Char Formation by Coal Injection and Its Behavior in the Blast Furnace

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1. Introduction

Blast furnace (BF) coal injection became a routine practice among European BFs; roughly, 40% of total energy required for the process is covered by auxiliary reducing agents. Pulverized coal (PC) remains the most commonly used auxiliary reductant. The key trend is increasing PC injection rates; over 200 kg tHM⁻¹ CO₁ PC on an annual basis is no rarity any more. Despite numerous measures for intensifying the coal conversion in the raceway, it is hardly possible to combust such a high amount of coal within a very short residence time of few tens of milliseconds. Recent computational fluid dynamics calculations showed that at PC injection rate of 240 kg tHM⁻¹, about 50% of the coal amount entering the raceway may leave it as so-called char.[2] Another theoretical study from 2011 calculated a maximum PC injection rate of 190–210 kg tHM⁻¹ for some BFs, considering that no ash deposition nor change in the gas flow distribution due to unburnt coal fines trapped in the coke bed takes place.[3] The following types of coal residues appear depending on different conditions and stages of its formation (Figure 1): 1) devolatilized coal particles (after light gases and tar have been released); 2) pyrolyzed (partly or completely) particles (caused by the thermal decomposition of the organic matter); and 3) not completely gasified particles (residues).

Char formation, transportation, and behavior outside the raceway may significantly affect the BF process both negatively with respect to process stability and positively by increasing the combustion efficiency by possible consumption of char. The knowledge on these phenomena was limited because the main efforts over the last few decades were focused on the complete conversion of PC within the raceway. A char morphology system was introduced for the characterization of char types.[4] However, few studies are devoted to the...
characteristics of char affecting its behavior and its possible conversion outside the raceway under the BF conditions.

This contribution focuses on the examination of char properties, synthetically produced from different coals, the extent of its transfer to the BF shaft and interaction with coke, sinter, and pellets under BF-simulating conditions. This research collaboration has developed and applied several methods and techniques for the production of synthetic char and conducted experiments using analytical, laboratory, pilot, and industrial facilities. Furthermore, a mathematical model was modified to investigate the char effect on the BF process. Findings related to the raceway measurements and monitoring are beyond the scope of this article.

2. Materials, Char Generation, and Characterization

Two coals that differ primarily in volatile matter (VM) and ash content were used (Coal 1 and Coal 2 in Table 4). Coals were ground and sieved to a particle size fraction of 90–200 μm. The compositions of coke and ferrous burden materials are given in Table 1 and 2.

To investigate the effect of conditions and stages of char formation on its properties and behavior, the synthetic char was produced under predefined conditions. These conditions were simplified compared to char generation in the BF. For this purpose, an experimental setup based on the drop tube furnace described by Ho et al. was developed. Char was produced under two different scenarios as shown in Table 3. PC is injected into the furnace’s reaction chamber and char is collected at the bottom outlet. In the first scenario, char was produced at a furnace temperature of 900 °C and in the second scenario at 1300 °C. Coal injection rate of 700 g h⁻¹ and air input of 5 L min⁻¹ were set constant. This represents a PC injection rate of ≈200 kg HM⁻¹.

Results of the char characterization along with the characteristics of parent coals are summarized in Table 4 and discussed in the following section.

2.1. Chemical Properties

Rising temperature increases the volatile yield in char. When increasing the furnace temperature from 900 to 1300 °C, the coals’ VM content decreases. Although Coal 1 contains a higher amount of VM, Coal 2 shows a higher volatile yield, especially at the higher temperatures.

2.2. Petrographic Properties

Maceral distribution analyzed by an oil immersion microscope indicated the complete disappearance of liptinite, which is characterized by the highest amount of hydrogen and VM, for all char samples. The proportion of vitrinite with the highest amount of oxygen decreases, whereas the inertinite content with a high amount of carbon increases with higher reaction temperature. The vitrinite reflectance of char increases with increasing reaction temperature as well.
2.3. Microstructural Properties

Table 5 shows the characterization of char structures defined by Bailey et al. A significant change in the microstructure was detected from coal to char. In general, char shows a porous structure, although Char 1 produced at 900 °C shows mostly the tenuinetwork or similar structure. With increasing furnace temperature, the amount of porous particle as well as their porosity increases; the wall thickness reduces, correspondingly. These changes are resulting in tenuisphere or crassispheric structures. Microstructure of both coals is similar, whereas the corresponding char microstructures vary in appearance. Thus, Char 1 is characterized by a smaller particle size compared to Char 2.

