

## Research article

## Investigating char agglomeration in blast furnace coal injection

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## ABSTRACT

Blast furnace iron manufacturers aim to reduce expensive coke usage through the injection of coal. This paper investigates contrasting agglomeration behaviour with a view towards optimising blast furnace operations and limiting furnace permeability issues.

A drop tube furnace (DTF) was used to investigate the performance of two coal particle size specifications that were representative of injection coal sizes: pulverised (100% < 300  $\mu\text{m}$ , 50% < 75  $\mu\text{m}$ ), and granulated (100% < 1 mm, 50% < 250  $\mu\text{m}$ ). A range of coals was subjected to DTF testing with issues arising from the injection of caking coals. Results show these coals exhibit signs of agglomeration, a potentially problematic effect concerning blast furnace permeability. Considering gasification reactivity upon leaving the blast furnace raceway, it was found that the agglomerated coal chars do not suffer from poor reactivity and are more reactive than the non-agglomerated chars. Pre-treatment through oxidation was found to be an effective means of eliminating agglomeration in the DTF as a result of the reduction in caking properties.

## 1. Introduction

Coke is a crucial ingredient in blast furnace operation, used as a principal source of both fuel and reducing agent in smelting iron ore [1]. However, due to expensive coking costs it is now commonplace for alternative reductants, primarily coal, to be injected in order to limit coke requirements. Prior to injection, coal is ground to either pulverised or granulated specification. Pulverised coal is typically 60% < 75  $\mu\text{m}$ , whilst granulated coal is coarser with top sizes of 1 to 2 mm [2]. Coal enters the blast furnace through injection lances within the tuyères causing the coal to be subjected to initial hot blast temperatures of around 1200 °C and heating rates of 10<sup>4</sup>–10<sup>6</sup> °C/s [3–5].

Aside from cost savings by reducing coke demands, coal injection provides a range of processing, economic, and environmental benefits with improved furnace operability, higher productivity, and reduced plant emissions [6,7]. Coal injection has been known to generate various furnace challenges such as reduced flame temperatures and impacts on slagging, however, one of the most problematic issues and the primary concern for this work is furnace instability as a result of lowered permeability [8–10]. As the blast furnace is a counter-current reactor, both burden descent and efficient gas ascension are vital to stable operations meaning any reduction in permeability is an issue. Following injection into the furnace, coal char particles that remain unburned after leaving the raceway region are prone to accumulating, often causing blockages and thus lowering permeability [8,11–13]. Schott

[14] explains that a key factor causing permeability issues is inefficient char gasification.

Under certain conditions, coal is prone to physical changes including swelling, fragmentation, and agglomeration [15–19] all of which will go on to impact particle reactivity. Upon the initial heating of a coal particle, caking coals are prone to developing plasticity, often occurring simultaneously with devolatilisation. With plasticity, the particle can become viscous and, as a result, there is the possibility of particles combining and resolidifying into larger particles called agglomerates [19]. Due to their now increased size, the particles have a smaller surface area available for reaction and thus are increasingly likely to leave the raceway region unreacted. Depending on the physical structure of the char leaving the raceway, an agglomerated char may be relatively unreactive, in turn increasing the likelihood of accumulation in the furnace – a factor in poor furnace permeability. Although Shampine et al. [20] determine that agglomeration has little effect on performance in typical combustors; no such conclusions have been drawn regarding effects in the blast furnace.

When studying char/agglomerate reactivity, it is important to consider the mechanisms that govern char reactions in the blast furnace. Under initial injection conditions in the raceway, coal oxidation rates are controlled by Regime III – the external diffusion of oxygen to the particle surface [21,22]. The high temperatures and relative abundance of oxygen available means that almost all oxygen transported to the surface of the particle is consumed [23]. As a result, surface area

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available for oxygen diffusion is the rate-limiting factor. Upon particles leaving the raceway and travelling into a limited oxygen/carbon-rich, lower temperature environment, the driving mechanism controlling char reactivity is Regime II – internal pore diffusion, with the physical structure of the char becoming the rate-limiting factor [21,22]. When discussing coal gasification in CO<sub>2</sub> Irfan et al. [24] state that the rate of gasification of a char particle in a high-carbon environment is governed by the accessibility of the reactant gas to the active sites located on the internal surface of the char. It is claimed that low reactivity will arise when a particle has a relative lack of large “feeder pores” thus resulting in gas diffusion in and out of the particle being driven through micropores as opposed to macro “feeder” pores. As a result of this, the physical structure of any char or agglomerate particle will impact reactivity in the blast furnace. Particles that react slowly will be susceptible to accumulating and impacting furnace stability [5].

This work aims to experimentally simulate coal injection and examine the possibility of agglomeration under blast furnace heating conditions by using a drop tube furnace (DTF). The experimental use of a DTF is common in investigating blast furnace coal injection [25–32] as a result of the high heating rates and low residence times provided that are akin to those in the blast furnace raceway region, whilst DTF temperatures of 1100 °C are suitable for replicating hot blast temperatures (900–1300 °C). Following DTF testing, the link between coal caking properties and agglomeration will be measured. Analysis of the physical structure of the agglomerated char and the gasification reactivity derived from this structure are assessed using scanning electron microscopy (SEM) and thermogravimetric analysis (TGA). In addition, heated pre-treatment is tested as a potential method of mitigating agglomeration. This study therefore extends our understanding of char agglomeration in blast furnace coal injection and the potential furnace impacts.

## 2. Material and methods

Four bituminous injection coals were chosen for analysis with the objective of including a range of volatile matter samples – one low volatile coal, two medium volatile, one high volatile. As both pulverised and granulated coal injection is common industrially, each coal sample was ground to both size specifications. A laboratory bowl mill was used to grind the raw coals prior to sieving (BS ISO 1953:2015) to the required size in accordance with industrial specifications. The specific sizes are detailed below:

Pulverised – 100% < 300 μm, 50% < 75 μm.

Granulated – 100% < 1 mm, 50% < 250 μm.

Following preparations for size, the samples were dried prior to proximate analysis (BS 17246:2010) and petrographic analysis (BS 7404-5:2009) with results shown in Table 1.

In order to simulate the injection of coal into the blast furnace, a drop tube furnace (detailed in [15,33] and shown in Fig. 1) was used. The drop tube furnace utilises high heating rates (10<sup>4</sup>–10<sup>5</sup> °C/s) and short residence times (35 ms–700 ms) that can be adjusted to resemble the initial blast furnace hot blast and raceway environments. The initial

heating of the injected coal particle is of particular importance to this work; therefore the aim was to provide a temperature similar to furnace hot blast conditions (typically 900–1300 °C [4]). As a result, an 1100 °C DTF operating temperature was selected with an air atmosphere. Coal samples were injected into the top of the 1100 °C furnace by means of a vibrating screw feeder at an addition rate of 30 g/h. The coal particles enter a nitrogen inlet gas before passing through the heated alumina work tube (1.36 m × 0.06 m) in an entrained laminar air flow (20 l/min). A particle residence time of 35 ms was selected by means of altering the length of a water-cooled collector probe to shorten the amount of time that the coal particles spend exposed to the heated furnace atmosphere. As the coal particles are entrained in the 20 l/min airflow, the distance required to set a specific residence time can be calculated via velocity of the gas flow and the desired residence time. The below equation was used where  $d$  = distance between injector probe and collector probe (cm),  $v$  = gas velocity (ms<sup>-1</sup>),  $s$  = residence time (ms), whilst a correction factor of 5 cm is applied to allow for mixing of inlet gases.

$$d = 5 + (vs)$$

The cooled probe acts to quench the coal/char reaction, before leading to a cyclone trap whereupon the resultant char is collected prior to further analysis.

