



## Research article

## A comparison of partially burnt coal chars and the implications of their properties on the blast furnace process

J.M. Steer<sup>a,\*</sup>, R. Marsh<sup>a</sup>, D. Sexton<sup>a</sup>, M. Greenslade<sup>b</sup><sup>a</sup> Cardiff University, School of Engineering, Queen's Buildings, The Parade, Cardiff CF24 3AA, United Kingdom<sup>b</sup> Tata steel UK, Port Talbot SA13 2NG, United Kingdom

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## ABSTRACT

Blast furnace coal injection is a vital part of modern ironmaking, reducing the amount of coke reductant required in the process and increasing its efficiency. However the injection of different coals or their blends, into the raceway formed by the hot blast, has technical issues due to the very short particle residence times and the limited availability of oxygen in this region. This makes complete burnout difficult and limits the range of coals suitable for this application, leading to partially burnt chars being carried out of the raceway into the blast furnace shaft and potentially into the off-gas system.

This paper explores the fate of these chars, from a range of different coals, looking at how this influences the selection for injection and the implication of these on the blast furnace. In particular, we have looked beyond the limitations of selecting coals based on proximate analysis alone by examining in more detail other physical and chemical properties and their potential effect on the process. A drop tube furnace (DTF) has been used to synthesise chars in a high heating rate environment, and although burnout and volatile loss values suggest suitability of some coals for blast furnace injection, additional problematic effects have been identified and measured such as char swelling and agglomeration which may impact the gas permeability of the furnace. A TGA/DSC has been used to measure the gasification of chars by the Boudouard reaction and compare the thermal impact of more reactive samples.

While other studies have concentrated on the combustion of injection coals to determine their suitability, this one focuses on the implications of the partially burnt chars formed by incomplete reaction in the raceway.

## 1. Introduction

The blast furnace ironmaking process uses carbon based reductants such as coke and coal in the process of reducing iron ore to iron. Coal is injected into the hot air blast which is directed into the furnace through tuyeres and plays a vital role in the process by reducing the reliance on expensive coking coals; improving the yield of iron per tonne of raw materials; and reducing environmental emissions associated with the coking process [1].

However, oxygen is rapidly consumed in the 'raceway' region, formed where the hot blast and coal are injected into the furnace. This limits the opportunity for coal to combust completely, which can result in limited burnout of the injected coals leading to partially burnt chars being carried into other regions of the furnace or out of the top as dust emissions in the off gas system where they can impact gas permeability through the coke and iron ore burden and thermal stability [2,3]. Much research work has concentrated on the importance of coal burnout reactivity but with respect to combustion [4,5], specifically Kalkreuth

et al. explained how higher volatile matter coals produce more reactive chars [6]. Work has also been carried out on the effect of those volatiles, by Ross et al. [7] and Hayhurst and Lawrence [8], who showed how the reaction environment and conditions influence the fuel particle devolatilisation behaviour especially under oxidizing conditions.

In addition to the reactivity of the coals, the physical structure and properties of the partially burnt coal chars affects the utilisation of these and is likely to impact the thermal stability of the furnace, which in turn will affect the production and cost of iron. Stubington and Linjewile described how devolatilisation can be accompanied by swelling, shrinking and fragmentation of the particles all of which will affect the behaviour and utilisation of the chars [9].

In particular, this paper looks specifically at the fate of partially burnt coal chars with respect to a blast furnace. The raceway is characterised by short residence times, high temperatures and high heating rates and these conditions influence the burnout and physical properties of the chars. This paper aims to look at the variation in these properties and the reactivity of these partially burnt chars, examining

\* Corresponding author.

E-mail address: [SteerJ1@cardiff.ac.uk](mailto:SteerJ1@cardiff.ac.uk) (J.M. Steer).

changes in the surface chemistry, investigating and considering the ways it might adversely affect the process; and to show the importance of the char gasification reaction, via the reverse Boudouard reaction, which occurs further up the shaft of the furnace.

## 2. Materials and methods

### 2.1. Materials

A range of coals, indicative of the type that might find themselves in blends for injection, were chosen ranging from the high rank semi-anthracitic LV1 to the lower rank high volatile bituminous HV1. Three particle size classifications were chosen, a typical granulated specification,  $100\% \leq 1000 \mu\text{m}$  with  $50\% \leq 250 \mu\text{m}$ ; a pulverised coal specification  $100\% \leq 106 \mu\text{m}$  and an intermediate size classification of  $100\% \leq 500 \mu\text{m}$ . The samples were milled to this specification using a TEMA™ disc mill and classified by dry sieving using the standard BS1016-109:1995.

### 2.2. Methods

The classified samples were dried at  $105^\circ\text{C}$  using BS11722:2013 until a constant weight and the volatile matter content was measured using standard BS15148:2005. Ash contents were carried out using the standard method BS 1171:2010.

A drop tube furnace (DTF) was used to characterise the devolatilisation and burnout behaviour of the coal samples at  $1100^\circ\text{C}$  in air for residence times at 35 ms to 700 ms as detailed by the authors in previous publications [10,11]. Particles were fed into the top at feed rates of 30 g/h, entrained in a laminar air flow at 20 l/min and collected at the bottom by means of a cyclone collector. The particle residence time was controlled by altering the distance of a moveable water cooled collection probe. The ash tracer method was used to calculate the burnout of the coals, sometimes referred to as the combustion efficiency.