2.4. Physical Properties

The average particle size changes only slightly by initial conversion from coal to char and stays virtually unchanged with further increasing temperature. This phenomenon may be caused by development of the char microstructure. Despite the particle weight losses through devolatilization and initial conversion, a char “skeleton” remains. Figure 2. This fact is in good agreement with the development of the bulk density and specific surface area measured by MIP (mercury intrusion porosimetry) and BET (Brunauer–Emmett–Teller) methods, respectively. A significant decrease in bulk density during the transformation from coal to char was observed accompanied by an increase of the specific surface area. However, the specific surface area as well as the bulk density only showed minor change when the furnace temperature was changed from scenario 1 at 900 °C to scenario 2 at 1300 °C. Further change might occur when conversion of the char skeleton takes place.

2.5. Conversion-Related Characteristics

The activation energy of coals and chars for oxidation in air was determined by means of the Arrhenius equation, and the reaction constants were obtained by laboratory trials using a Tammann furnace setup and a thermogravimetric facility described by Born et al. The analysis of results showed that the activation energy reflects the effect of two parameters acting in opposite directions: specific surface area and VM content, as can be derived from Table 4. Due to the sharp increase in specific surface during the initial stage, the activation energy

| Table 4. Results of char characterization. |
|-------------------------------------------|
| Units | Coal 1 | Char 1900 | Char 11300 | Coal 2 | Char 2900 | Char 21300 |
| V.M. wt% | 17.20 | 11.80 | 6.60 | 13.80 | 8.90 | 3.20 |
| Cx wt% | 77.90 | 82.80 | 87.40 | 77.60 | 81.30 | 88.00 |
| Ratio Cx/VM | – | 4.53 | 7.02 | 13.24 | 5.62 | 9.13 |
| Ash wt% | 4.5 | 5.4 | 6.0 | 7.6 | 9.8 | 10.6 |
| Volatile yield % | – | 36.4 | 61.6 | – | 36.0 | 77.0 |
| Specific surface m² g⁻¹ | 2.30 | 24.20 | 28.20 | 4.20 | 25.60 | 32.30 |
| Average diameter μm | 139.0 | 125.0 | 122.0 | 121.0 | 119.0 | 120.0 |
| Bulk density g (cm³)⁻¹ | 0.76 | 0.22 | 0.28 | 0.65 | 0.36 | 0.38 |
| DTG in CO₂ wt% min⁻¹ | 1.60 | 3.49 | 4.80 | 1.52 | 1.98 | 2.54 |

Maceral distribution
- Vitrinite vol% 80 | 61 | 45 | 81 | 44 | 26 |
- Liptinite vol% 7 | 0 | 0 | 6 | 0 | 0 |
- Inertinite vol% 13 | 39 | 55 | 13 | 56 | 74 |
Reflection degree % 1.3 | 2.5 | 5.3 | 1.7 | 3.7 | 6.1 |
Activation energy kJ mol⁻¹ 36.30 | 20.51 | 31.15 | 43.82 | 35.67 | 38.18 |

*Char produced from Coal 1 according to scenario 1 in Table 3.

Table 5. Different char structures defined by Bailey et al.,[4] after the study by Yu et al.[6]
decreases significantly. Then, the specific surface is relatively unchanged, whereas the VM content continues to decrease linearly and causes in such a way, a slight increase in the activation energy.

A thermogravimetric study [derivative thermogravimetry (DTG) in CO$_2$ in Table 4] shows a higher reaction rate due to the increased specific surface.

Char formation under BF raceway-simulating conditions was examined using the MIRI (Multifunctional Injection Rig for Ironmaking) injection rig at IEHK, RWTH Aachen University, Germany and a pilot coal injection rig at CanmetENERGY in Ottawa, Canada described by Babich et al. and Wing Ng et al., respectively. For the MIRI, conversion degree of PC can be determined by several methods; in this article, it was calculated based on off-gas analysis using a nitrogen balance as follows

$$\eta_{\text{Gas}} = 100 \times \frac{\text{CO}_2 + \text{CO} + \text{CH}_4}{\phi_{\text{CO}_2}} \quad (1)$$

with

- $\eta_{\text{Gas}}$ conversion degree based on off-gas;
- CO, carbon monoxide amounts in off-gas [vol%];
- CO$_2$, carbon dioxide amounts in off-gas [vol%];
- CH$_4$, methane amounts in off-gas [vol%]; and
- $\phi_{\text{CO}_2}$, theoretical maximum amounts of CO$_2$ produced.