The extent to which the agglomeration effect found in the drop tube furnace will be observed in the blast furnace raceway is uncertain as the higher raceway temperatures may combust the coal to the extent that agglomeration is not present in particles exiting this region. However, it is well understood that the short residence times, varying raceway size, and competing raceway reactions could present conditions where coal is only partially consumed, allowing the potential for agglomeration in these partially burnt chars.

It is important to note that, as a result of the nature of agglomeration, there is potential for repeatability issues when creating chars/quantifying char agglomeration. All DTF runs and agglomerate quantifications were carried out a minimum of twice per sample with the averages shown.

Following char collection, back-scattered and secondary electron SEM images of the raw coal samples and the post DTF chars were attained using an FEI XL30 Environmental SEM with the aim of identifying smaller examples of particle agglomeration, in addition to linking the char reactivity with the char physical structure. In order to test char reactivity in a carbon-rich environment, a Mettler-Toledo TGA/DSC was used under a CO<sub>2</sub> flow rate of 100 ml/min. Prior to TGA analysis, char samples were devolatilised under nitrogen in order to remove the impact of volatiles and test the reactivity of the remaining carbon/mineral structure. Samples of 10 mg were held at 900 °C in CO<sub>2</sub> for 420 min whereupon mass loss was measured vs. time and used in order to calculate char conversion ( $x$ ). The equation used to calculate conversion is shown below where  $m_0$  = initial sample mass,  $m$  = instantaneous mass, and  $m_{ash}$  = mass of the char ash.

$$x = \frac{m_0 - m}{m_0 - m_{ash}}$$

A commonly used gasification figure was selected in order to indicate a char's reactivity;  $t_{0.5}$  – the time in minutes taken for the chars to reach 50% conversion with a lower number signifying a more reactive char [34–36].

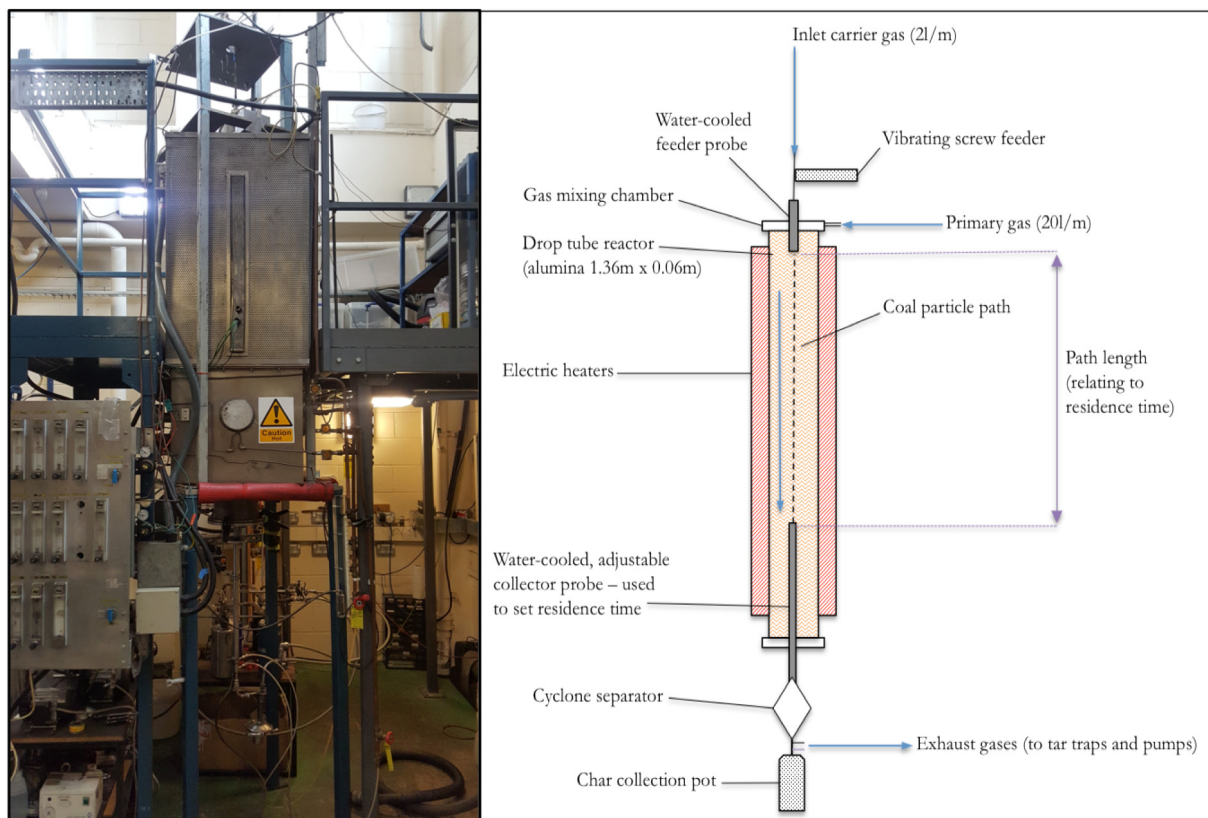
Specific surface area determinations were carried out using a Quantachrome Nova 2200e surface area and pore size analyser. 0.5 g of char sample was dried prior to vacuum degassing at 120 °C for 3 h. Following this the sample cells were analysed using BET theory with nitrogen used as the adsorbate gas. 5 specific surface area runs were collected for each sample and checked for consistency with the average used as the final specific surface area result, given in m<sup>2</sup>/g.

In order to test the impact of coal pre-heating on agglomeration, the raw coal with the strongest agglomerating tendencies was selected for

**Table 1**

Proximate and petrographic analyses of dried coal samples.

Coal	Proximate analysis			Petrographic analysis		
	Volatile content (wt%)	Fixed carbon (wt%)	Ash content (wt%)	Vitrinite (vol%)	Liptinite (vol%)	Inertinite (vol%)
LV1	9.1	79.7	11.2	83	1	14
MV4	17.6	77.2	5.2	72	6	20
MV3	20.2	70.3	9.5	78	1	20
HV1	34.5	58.3	7.2	71	10	17



**Fig. 1.** Image of drop tube furnace alongside schematic diagram. Diagram shows main inlet gases entering top of the furnace with exhaust gases leaving through collector probe. Collector probe adjustable in order to set variable residence time.

pre-treatment at 300 °C for 60 min. This was carried out under both an inert (N<sub>2</sub>) and oxidative (air) atmosphere in order to clarify whether the impacts on agglomeration are due to thermal or oxidative impacts. Initially, testing via TGA analysis was carried out (300 °C/60 min) in order to establish the extent of oxidation in air that can be seen as sample weight gain over time. Following TGA testing, larger quantities of the coal were pre-heated to create samples suitable for DTF injection. This was done in a box furnace at 300 °C for 60 min. Following this heat treatment, the caking properties of the samples were tested prior to DTF injection and agglomeration analysis.