The petrographic maceral analysis was carried out in accordance with ISO7404 by preparing a polished particulate block and carrying out a point count under reflected light microscopy to identify the different macerals present. Particle size analysis work was carried out using a Malvern Mastersizer 3000 laser diffraction particle analyser, capable of measuring between 0.01 and  $3500 \mu\text{m}$ , using a wet cell accessory with obscuration levels between 4 and 8%.

The gasification reactivity was determined by the reverse Boudouard reaction where the carbon in the char is reacted with carbon dioxide gasifying it to carbon monoxide. A Mettler-Toledo TGA/DSC 3+ was used to monitor the weight loss by first heating to  $900^\circ\text{C}$  in nitrogen and holding for 7 min to devolatilise the sample then switching to a  $\text{CO}_2$  flow rate of 100 ml/min until complete conversion was obtained. The gasification metric used to compare the reactivity of the samples was defined as  $t_{0.5}$ , the time taken in minutes to achieve 50% conversion of the sample.

A Kratos Axis Ultra DLD system was used to obtain XPS spectra using monochromatic Al X-ray source operating at 144 W. Pass energies of 160 eV were used to collect data for survey spectra, and 40 eV for the high resolution scans. The system was operated in the hybrid mode, which utilises a combination of magnetic immersion and electrostatic lenses and acquired over an area approximately  $300 \times 700 \mu\text{m}$ . A magnetically confined charge compensation system was used to minimize charging of the sample surface, and all spectra were taken with a  $90^\circ$  take off angle. A base pressure of  $\sim 1 \times 10^{-9}$  Torr (0.133  $\mu\text{Pa}$ ) was maintained during collection of the spectra. In all cases a binding energy of 284.5 eV was used for the C1s peak to account for peak shifts due to differences in sample charging.

## 3. Results and discussion

### 3.1. Variation in the gasification reactivity of chars obtained from different coals

Different types of coal have been shown to vary in the reactivity of the partially burnt char formed when the coal is not completely combusted [2,12,13]. As there is limited scope for complete combustion in the raceway region of a blast furnace, it is important to consider both the burnout and the gasification reactivity that takes place further up the shaft, as the char residue could be very important to utilisation of the injected reductant and the furnace performance.

Most coals used for blast furnace injection are milled to a pulverised size classification [14] and it is well understood that the increases in the surface area can lead to improvements in combustion reactivity and therefore utilisation of the coal in the raceway region of the furnace [15–17]. However, due to the extra cost, energy and wear on grinding equipment some ironmakers use larger granulated classifications that require less milling [18–20].

In a previous paper [10] the authors described how the process of grinding the coals affected the physical properties of the residual char, and in particular the surface chemistry, and that in terms of burnout the two effects might counteract each other to give burnouts in larger classifications of coals similar to smaller particle size classifications with higher surface areas.

Potentially large quantities of partially burnt coal chars could arise from the incomplete burnout of coal and their physical properties could have a detrimental effect on the ironmaking process. If lower reactivity chars are retained in the furnace by the burden then this could result in the accumulation of these chars in different parts of the shaft which impact the furnace thermal stability and the efficiency of iron production.

A drop tube furnace (DTF) was used to produce chars with similar properties as chars exiting the raceway in a blast furnace, so that the suitability and impact of coals with different particle size classifications could be compared to establish the impact of grinding. The high temperature and dynamic raceway environment cannot be replicated easily, but the DTF technique has been used by many researchers to mimic the short residence times, high temperatures and high heating rates as closely as possible in the laboratory environment; Li et al. compared it to a pulverised coal injection rig and found the high particle heating rate conditions ( $10^4 \text{ K/s}$ ) are comparable and useful to compare with this environment [2,21,22].

Fig. 1 compares the gasification reactivity of chars formed from different coals classified to three different sizes after a 35 ms residence time in a drop tube furnace at  $1100^\circ\text{C}$ . It is evident from the results that the coal rank impacts on the char reactivity. The higher rank LV1 and LV2 coals formed less reactive chars (longer  $t_{0.5}$  reaction times) and for these coals in particular there is a bigger difference in the reactivity between the small and large size classifications. Gibbins et al. described how coal petrography affects the thermal annealing, showing that high vitrinite coals tend to exhibit thermo-deactivation on heat treatment more than those with high inertinite content [23]. However, despite this LV2 had one of the lowest vitrinite contents, but a lower gasification reactivity than many of the coals tested and illustrates the difficulty assigning relationships that cover all coals. In comparison, for the lower rank coals the gasification reactivity is much higher (shorter  $t_{0.5}$  reaction times) and the particle size classification has less influence on the reactivity.

This has important potential implications for two reasons, the first is the effect on the reactivity of partially burnt char entering the upper furnace, as the rate of consumption of less reactive material will be slower; the second is the physical size of the particles, whose upward flow will be restricted through the furnace and might potentially block the pores and permeability of the coke burden. In turn, this could impact the distribution of heat, descent of the burden and overall stability

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