PC burnout for the Canmet rig was calculated from ash content of the parent coal and that of collected residues; it is defined as the ratio of mass loss of the total combustibles to the total combustibles present in the parent coal.

$$\text{Burnout} \% = \frac{(1 - A_0)}{(1 - A_1)} \times 100 \quad (2)$$

with

- $A_0 =$ mass fraction of ash in parent coal (db) and
- $A_1 =$ mass fraction of ash in residue (db).

The main results of the combustion tests performed are shown in Table 6 and Figure 3, 4, and 5 are summarized as follows: 1) the conversion degree of the investigated coals is low; 2) the oxygen enrichment of blast does not have significant effect on PC conversion; and 3) char collected from the combustion tests showed no relationship between oxygen enrichment and specific surface nor cumulative pore volume (BJH).

Analysis of the aforementioned char formation and its characteristics allow to state the following: 1) the synthetic char produced at predefined conditions shows similar tenuispheric or crassispheric structure to the char produced under
the raceway-simulating conditions using the MIRI rig (Figure 6) and can be used as an analogue of the BF char; 2) in general, depending on the rate-controlling step—oxygen diffusion or chemical reaction—coal particles can react from the surface and become smaller in size, irregular and more dense, or from the inside and become a more porous structure. Mostly, the latter combustion mechanism was confirmed: a significant change in the microstructure from coal to char was detected (microscopic analysis showed a highly porous particle). A carbon skeleton remains after devolatilization and partial combustion; 3) physical properties are changing at the initial stage of char formation, as well. However, there was no significant change in physical properties detected when comparing char generated at 900°C with char generated at 1300°C.

3. Effect of Char on Coke and Iron Burden

3.1. The Interaction of Char with Coke

The activation energy of coke (cylindrical samples of $d = 8$ mm and $h = 10$ mm) covered with 10 wt% of char was determined in an ambient air atmosphere at different isothermal conditions using a laboratory setup described by Babich et al.\cite{9} Results presented in Table 7 show that the activation energy of coke decreased significantly due to the presence of char. Thus, the starting reaction temperature of coke and, consequently, the thermal reserve zone temperature will be shifted to lower values. This might influence the thickness of the coke slits in the cohesive zone as well as the stability of the coke in the deadman area of the BF. It was confirmed by the mathematical modelling discussed in the next chapter.

A study on the interaction of coke surface with char under raceway conditions was conducted using the pilot Coke Bed Simulator (COBESI) at IEHK, RWTH Aachen University described by Born et al.\cite{2} Results showed that Coal 1 affects the gas composition in front of tuyère more than Coal 2, but no clear evidence of char impact on coke surface can be detected by means of a thermal vision camera.

3.2. The Effect of Char on the Reduction Behavior of Iron Burden

The reduction behavior of pellets was investigated using the same experimental setup as for the study on the activation energy of coke (Section 2.5), described by Babich et al.\cite{9} Pellet samples of $\approx 10$ g and size $12–13$ mm were covered with char and reduced

| Table 7. Activation energy of coke and coke with char, kJ mol\(^{-1}\). |
|-----------------|-----------------|-----------------|-----------------|-----------------|
| Coke           | Coke + Char 1,000 | Coke + Char 1,100 | Coke + Char 2,000 | Coke + Char 2,100 |
| 102.99         | 62.87           | 51.30           | 52.33           | 50.17           |

Figure 4. BET surface area of residue versus O/C, results of Canmet injection trials for Coal 2.

Figure 5. BJH cumulative micropore volume of residue versus O/C ratio, results of Canmet injection trials for Coal 2.

Figure 6. Microstructure of chars produced from Coal 2 by MIRI, SEM images.
at a temperature of 1050 °C in reducing atmosphere using CO. The reduction degree of pellets was increased significantly in the presence of char, Table 8. It may be caused by the Boudouard reaction wherein CO₂, which is produced during reduction of hematite, can be transformed to CO almost immediately since temperature was above 1000 °C. Consequently, the reduction of pellets was not limited by the transport of CO₂ from the pellet surface.

The reduction behavior of sinter was investigated using the HOSIM experimental test setup, described by Born et al.² Sinter samples of 500 g and size of 10–13 mm were coated with 1 wt% of char using a binder solution spray. These samples were reduced nonisothermally at temperatures of 430–910 °C at a heating rate of 1 °C min⁻¹ in CO/CO₂/H₂/N₂ atmosphere until O/Fe ratio of 0.5 was reached. Results showed that the presence of char influenced the mass transfer of reduction gas for reduction of sinter by physically limiting gas diffusion into the pores, resulting in rising reduction time (Table 9). The effect was higher at elevated test temperatures, at 900 °C, indicating that the temperature-dependent diffusional limitations were dominant. The char accumulation in the sinter pores was observed by macrostructural analysis (Figure 7). In this temperature range, the role of char in chemically influencing the reduction behavior was not the governing mechanism as observed by chemical and microstructural analyses.