### 3. Results and discussion

#### 3.1. Drop tube furnace agglomeration

A drop tube furnace was used in order to simulate the initial heating of injection coals in the blast furnace with key parameters being temperature, heating rate, and residence time. The DTF temperature of 1100 °C is applicable to initial hot blast temperatures whilst the short residence time of 35 ms is the estimated duration of the blast furnace raceway region. The coal chars collected at this residence time are not fully devolatilised and only partially burnt; as a result, the term char is used to describe the partially burnt residue collected from the DTF. The product coal chars were collected via water-cooled probe before being analysed for signs of agglomeration. Agglomerates were visibly present in a number of char samples with large-scale agglomeration occurring upon particle resolidification, on occasion causing gas flow issues and blockages.

In order to determine how much each coal agglomerates during DTF injection; it was necessary to calculate the char agglomeration percentages (shown in Fig. 2) with sieve classification used to do this. A 1 mm sieve was used to separate any char particles > 1 mm with these

particles categorised as agglomerates (and visually inspected to confirm). Following the separation of these > 1 mm agglomerates, the agglomerate mass as a percentage of the total char mass was calculated to give the final agglomeration figure shown in Fig. 2. Although < 1 mm agglomeration was present in the chars, quantification of these smaller agglomerates was not possible due to potential confusion between agglomerated material and individual swollen or unburned char particles.

As depicted in Fig. 2 it is clear that certain coals are prone to agglomerate when subjected to conditions comparable to initial blast furnace injection. The lowest volatile coal LV1 saw no occurrences of agglomeration in either the pulverised or granulated sample whilst MV3 saw limited agglomeration of approximately 1% in only pulverised injection. Agglomeration occurred at consistent levels in HV1 at both particle sizes with 6–8% of the total chars being made up of agglomerates, whilst MV4 saw agglomeration in both samples with almost one quarter of the pulverised char characterised by > 1 mm agglomerated particles. Agglomeration in the DTF occurs as a result of a coal's performance during devolatilisation and the plastic stage resulting in particle combinations upon collision.

Agglomeration on this scale has the potential to be detrimental to blast furnace operations, negating the positive impacts gained by grinding the coal prior to injection. As a result of the potential scale of agglomeration, this detrimental effect is likely to occur irrespective of agglomerate/char reactivity. Typically, volatile matter content is a key factor in selection of coals for blast furnace injection. These results show that volatile content is not a reliable indicator of how a coal will perform in the furnace with two similarly volatile coals (MV3 & MV4) behaving very differently with regards to agglomeration. Extensive agglomeration is likely to result in greater instances of char accumulation and blockages due to the increased size of agglomerated material. Increased occurrences of blockages will result in gas flow issues causing

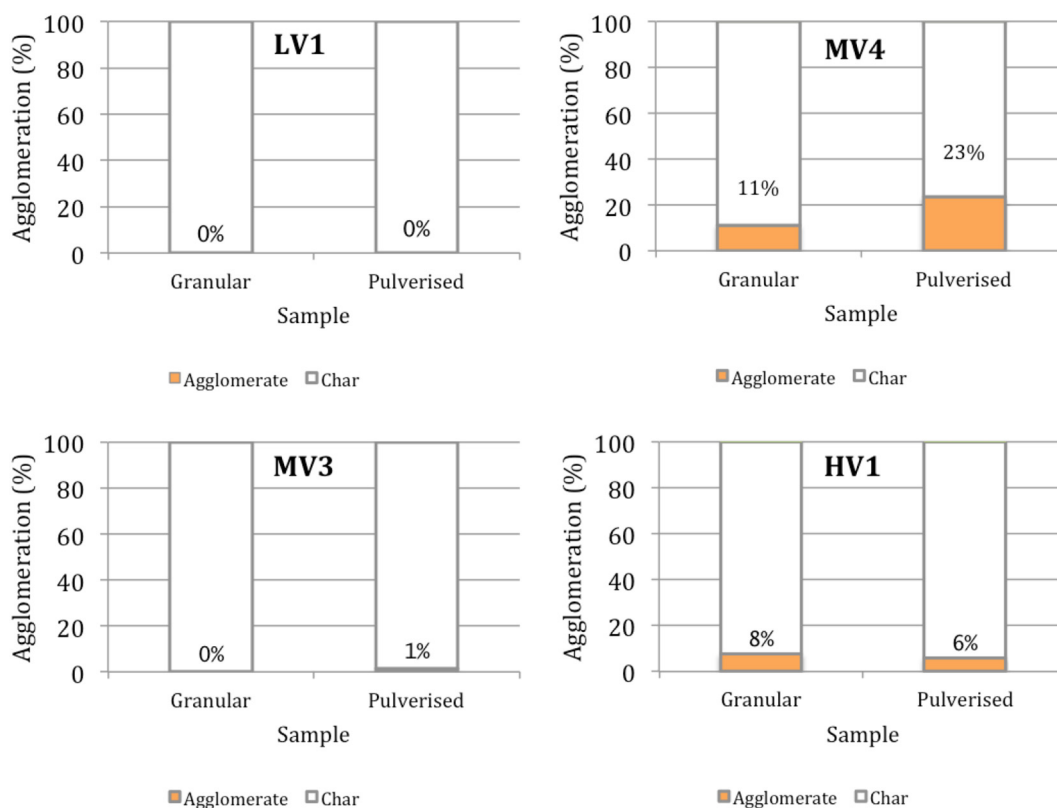


Fig. 2. Drop tube furnace 35 ms char agglomeration percentages. Chars created under DTF conditions of 1100 °C, 35 ms residence time under an air atmosphere.

temperature and pressure build-ups, instability, and an overall reduction in furnace permeability – potentially exacerbating an already problematic issue. Although the precise form of agglomeration in the blast furnace remains uncertain, these results show that the initial heating conditions found in the hot blast region are prone to resulting in coal agglomeration. Anecdotal evidence provided by blast furnace operators has shown MV4 to perform poorly in the blast furnace, impacting stability and permeability.

Although not studied in the same depth as 35 ms residence time, preliminary testing on 100 ms DTF chars does not show signs of agglomeration via sieve analysis, believed to be due to increased combustion, however, examples of fine agglomerates are present in SEM imaging.

### 3.2. Blend agglomeration

It is a common practice to blend coals prior to blast furnace injection with blends allowing for greater variability and control over the coal properties injected [32,37]. Due to the prevalence of blending in industry, the effects on agglomeration are of much relevance to this study. In order to test blend agglomeration in detail, the most susceptible coal – MV4 was used as the coal to be blended with blend ratios of 1:3, 2:2, and 3:1.

