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The effects of particle grinding on the burnout and surface chemistry of coals in a drop tube furnace

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HIGHLIGHTS

- Some larger coal particle sizes had better burnouts than smaller sizes.
- More surface oxygen bonding on the chars of larger particles compared to smaller ones.
- Grinding reduced sp^2 carbon bonding correlating with better burnout at low conversion.
- Smaller particle size coals tend to swell while larger size coals tend to fragment.
- More mineral phase changes occurred in the larger size coals.

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ABSTRACT

Grinding coals to a pulverised coal specification for blast furnace injection can be costly, which is why some iron manufacturers choose a larger granulated coal size specification. However, there is a concern that these coals may have lower burnout in the raceway region so there is a technical and economic balance with coal grinding. This paper investigates how the process of grinding alters the physical properties, plus the surface chemistry, of coals and their chars formed in a drop tube furnace; it was found that in many cases the larger particle size coals gave improved combustion burnout compared to smaller sizes.

The physical properties of the chars, formed from grinding coals to different sizes, resulted in char swelling in the smaller particle sizes, compared to char fragmentation for the larger size classifications. Minerals phases associated with better coal reactivity were found to undergo higher conversion to other chemical forms with the larger size coals, suggesting a potential catalytic or synergistic contribution to their burnout. A closer look at the surface chemistry suggests that the action of grinding coals has an important effect on the surface chemistry. The XPS spectra of the chars, formed in a drop tube furnace, indicated that grinding the coals to a smaller particle size reduced the carbon–oxygen and carbon–mineral interactions compared to the larger sizes and correlated with the higher burnouts. An increasing trend was identified for the carbon sp^2 bonding with larger size and higher rank coals which correlated with their burnout at low carbon conversions; however, this did not hold at higher conversions, suggesting other factors were more dominant.

1. Introduction

Coal injection is a widely established technique used in blast furnace ironmaking, but grinding the coal into a pulverised coal specification costs energy, time, and money [1]; in a study using a Western bituminous coal (Oxbow), the U.S. department for energy found there was a 60% increase in the power required to grind it to a pulverised specification [2]. For this reason some manufacturers choose a larger granulated size specification typically in the region of 100% < 2000 μm and 20% < 75 μm to save grinding costs compared to general pulverised specification of 80% < 75 μm [3–6]. However, manufacturers using granulated coal injection risk unburnt materials carrying through the furnace into the top gas and reducing the amount of coal utilised, particularly with coals that combust less well in a blast furnace raceway, such as those with lower volatile matter content [7,8]. This paper seeks

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to investigate the effect of particle size on the coal burnout in the context for blast furnace coal injection.

Grinding coal particles to a small particle size has been found to improve their combustion by increasing the surface area, pore volume and devolatilisation [6,9,10], but can also lead to maceral and mineral segregation effects. The maceral content of a coal affects the combustibility (temperature of combustion) and the ordering is considered to be liptinite < vitrinite < inertinite [11]. Work by Morgan et al. and Yu et al. found that burnout temperatures were reduced for smaller coal particle sizes [12,13], but the reactivity of the corresponding chars was intrinsically different due to maceral segregation effects although they were from the same coal [14]. In comparison Cloke et al. discussed this effect, noting that inertinite and fusinite concentrate in the smaller size fractions because they are brittle compared with other macerals, but they also noted that there were exceptions to this due to maceral associations with the mineral matter [15].

Segregation of the mineral constituents by grinding coal also plays an important role as these represent the non-combustible portion of the coal; and due to their inherent hardness, increasing amounts can affect the grinding which in turn increases abrasion and wear to the milling equipment [11,16]. Studies have shown that although the highest ash levels tended to occur in the smallest size fraction this was not the same for all coals as their organic affinity varies [15,17].

Thermal fragmentation of coals is a significant effect related to particle size and composition, but limited work has been carried out relating to surface chemistry and drop tube furnace burnout [18]. Particles have been found to fragment extensively depending on the coal type, particle size, furnace temperature and volatile matter content. Dacombe et al. found that fragmentation increased with particle size and correlated with a decreasing compressive strength, while Senneca et al. and Zhang et al. identified the role of internal overpressure and higher inner pressure due to the volatile content of the larger particles and the contribution to fragmentation [9,19,20].