Furthermore, the disintegration index identified as the fraction of material less than 3.15 mm after tumbling for 300 rotations was determined. No remarkable differences in the fraction of reduced sinter below 3.15 mm after tumbling can be found for samples with and without char. This result can be explained by the test temperature conditions being low for char gasification.

The opposite effect of char on the pellets and sinter reducibility described earlier is attributed to different test conditions. Pellets were reduced at temperatures above the starting temperature of solution loss reaction, wherein formed CO accelerated the reduction. Sinter was reduced at lower temperatures with unreacted char blocking the pores and hindering the reduction process. This explanation will also be confirmed in the next section.

### 3.3. Softening and Melting Behavior

Softening and melting trials of the iron burden in an inert atmosphere (100% N₂) in the temperature range 1000–1200 and 950–1200 °C with the reduction prestep in CO/N₂ atmosphere at 1000 and 950 °C were performed using metallurgical reduction facilities at Tata Steel and CRM.² It was observed that coating the iron burden materials with 1 wt% of char during the reduction step increased the softening and melting temperature of sinter and pellets. The softening and melting temperatures were raised by 30–100 and 15–50 °C, respectively, depending on the type of char and iron burden material. The reduction degree of both sinter and pellets increased as well. This confirms the statement done in previous section that the char effect on the pellets and sinter reducibility is attributed more to the test conditions than the type of iron burden or test facilities. This additional reduction was sensibly higher for sinter than for pellets due to the higher macro porosity of sinter.

Furthermore, the presence of char inhibited coalescence of pellet boundaries, and helped maintaining the interpellet void, thereby enhancing gas permeability as shown in Figure 8. However, in the base case without char, the pellet boundaries coalesced, thus mitigating gas flow.

The modification of the ferrous burden reduction degree as well as the softening and melting temperatures can result in a displacement of the cohesive zone in the BF; however, this was challenging to estimate as the test procedure and environment (reduction in a separate device and softening under nitrogen) were far from the actual BF conditions.

### Table 8. Reduction degree of pellets covered with char.

| Char type added to pellet | None | Char 1₉₀₀ | Char 1₃ₐ₀₀ | Char 2₉₀₀ | Char 2₃ₐ₀₀ |
|--------------------------|------|-----------|-----------|-----------|-----------|
| Reduction degree [%]     | 48.0 | 69.5      | 81.5      | 94.0      | 89.0      |

### Table 9. Reduction time of sinter coated with char to reach O/Fe = 0.5.

| Char type added to sinter | None | Char 1₉₀₀ | Char 1₃ₐ₀₀ | Char 2₉₀₀ | Char 2₃ₐ₀₀ |
|--------------------------|------|-----------|-----------|-----------|-----------|
| Reduction time [min]     | 185  | 190       | 194       | 195       | 204       |

Figure 7. Presence of Char 1₉₀₀ along the sinter pores and boundaries; white phase: metallic iron, light gray phase: wustite, and gray circles: char.

Figure 8. Pellet boundary after softening; left: without char and right: in the presence of char.
4. Char Transfer in the BF and Its Effect on the Cohesive Zone Characteristics and Shaft Permeability

The char is assumed to affect the BF process by two complementary mechanisms: 1) char particles are transported by high velocity gas, then accumulate in the BF where the velocity is lower, thus affecting the permeability; 2) small particles of char transported by the gas attach to coke and burden particles, where they can be trapped inside the pores, thus affecting the coke reactivity and burden properties.

4.1. Industrial Trials

Measurements at a BF with a hearth diameter of 12 m injecting about 240 kg tHM\(^{-1}\) of a mixture of hard coal and lignite (75/25% share) were performed to evaluate the extent of char transfer within the BF shaft. The results of temperature, gas composition, and pressure measurements made using a multipoints vertical probe (MPVP) described by Bailly et al.\(^{[10]}\) are presented in Figure 9. During the trial period, a charging pattern was used, which led to a marked inverted V-shape of the cohesive zone with a strong central gas flow and a low gas flow at the wall. This led to a lower reduction of burden at the furnace wall. The root (bottom) of the cohesive zone was detected less than 2.5 m above the tuyère level which is considered rather low.