In order to establish additional confidence in the DTF agglomeration method, theoretical blend agglomeration percentages were estimated based upon the agglomeration found in the whole coal samples. Following testing, the actual blend agglomeration figures were determined and found to show strong correlation with the theoretical values ( $r = 0.89$ ) with relatively linear performance based on blend variations.

The agglomeration results for each blend tested are shown in Table 2. As is to be expected considering the previous agglomeration performances, the addition of MV4 to a blend results in an increase in the amount of DTF agglomeration. It is clear that MV4 consistently and

Table 2

35 ms DTF agglomeration percentages (shown in bold) for various MV4 blends.

Particle size	Blend ratio	MV4 100%	MV4 75%	MV4 50%	MV4 25%	MV4 0%
DTF agglomeration values						
Granulated blends	HV1 0%	<b>11</b>	–	–	–	–
	HV1 25%	–	<b>10</b>	–	–	–
	HV1 50%	–	–	<b>8</b>	–	–
	HV1 75%	–	–	–	<b>7</b>	–
	HV1 100%	–	–	–	–	<b>8</b>
	MV3 0%	<b>11</b>	–	–	–	–
	MV3 25%	–	<b>8</b>	–	–	–
	MV3 50%	–	–	<b>6</b>	–	–
	MV3 75%	–	–	–	<b>3</b>	–
	MV3 100%	–	–	–	–	<b>0</b>
Pulverised blends	LV1 0%	<b>11</b>	–	–	–	–
	LV1 25%	–	<b>5</b>	–	–	–
	LV1 50%	–	–	<b>3</b>	–	–
	LV1 75%	–	–	–	<b>0</b>	–
	LV1 100%	–	–	–	–	<b>0</b>
	MV4 100%	<b>23</b>	–	–	–	–
	MV3 0%	–	–	–	–	–
	MV3 25%	–	<b>26</b>	–	–	–
	MV3 50%	–	–	<b>16</b>	–	–
	MV3 75%	–	–	–	<b>16</b>	–
MV3 100%	–	–	–	–	<b>0</b>	
Pulverised blends	LV1 0%	<b>23</b>	–	–	–	–
	LV1 25%	–	<b>23</b>	–	–	–
	LV1 50%	–	–	<b>19</b>	–	–
	LV1 75%	–	–	–	<b>0</b>	–
	LV1 100%	–	–	–	–	<b>0</b>

reliably agglomerates in the DTF with 13 out of 15 blend variations agglomerating. The exceptions to this are the MV4:LV1 blends at 1:3 ratios at both particle sizes. It appears that only by blending MV4 with a particularly unreactive coal can agglomeration be eradicated, however,

minor blend variations can and do reduce the agglomeration effect.

The blend performance of the granulated size specification is more predictable in comparison to the pulverised blends. This is not unsurprising considering the nature of the agglomeration mechanisms being strongly linked to chance collision and combinations. As is the case for the whole coal samples, the pulverised coal blends consistently see higher levels of agglomeration. This is not likely to be a chemical effect (due to precautions taken in the grinding method), rather an impact of increased numbers of individual particles being injected at a given time. Additionally, the violent fragmentation effect that occurs during injection of larger granulated coals [15] will serve to separate individual particles during injection thus disrupting particle combinations.

### 3.3. Coal caking properties

After establishing measurable and consistent agglomeration in the DTF, it is important to consider the relevant properties producing the effect. Due to the process that occurs during the plastic stage of development with metaplast formation, particle softening and flowing, the caking properties of the coal should be tested with the aims of finding a reliable indicator of coal agglomeration. Petrographic analysis is often cited as an integral component of coal analysis and can often be used as an indicator of a coal's plastic properties. Vitrinite and liptinite macerals are generally considered to show varying levels of fluidity as opposed to inertinite, which is unlikely to plasticise [38,39]. However, regarding these coal samples, there are no strong indicators that would suggest agglomeration of one coal over another in the proximate or petrographic analyses shown in Table 1.

A test of a coal's caking properties called the agglomeration index [40,41] was used in order to test for correlation between DTF agglomeration and caking. This index was selected due to a heating rate more akin to blast furnace injection than a number of alternative procedures that were designed with coke making in mind. The test utilises a volatile matter residue as produced in BS 562:2010 with the user inspecting the residue whilst referring to set criteria including strength, swelling, and lustre in order to assign a caking score as detailed in Table 3. It is noted that samples with scores of 5, 6, and 7 are defined by the test as caking coals.

Following the creation of the volatile matter test buttons shown in Fig. 3, the residues were analysed using the criteria listed in Table 3.

Results from the index are as follows (low-high caking score):

Results from the index (shown in Table 4) correlate directly with the amounts of agglomeration found within the drop tube furnace chars for the four coals. For example, LV1 remained entirely as powder resulting in a low classification as a non-caking, non-agglomerating coal, whilst MV3 formed a weak, loosely formed button with limited swelling.

**Table 3**  
Caking test scoring criteria [40,41].

Class	Group	Appearance of residue from standard method volatile matter coal determination (BS ISO 562:2010)	Caking score
Nonagglomerating – button shows no swelling or cell structure and will not support a 500 g weight without pulverising	NA (nonagglomerate)	NAA – non coherent residue	1
		NAB – button shows no swelling or cell structure and after careful removal from the crucible will pulverise under a weight of 500 g carefully lowered on button	2
Agglomerating – button shows swelling or cell structure or will support a 500 g weight without pulverising	A (agglomerate) – button dull black and sintered, shows no swelling or cell structure; will support a 500 g weight without pulverising	Aw (weak agglomerate) - buttons come out of crucible in more than one piece	3
		Af (firm agglomerate) - buttons come out of crucible in one piece	4
		Cp (poor caking) – button shows slight swelling with small cells, has slight grey lustre	5
	C (caking) – buttons shows swelling or cell structures	Cf (fair caking) – button shows medium swelling and good cell structure, has characteristic metallic lustre	6
		Cg (good caking) – button shows strong swelling and pronounced cell structure, with numerous large cells and cavities, has characteristic metallic lustre	7

Conversely, HV1 had a relatively high index rating due to high strength, some swelling, and a strong lustre (though ultimately did not score higher due to limited swelling). Meanwhile, MV4 was awarded the highest possible caking score due to extensive swelling. The order of these result scores correlates with the amounts of agglomeration that occurred in the DTF.

In order to improve confidence and strengthen the link between caking and agglomeration, caking index scores were generated for 36 varied coal samples including single coals, blends, and pre-heated samples. Following this, the samples were injected into the DTF for 35 ms with the agglomeration per sample quantified. The caking scores were plotted against the agglomeration percentages with the results shown below in Fig. 4.

This study of caking properties and the link with DTF agglomeration shows a clear positive association with a Spearman's rank correlation coefficient of 0.85. The general trend is that with an increase in caking properties, there is a higher possibility of not only agglomeration occurring, but also the extent to which it occurs.