Some coal particles exhibit a swelling effect which has also been found to be related to particle size. It has been found that small particle sizes have high swelling [21] and that in high vitrinite bituminous coals a greater swelling effect occurred due to the higher content of this maceral, producing more char cenospheres [22]. However, although more porous chars are associated with the swollen chars, agglomeration has also been observed in some coals and may also influence burnout [10].

This paper aims to investigate reasons for differences in the burnout of coal samples, ground and classified to three different sieved particle sizes, <106 μm, <500 μm and <1000 μm, using a drop tube furnace. The different sizes had different physical properties and laser diffraction was used to measure the particle size distributions to investigate fragmentation and swelling effects related to burnout. In comparison to the state of the art, this work relates the coal burnout with the effect of grinding on the surface chemistry and physical properties using X-ray photoelectron spectroscopy (XPS) to identify bonding interactions that contribute to improvements in combustion.

2. Materials and methods

Five coal samples were milled using a TEMA™ disc mill and classified by dry sieving using the standard BS1016 109:1995 into the following three different ranges.

1. 100% < 106 μm
2. 100% < 500 μm
3. 100% < 1000 μm 50% < 250 μm

The classified samples were dried at 105 °C using BS11722:2013 until a constant weight and the volatile matter content was measured using standard BS15148:2009. Ash contents were carried out using the standard method BS 1171:2010. 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. The samples ranged from high rank semi-anthracitic LV1 to the lower rank high volatile bituminous HV and were chosen based on their variation in proximate analyses shown in Table 1.

A drop tube furnace (DTF) was used to characterise the devolatilisation and burnout behaviour of the coal samples at 1100 °C in air for residence times between 35 ms and 700 ms. The high heating rate and short residence times in the DTF environment closely resemble those experienced when coal is injected into the blast air of the blast furnace raceway making this a particularly relevant technique [23–26]. 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 up to a maximum path length of 1.2 m from a water cooled inlet feeder.

The ash tracer method was used to calculate the burnout of the coals, sometimes referred to as the combustion efficiency [27,28]. This method assumes that the coal ash remains conserved in the char residue in the test conditions and that no ash species are volatilised. This was tested for all the coal samples at 1100 °C. It is important to note that because the burnout figures are calculated using the ash tracer method, there is room for error propagation which can lead to repeatability issues [29]. The average standard deviation for all the drop tube furnace burnouts was 2.4%, with a range from 0.1% to 4.1%.

The burnout (%) is calculated from the ash balance of the initial content of ash in the coal (A_0) and the ash content of the residue collected post DTF (A_1).

$$\text{Burnout } (\%) = \frac{10^4(A_1 - A_0)}{A_1(100 - A_0)}$$

Scanning electron microscope (SEM) images were obtained using a FEI SEM–EDX instrument XL30 ESEM FEG at 512 × 384 resolution in back scattered and secondary electron detection modes. Particle size analysis work was carried out using a Malvern Mastersizer 3000 laser diffraction particle analyser, capable of measuring between 0.01 and 3500 μm, using a wet cell accessory with obscuration levels between 4% and 8%.

Table 1
Average analyses of different particle size coals (dried).

Coal type	Proximate analyses				Petrographic analyses			
	Volatile matter content (wt%)	Ash content (wt%)	Fixed carbon content (wt%)	Gross calorific value (MJ/kg)	Vitrinite (vol%)	Liptinite (vol%)	Inertinite (vol%)	Mineral matter (vol%)
LV1	8.2	5.8	86.0	34.6	83	1	14	2
LV2	12.5	8.6	78.9	31.9	60	0	39	1
LV3	14.4	4.7	80.9	34.2	78	1	18	3
MV	24.4	7.8	67.8	30.8	52	1	46	1
HV	33.0	6.9	60.1	31.9	71	10	17	2