The effect of high PC injection rates on the inner situation of the hearth and the coke size change between the BF top and bosh was evaluated by coke core boring and scrapping exercises at a tuyère of the same BF using an equipment and procedure described.\(^{[11]}\) The raceway coke is in general smaller than the regular coke, which is obtained through coke scrapping. This can be caused by coke degradation and combustion inside the raceway. However, raceway coke contains less fines. The analysis of coke core-drilling samples also indicated that unreduced materials (pellets) arrive below the cohesive zone and can reach the tuyère level. The conducted exercises gave indications that the raceway depth under the conditions mentioned with high PC injection rate makes up about 1.0 m and coke size decreases by half or by about 30 mm between the top and the raceway. For comparison, measurements by image-processing technique at a dissected BF with hearth diameter of 14 m operated with an average PC rate of 180 kg tHM\(^{-1}\) showed that the mean size of coke in the deadman was reduced by 36% from its original size and approximately about 33 mm.\(^{[12]}\)

4.2. Mathematical Modelling

The mathematical model MOGADOR (model for gas distribution and ore reduction)\(^{[13]}\) was modified by the implementation of char behavior in the BF shaft according to the aforementioned two complementary mechanisms. For “accumulation” mechanism, a formula calculating the static holdup of powder in a packed bed of particles\(^{[14]}\) was added to model equations, whereas for “transport” mechanism, char was assumed to follow the flow of gas in the BF. Kinetic data of char conversion were modified in the model using the experimental results presented.
in Section 2 and 3, i.e., reducibility and softening tests, coke volatilization, and gasification rate.

**Figure 10** shows the results of calculations considering that char accumulation blocks some of the burden voids and decreases the global permeability: void fraction is locally decreased and total pressure loss in BF increases. It can be seen that the char accumulates mainly in the cohesive zone, deadman, and hearth. In these zones, the gas velocity decreases, which favors particle deposition. The char was considered here as chemically inert.

Simulations considering the char reaction kinetics showed that the cohesive zone was positioned slightly higher in the BF than in the reference case without char. It is because char being more reactive than coke, can be gasified earlier via the Boudouard reaction. Pressure drop is increased due to the presence of char that fill the voids and a decrease on bed permeability. The char consumption will increase the thickness of cohesive zone, caused by the Boudouard reaction taking place in a larger area compared to the reference. The experiments, however, indicate a lower pressure drop due to the presence of char in the pellet void channels, which is not considered in the model.

**5. Conclusion**

The formation of char by coal injection and its interaction with coke and iron burden were systematically studied by the collaborative work of European leading steel producers and researchers. The following conclusions can be drawn:

1) microstructural analysis revealed that a char “skeleton” remains after coal devolatilization and initial conversion. Its physical properties such as particle size, specific surface, and bulk density hardly change with increasing reaction temperature. The activation energy of char is lower and its reactivity in CO$_2$ atmosphere is higher compared to the coal; 2) the activation energy of coke significantly decreases in the presence of char, and thus, the solution loss reaction temperature of coke might be shifted to lower value; 3) the effect of char on burden reduction is temperature-dependent: at lower temperatures around 900 °C, the presence of char influences the reduction of sinter by physically limiting gas diffusion into the pores, resulting in rising reduction time. At temperatures above 1000 °C, reduction is enhanced in the presence of char for both sinter and pellets; 4) measurements at an industrial BF operating with PC injection rate of about 240 kg tHM$^{-1}$ indicate that operation is driven by predominant gas flow through the center as compared to that at the wall. The cohesive zone has an inverted V-shape with a low position of the root and high in the center. The reduction of the burden is significantly delayed at the BF wall relative to the center; 5) the char accumulates mainly in cohesive zone, deadman, and hearth according to the mathematical simulation considering char as a chemically inert powder. Simulations considering the char reaction kinetics showed that the cohesive zone was positioned slightly higher than in the reference case without char; and 6) further work is in progress targeting a deeper understanding of the correlations between raceway characteristics, char paths through the BF, and its evolution and identification in the BF dust.

**Acknowledgements**

The authors acknowledge the European Commission for financial support of this study (Contract No. RFSR-CT-2014-00001). Open access funding enabled and organized by Projekt DEAL.
Conflict of Interest
The authors declare no conflict of interest.

Keywords
char, coke and iron burden, conversion degree, gas permeability, pulverized coal, reactivity and reducibility

Received: January 21, 2020
Revised: March 23, 2020
Published online: September 23, 2020

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