A clear mid point in caking/agglomeration can be seen at a caking score of 4. The index above defines all coal samples that score 5, 6, or 7 as caking coals. All 17 samples defined as caking coals consistently agglomerate in the DTF. Conversely, all 10 samples with low caking scores of 1, 2, or 3 do not agglomerate at all. A middling score of 4 shows the only level of uncertainty in DTF agglomeration. Of the samples with this caking score, 4 samples agglomerate and 5 do not. It is clear that the caking component in a coal is heavily linked to whether the sample is prone to agglomeration under high heating rate, short residence time conditions.

### 3.4. Structure of agglomerated particles post DTF

Following the establishment of agglomeration as a consistent effect, the resultant impacts should be considered. In order to do this it is prudent to explore the physical structure of the chars and how char reactivity is impacted. In order to do this, scanning electron microscope images were used to examine the prevalence of agglomeration in the chars following collection. Additionally, the link between char reactivity and physical structure can be assessed. The following SEM images show a number of char particles that are representative of the coal chars with the images taken from 35 ms granulated and pulverised chars. Large-scale > 1 mm agglomerates are removed from the chars following DTF collection to allow study of smaller examples of char agglomeration. Particular attention has been paid to the MV4 char due to the prevalence of the agglomeration effect in this coal. The SEM images are labelled as follows: I = agglomerated particle(s), II = reacted singular particle, III = physically unreacted particle, and IV = mineral matter.



Fig. 3. Caking test residues. Clockwise from top left: LV1 residue, MV3 residue, MV4 residue, HV1 residue. CM ruler included for scale.

Table 4  
Caking index results per coal.

Coal	Caking score	Coal characteristics
LV1	1 - NA, NAa	Non agglomerate, non coherent residue
MV3	4 - A, Af	Some swelling, limited cell development, weakly formed, dull button
HV1	6 - C, Cf	Some swelling and porosity, strong lustre
MV4	7 - C, Cg	Strong swelling, and pronounced interior cell structure

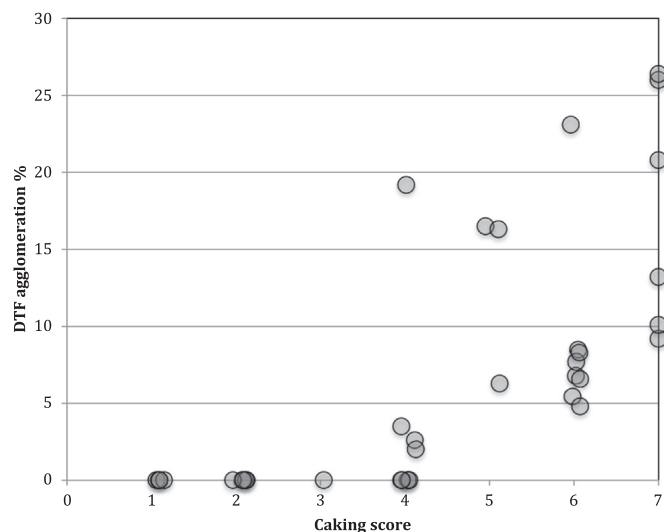


Fig. 4. Caking score vs. DTF agglomeration percentage.

Fig. 5 confirms the lack of agglomeration in the LV1 sample with mostly physically unreacted particles present (signified by III). Both granulated and pulverised samples strongly resemble milled raw coal exhibiting angular shape and solidity suggesting that these coals are particularly unreactive upon initial injection. The physical make-up of the char alone would suggest these particles would be unreactive following their transition deeper into the blast furnace, as confirmed by TGA reactivity.

The strongly agglomerated MV4 chars are shown in Fig. 6 and provide evidence of small-scale particle agglomeration (evidenced by I in the figure). Due to the thermofluidity attained by this coal upon heating, the particles have undergone significant physical change when compared to an angular, milled raw coal particle. Fig. 6(a and b) show typical examples of fine agglomeration formed of two rounded particles. The point of original contact is evident with a bridged connection between the two. The images also show examples of singular particles (denoted by II) though the volume is mostly characterised by coarse multi-particle conglomerates as evidenced by Fig. 6d. The agglomerated particles do not appear to contain many large “feeder” pores, and are not as well developed or hollow as swollen particles often are. Instead, they appear to be characterised by a surface comprised of smaller micro-pores.

The MV3 samples shown in Fig. 7 do not exhibit examples of agglomerated particles, they are characterised by singular particles at a range of sizes and pore structures. The granulated MV3 char shows a number of coarse particles that make up a large percentage of the total mass of the char shown and would likely have a sizeable impact on the char’s performance. These particles are solid and reasonably angular with very little concerning pore development. The pulverised sample contains relatively fewer coarse particles and some developed char that would theoretically improve reactivity when compared with the

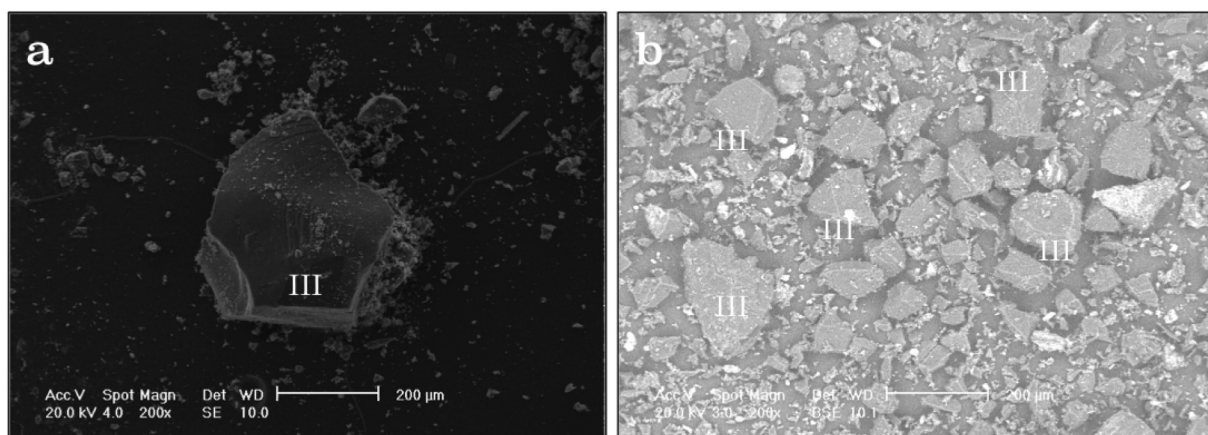


Fig. 5. SEM images of post DTF LV1 chars: a) granulated, b) pulverised.

