions have not been previously studied systematically.

In this study, a bio-coke with 7.5 wt% charcoal addition and a corresponding base blend coke were produced in a pilot-scale coke oven. Both cokes were subjected to conditions simulating BF gas composition-temperature profiles to 1673 K, annealing under N_2 to 2273 K or gasification with subsequent annealing at 2273 K. This paper aims to develop an understanding of the effect of charcoal addition on coke degradation under simulated BF conditions.

2. Materials and methods

2.1. Materials

The bio-coke studied in this work was produced from a medium volatile blend of moderate inertinite content coal (36.9 vol%) with addition of 7.5 wt% charcoal (biomass) and 2 wt% of coal tar pitch (CTP). The CTP was added to improve the thermoplastic and caking property of the blend during carbonization. The carbonization of the base coal blend and coal blend with charcoal was carried out in a pilot-scale moving wall coke oven [23]. The carbonization was carried out as two by two half-compartment charges; each half compartment charge was packed to a volume of 450 mm × 440 mm × 990 mm at a dry bulk density of 825 kg/m^3. The oven initial flue temperature was 1043 K and the total coking time was 20.1 h. For each compartment the charge centre temperature at the end of coking was 1300 ± 2 K. At the end of coking, the cokes were pushed and water quenched. Product coke yield from each compartment was 100 to 105 kg. The coke samples were subsequently stabilised by three drops through 4 m.

The charcoal in the bio-coke was prepared by pilot-scale retort pyrolysis of high density wood pellets (mixed Australian hardwood species) [6]. The product charcoal was crushed and screened to provide a −1.0 + 0.5 mm fraction for the coking trials. The coke samples examined in this test work were as received crushed and sized samples (−21 + 19 mm). The coke samples are identified as: Coke A (base blend coke produced without charcoal addition) and Coke B (coked with addition of 7.5 wt% charcoal and 2 wt% coal tar pitch).

The proximate analysis of cokes and charcoal was conducted following Australian Standard AS 1038.3–2000. The Coke Reactivity Index (CRI) and Coke Strength after Reaction (CSR) were determined following Australian Standard 1038.13-1990 on the specific samples provided for this study. The cold strength of coke was also measured using I-drum tumbler (130 mm internal diameter, 700 mm length). A 150 g coke sample with a particle size of −21 + 19 mm was tumbled in an I-drum tumbler rotated at 20 revolutions/min for 30 min. The tumbled sample was sieved over a 10 mm perforated plate laboratory sieve; the weight percentage of +10 mm of the original sample was reported as I-drum tumbler strength and noted as I*600. The Brunauer-Emmett-Teller (BET) surface area of cokes and charcoal was determined from gas adsorption isotherms, using N_2 as adsorptive with a Micromeritics TriStar 3000 Analyzer. Coke and charcoal samples (−1 + 0.5 mm) in this analysis were vacuum degassed for 12 h at 423 K. The ash composition of cokes and charcoal was analysed using XRF. The proximate, CSR/CRI, I*600, BET surface area and ash composition of cokes are summarised in Table 1.

2.2. Annealing and gasification of cokes under simulated blast furnace conditions

2.2.1. Annealing

The coke samples (200 g each load, particle size −21 + 19 mm) were heat-treated in a graphite furnace for 2 h at 1673, 1873, 2073, and 2273 K under N_2 atmosphere. The heating rate to the nominated treatment temperature was fixed at 25 K/min. The samples were contained in a graphite crucible, into which 1 L/min of N_2 (99.99%) was continuously blown through a graphite ducting tube with the outlet touching the bottom of the crucible. Heat treatment time was commenced from the time the furnace temperature reached the designated value. The samples without heat treatment were labelled as Coke 1 (i.e. samples Coke A1 and Coke B1 for original Cokes A and B). The four coke samples, after annealing at temperatures from 1673 to 2273 K, were labelled sequentially as:

- Coke 2 (Coke A2 and Coke B2 annealed at 1673 K)
- Coke 3 (Coke A3 and Coke B3 annealed at 1873 K)
- Coke 4 (Coke A4 and Coke B4 annealed at 2073 K)
- Coke 5 (Coke A5 and Coke B5, annealed at 2273 K)

2.2.2. Gasification

The coke samples (200 g each load, particle size −21 + 19 mm) were contained in a silicon carbide reaction vessel, reacted under conditions simulating the BF gas composition-temperature profile from 1173 to 1673 K. The gas composition-temperature profile was based on a vertical probing reported by Van der Velden et al. [24]. In the current experiments, water and hydrogen were excluded from the gas atmosphere, and the total gas flow rate was fixed at 5 L/min. The composition of the CO-CO_2 (with balance N_2) gas mixture was varied with temperature according to Fig. 1. The temperature interval between 1173 and 1273 K corresponded to the coke passage through the high temperature thermal reserve zone, and from 1373 to 1673 K approximated the coke passage through the softening-melting zone. Samples were heated from room temperature to 1173 K in a N_2 atmosphere, then the CO-CO_2-N_2 gas mixture was introduced into the reactor to start the gasification. The four coke samples after gasification to 1273, 1473 and 1673 K, were labelled sequentially as:

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