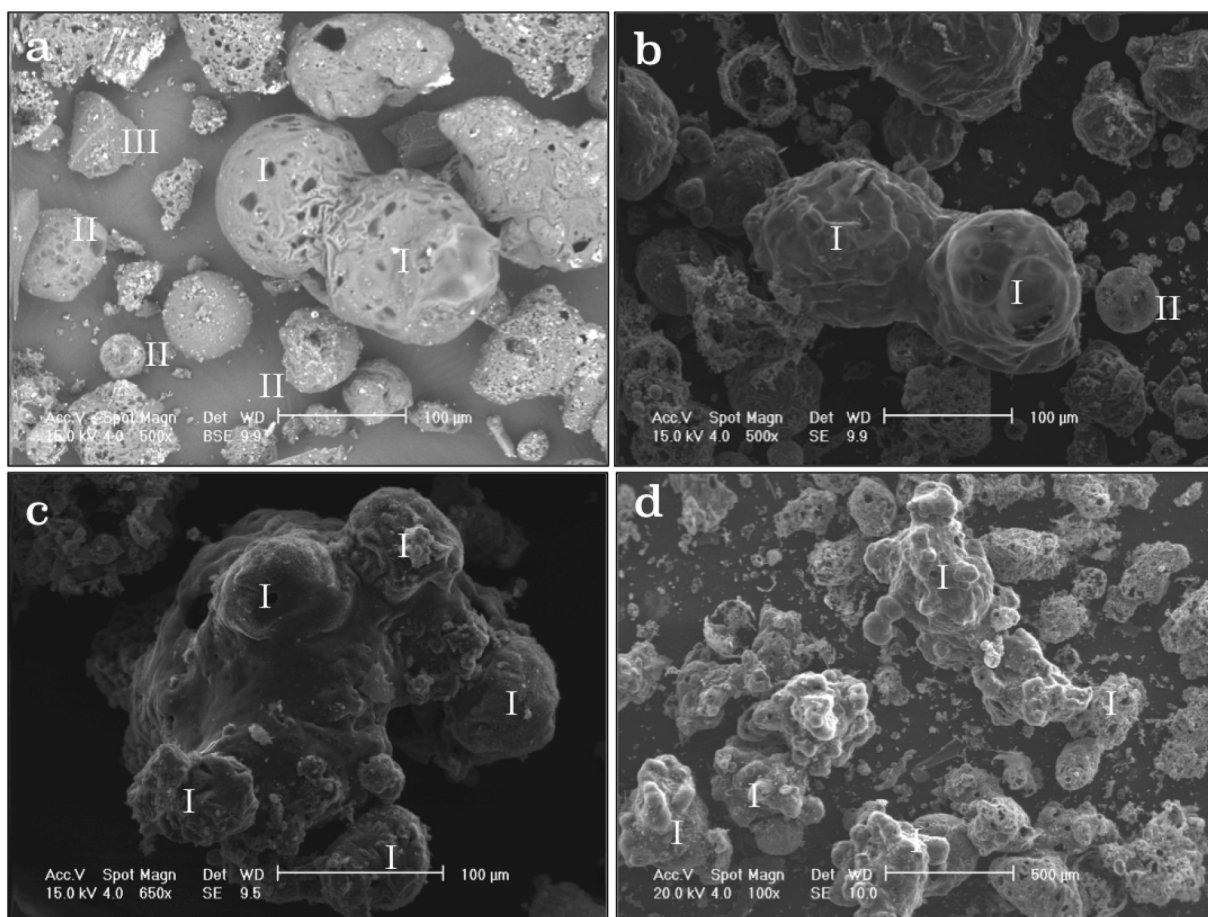


Fig. 6. SEM images of post DTF MV4 chars: a & b) granulated, c & d) pulverised.

granulated sample.

The images shown for the HV1 chars in Fig. 8 show similar char structures at both the granular and pulverised particle sizes. Examples of agglomerated material are present (I), however, when compared with those seen in the MV4 chars, the overall size and prevalence of the agglomerates appears to be lower, a factor that could improve reactivity. There are more examples of singular particles than agglomerated material in this char. The presence of large pores provide an effective means of reactant gas diffusion into the char particle, and also of product desorption out of the char. Additionally, these large pores could be used to make inferences as to the caking properties of the coal. As mentioned previously, the caking score of HV1 is lower than MV4

due to reduced swelling. Coal particles can rupture during volatile matter liberation [26], leaving large pores that can be described as “blowholes” [42]. The presence of these blowholes in the HV1 can explain the limited swelling of the HV1 caking button due to the sample effectively expelling volatile gas through these pores, an effect that may result in a collapsed particle.

### 3.5. Char gasification reactivity

When evaluating the likely impacts of injection coal agglomeration on the blast furnace, it is vital to consider char reactivity. Although there are a number of other potential impacts including furnace

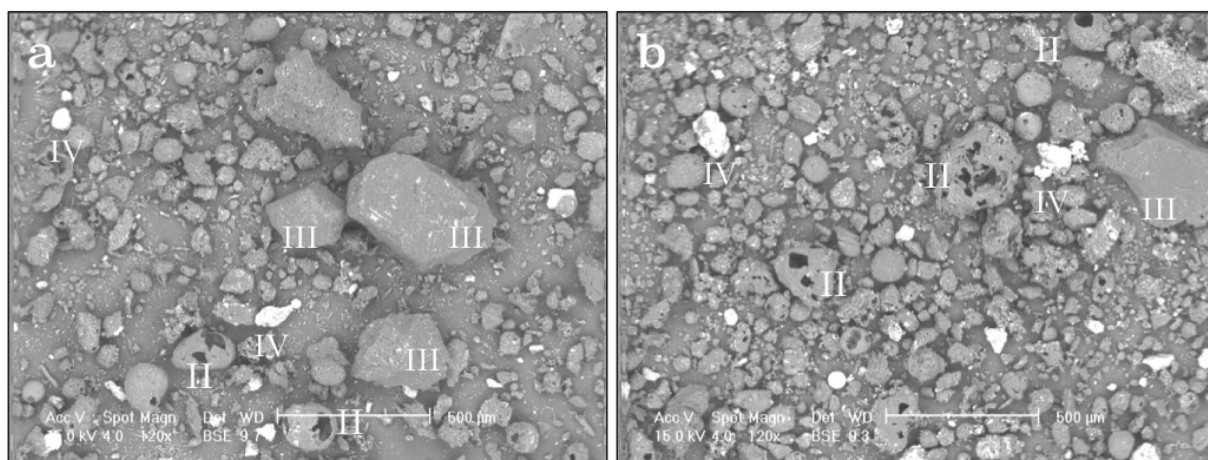


Fig. 7. SEM images of post DTF MV3 chars: a) granulated, b) pulverised.

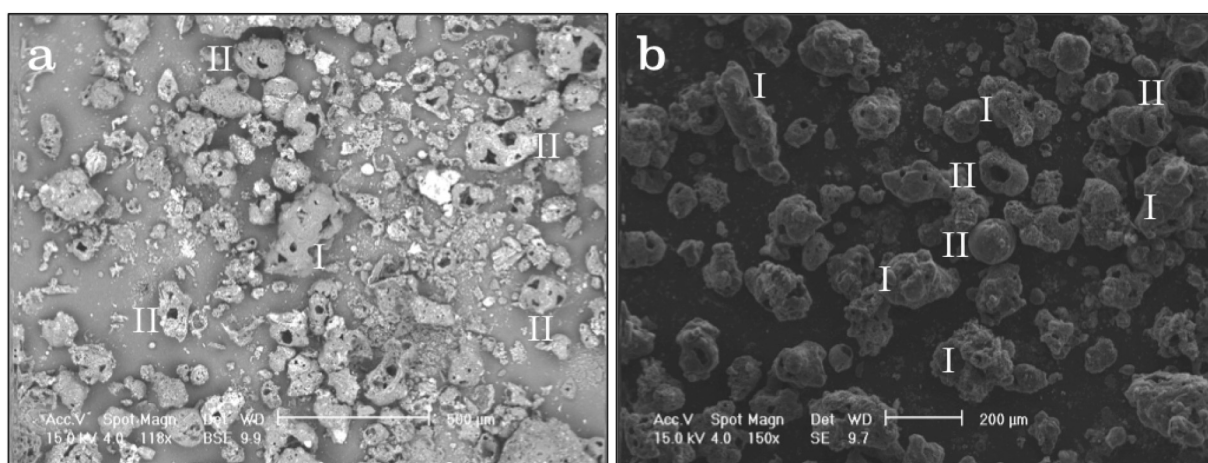


Fig. 8. SEM images of post DTF HV1 chars a) granulated, b) pulverised.

blockages, these cannot be accurately tested due to uncertainty in what form agglomeration would take in the blast furnace. Regarding gasification reactivity, as char particles leave the raceway region and enter the deeper recesses of the furnace, they are subjected to a lower temperature/carbon-rich environment. Under these conditions it is typically considered that the structure of the char particle will be integral to the resultant reactivity. A TGA was used to test the reactivity of the char with CO<sub>2</sub> via the reverse Boudouard reaction and defined as the time taken in minutes for the original sample mass to reach 50% conversion with a lower number signifying higher reactivity.

The various char  $t_{0.5}$  times are listed in Table 5 including both unground and ground chars. Due to the increased surface area available

Table 5

TGA char gasification: unground & ground chars. 900 °C under CO<sub>2</sub> atmosphere with  $t_{0.5}$  denoting time taken to reach 50% conversion.

Particle size	DTF 35 ms char	Unground char specific surface area (m <sup>2</sup> /g)	Unground char gasification time: $t_{0.5}$ (min)	Ground char gasification time: $t_{0.5}$ (min)
Granulated	HV1	17	70	58
	MV4	10	113	93
	MV3	30	166	136
	LV1	5	207	177
Pulverised	HV1	40	62	61
	MV4	40	72	61
	MV3	75	107	76
	LV1	5	214	168

for reaction, the pulverised samples are generally more reactive than their granulated counterparts per coal, reaching  $t_{0.5}$  after less time. It is notable that the agglomerated chars are generally more reactive than those that have not undergone agglomeration. There are two possible explanations for this. Firstly, the relative lack of physical change seen in the non-agglomerated chars during heating results in a solid, poorly developed structure that is more detrimental to reactivity than agglomerated materials in the char. Alternatively, the physical char structure may be of less importance than previously considered, leaving the intrinsic carbon reactivity of the char as the dominant factor in reactivity. Based on the unground  $t_{0.5}$  times, it is clear that for this range of samples, the impacts of agglomeration on gasification reactivity are not significantly detrimental. This suggests that small examples of agglomerated materials will not remain in the blast furnace for any greater time extent than standard char particles would. As a result, the effect of agglomerated chars with regard to reactivity in the blast furnace is likely to be similar to chars derived from existing coal injectants.

With the aim of further clarifying the relevance of char physical structure for these samples, the chars were ground in a mortar and pestle in order to remove any physical structure that had developed during DTF injection. Following grinding they were subjected to the same TGA gasification program with their gasification  $t_{0.5}$  times calculated and plotted alongside those of the unground chars as displayed in Fig. 9.

It is evident via the data plotted in Fig. 9 that there is a strong relationship between the char reactivity per sample irrespective of



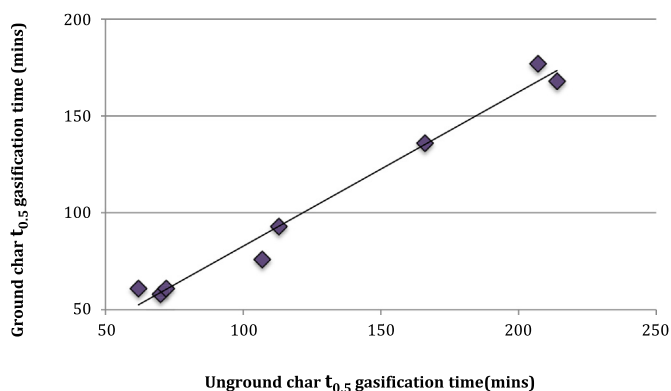


Fig. 9. Char gasification performance in TGA:  $t_{0.5}$  data for the range of chars, unground (as received from DTF) vs. ground chars.

physical structure ( $r = 0.99$ ). As would be expected, the ground chars are generally more reactive than their unground counterparts. Despite this, there are no large changes in the char order of reactivity once the physical properties of the char are removed. This suggests that for the range of samples tested, the agglomerated chars will be reactive regardless of agglomeration – the intrinsic reactivity of the char material is the principle factor behind gasification reactivity.

### 3.6. Agglomerate mitigation – pre-oxidation

The strongly agglomerating MV4 coal was subjected to pre-heating in both oxidative and inert environments in order to establish whether any effects on agglomeration are as a result of oxidative or thermal effects. Potential thermal effects include the loss of volatile matter content, whilst oxidation may lead to increased oxygen functional groups developing, both factors that can result in a loss of coal caking properties. A temperature of 300 °C was selected in order to limit volatile matter loss and ensure that the coal's plastic properties were not engaged (MV4 shows initial plasticity at approximately 420 °C via Gieseler plastometer). Prior to larger-scale testing, a TGA was used in order to test the effects of varying atmosphere at 300 °C with air and nitrogen used as oxidative and inert atmospheres respectively. TGA results (Fig. 10) show the differing impacts of pre-heating on the coal with varying atmosphere. As expected, heating in an oxidative environment results in a weight increase of approximately 2.5% due to oxygen adsorption onto the coal surface. Meanwhile, heating in an inert atmosphere sees minimal weight change with a slight decrease due to minor volatile loss (volatile matter loss for the oxidised sample hidden

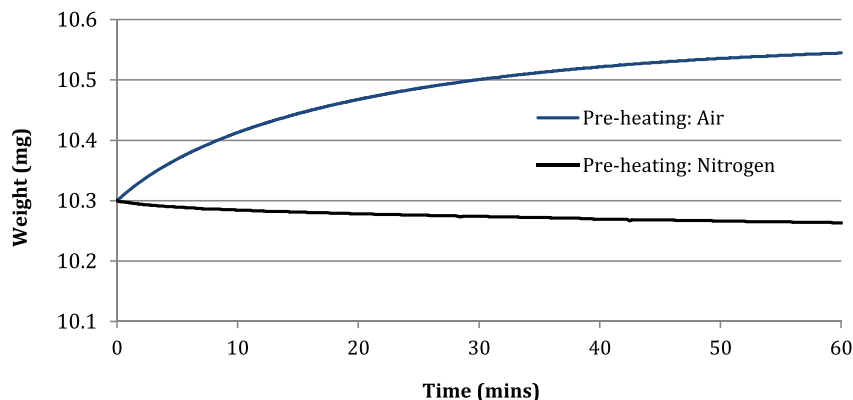


Fig. 10. TGA analysis of pre-heating program on MV4 coal.

Table 6  
Analysis of MV4 coal prior and post pre-heating.

Coal	Volatile matter (wt %)	Fixed carbon (wt %)	Ash content (wt %)	XPS oxygen % ( $O1_s$ )	Caking score
MV4	17.6	77.2	5.2	8.9	7 - C, Cg (caking)
MV4 ( $N_2$ )	16.9	77.9	5.2	n/a	7 - C, Cg (caking)
MV4 (Air)	16.9	77.8	5.3	16.1	2 - NA, NAb (non-caking)

by weight gain).

Prior to DTF injection, the pre-heated coal samples were analysed in order to clarify the impacts of heating on relevant coal properties including the effect on caking. Surface atomic oxygen percentages were found using XPS to analyse  $O1_s$  peak spectra intensity for the samples.

As shown in Table 6, volatile matter content losses are consistent in the samples whilst surface oxygen is almost doubled as a result of oxidation. The impact on the coal's caking properties highlight oxidation as the relevant factor in reducing caking tendencies with the sample heated in nitrogen seeing no reduction in caking score. Conversely, the pre-oxidised MV4 has significantly reduced caking properties with a caking score of NA, NAb due to a weak, non-swelling caking button. This is most likely a result of increased oxygen functional groups in the coal cross-linking with hydroaromatic ring structures, in turn shortening or eliminating the plastic phase of coal development. As a result of lack of plasticity, swelling and other caking parameters are limited.

With the pre-oxidised sample effectively reducing the caking properties of the MV4, the sample was injected into the DTF at a residence time of 35 ms at 1100 °C in order to test the impact on char agglomeration.

Fig. 11 depicts the amount of agglomeration found in the DTF chars for the pre-oxidised MV4 when compared with the “raw” MV4 coal. As discussed previously, the “raw” sample is characterised by prevalent agglomeration after DTF injection, both to the naked eye and in SEM images. In contrast, once MV4 has been pre-oxidised and its caking properties reduced, all agglomeration in the DTF is eliminated, including smaller-scale agglomerates as shown in the following SEM images that are characterised largely by clearly singular particles (Fig. 12). The char particles are limited to singular particles that are reasonably well rounded. This is in contrast to the standard MV4 char particles shown in Fig. 6 that are characterised by conjoined clusters of individual particles.

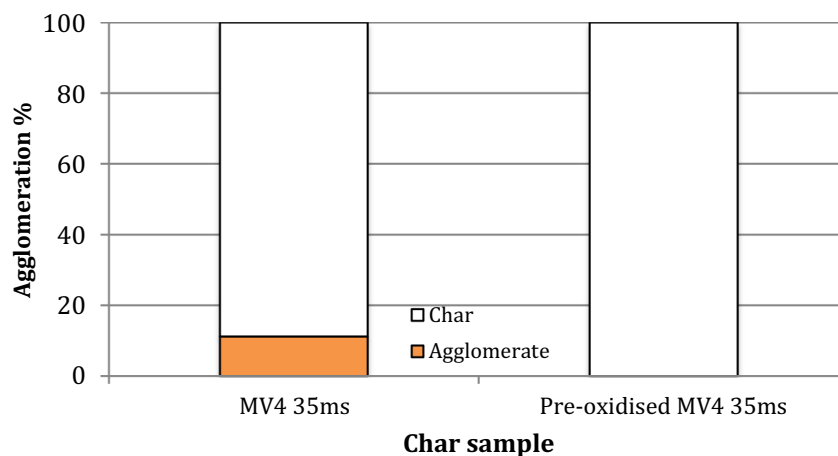


Fig. 11. DTF char agglomeration mass percentages: MV4 vs. pre-oxidised MV4.

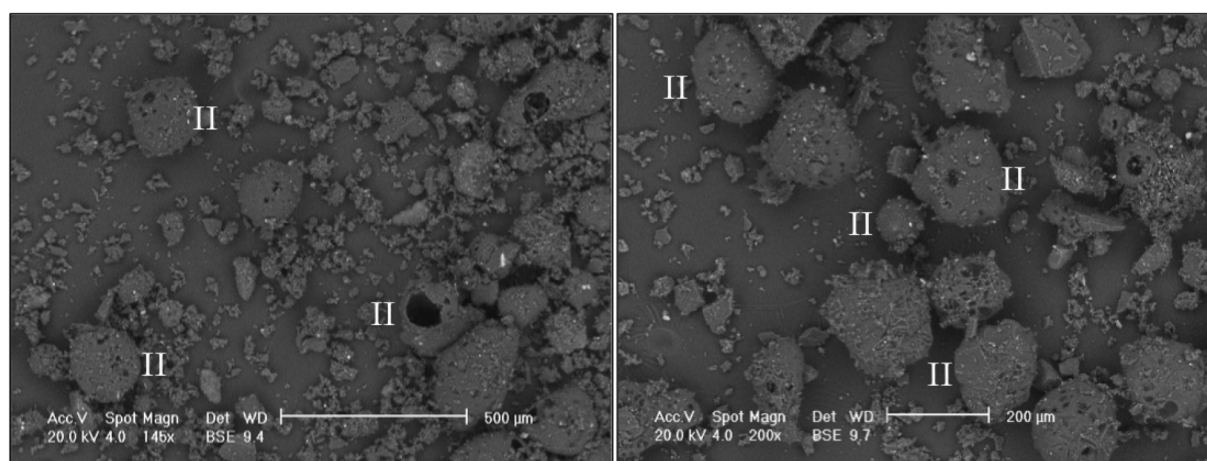


Fig. 12. SEM images of pre-oxidised MV4 DTF char (note that no agglomerated particles (I) are present in the pre-oxidised MV4 char).

#### 4. Conclusions

The results propose that coal injection under blast furnace heating conditions can be susceptible to particle combination and agglomeration. Two of the four coals tested consistently agglomerate at both granulated and pulverised size specifications. Regarding coal blends, the agglomeration effect often occurred as a factor of the parent coal's tendencies to agglomerate. The blending of coals served to effectively reduce char agglomeration with the addition of a non-agglomerating coal to the blend ratio.

Injection of a range of 36 coal samples shows a strong correlation between caking properties and DTF agglomeration. These findings suggest that coals with greater caking properties are prone to agglomerate more than non-caking coals under high heating rate conditions. The test results show that all coals defined as “caking coals” agglomerate consistently in the DTF.

With respect to the possible impacts of agglomeration, of which there are many (blockages, accumulation, permeability concerns), char reactivity was tested in order to establish how agglomerated chars behave upon leaving the raceway region, entering a carbon-rich, lower temperature environment. It was found that the chars characterised by agglomeration were, contrary to expectation, more reactive than their non-agglomerated counterparts. The effect of char grinding to remove the physical differences between the various samples was minimal, signifying that for this range of chars the inherent substance reactivity was the more dominant factor than the physical structure.

Additionally, pre-oxidation of the agglomerating MV4 coal sample

was found to effectively reduce caking properties which in turn eradicated all signs of agglomeration in the DTF chars as found by sieve analysis and SEM imaging.

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