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Hazem F. Labib
University of Wollongong, hfa809@uow.edu.au

Brian J. Monaghan
University of Wollongong, monaghan@uow.edu.au

Raymond L. Longbottom
University of Wollongong, rayl@uow.edu.au

Sheng J. Chew
BlueScope Steel Research

Peter R. Austin
BlueScope Steel Research, Peter.Austin@bluescopesteel.com

FLOW OF SLAG THROUGH COKE CHANNELS AT 1500°C

Hazem F. Labib[1], Brian J. Monaghan[1], Raymond J. Longbottom[1], Sheng J. Chew[2] and Peter R. Austin[2]

[1]PYROmetallurgical Group, Faculty of Engineering, University of Wollongong,
Wollongong, NSW 2522, Australia,
hfa809@uowmail.edu.au
[2]BlueScope Steel Research,
P.O. Box 202, Port Kembla, NSW 2505, Australia.

ABSTRACT

The ironmaking blast furnace forms part of the dominant process route for steelmaking worldwide. In the lower zone of this shaft reactor, liquid iron and slag descend counter-current to reducing gases through a packed bed of coke. The fundamental characteristics of these liquid flows are not fully understood, but influence the product quality, production rate, fuel use and asset life of the process, and hence the greenhouse gas emissions and competitiveness of the integrated steel industry. The present study aimed to establish the criteria for the passage of slag through the narrow pore necks that form between coke particles.

To simulate the flow of slag through the pore necks, a new experimental technique was developed. This involved melting a slag pellet in a coke funnel with a channel of known diameter cut out of synthetic coke. Synthetic coke was used to minimise experimental uncertainty associated with the use of variable industrial coke and allow control of the coke mineralogy. Coke and slag were heated to 1500°C under argon and held at temperature for 30 minutes. After cooling, the penetration of slag into, or passage of slag through the channel was determined and the interactions of the slag with coke were characterized. Variables assessed included slag composition in the CaO-SiO_2-MgO-Al_2O_3 system, coke mineralogy and channel diameter.

For the slags and cokes studied, the minimum channel diameter that allowed slag to flow was between 4.4 and 5.0 mm. The results were in good agreement with a simple gravity and capillary force balance.

INTRODUCTION

At present, our knowledge of the ironmaking blast furnace does not extend to a full description of the material flow, chemistry and internal physical structure of the lower region. This is in part due to the highly complex inter-dependent thermal, physical and chemical phenomena within the furnace, and the lack of accessibility to the inside of the furnace to allow direct observation. High gas permeability is required for high iron production rates that characterise modern blast furnace ironmaking. In the lower zone, the permeability is significantly affected by the liquid (iron and slag) flows and their holdup. The lower zone of a blast furnace is defined to lie between the cohesive zone where ferrous materials undergo meltdown, and the hearth where liquid metal and slag accumulate before being drained from the furnace. Reducing gas, lump coke, liquid iron and slag, and fine coke and un-burnt coal co-exist in this region. An improved understanding of the liquid holdup and flow phenomena in the lower zone offers possibilities for enhanced iron production rates. The liquid flow in the lower zone of a blast furnace is often approximated to flow through a coke packed bed. Therefore, fundamental studies of liquid flows in a packed bed and the channels developed between packed coke particles should offer insights into blast furnace performance and operation.

In a coke packed bed, the voids (pores) formed between coke particles have a complex nature in terms of the way they are interconnected and the size of the connecting necks. The interconnectivity of pores and pore neck sizes determine the path of the liquid flowing through the bed and the possible sites of blockage. This is illustrated in Fig. 1, where 7 types of voids and liquid flows are indicated.

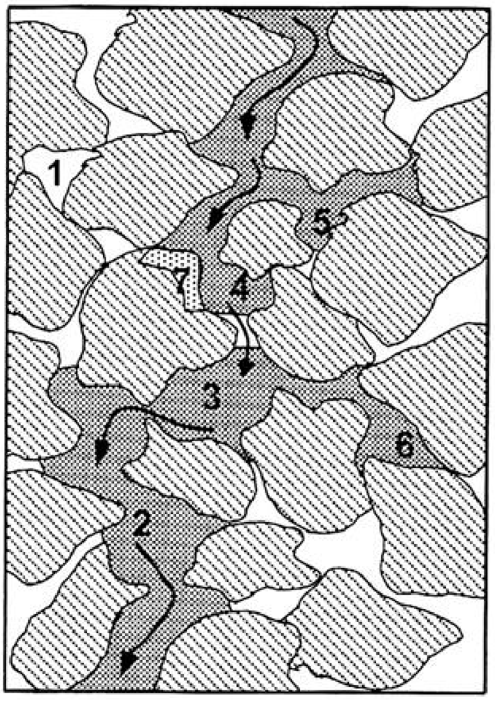

Fig. 1: Schematic representation of liquid flow through a coke bed – hatched gray: coke particles, dark gray: liquid. The numbers refer to the different sites: (1) inaccessible voids, (2) plug flow type, (3) and (4) semi batch flow, and (5) to (7) dead volume (Husslage et al., 2005)

There are few high temperature experimental studies in the literature focussed on the flow of liquid iron or slag through a packed bed (Husslage et al., 2005; Takatade, 1984). In these studies, the flow of hot metal and slag through laboratory scale coke packed beds was investigated over the temperature range 1400°C to 1660°C. The main concept in these experiments was to measure supply and drainage of slag or liquid iron through a hot coke bed so that key flow characteristics and liquid holdup could be established and related to the bed packing properties.

Results showed that the static holdup, which is the volume fraction of liquid that remains in the bed after the liquid supply has stopped, increases when smaller coke particles, and hence smaller pore neck sizes, are used to pack the bed. It was also found that beds packed with coke particles under a certain size range may lead to complete blockage of the bed by molten slag or molten metal. The reported lower limit of coke particle sizes were 8.0-10.0 mm and 4.0-6.0 mm for liquid slag and liquid metal respectively (Husslage, 2004)

In order to develop an improved understanding of liquid flow between coke particles in the lower zone of the blast furnace, the current work focused on the experimental study of the flow through narrow channels in coke. Metallurgical coke, however, is a complex material. The heterogeneous nature of coke with respect to mineral and maceral composition, dispersion and morphology, and physical characteristics such as bulk density and porosity makes it difficult to control localised properties and assess their effects in high temperature reactions in laboratory experiments (Longbottom et al., 2011). To minimise the effect of coke's inherent heterogeneity obfuscating experimental results, a synthetic coke (coke analogue) has been developed (Monaghan et al., 2010a; Monaghan et al., 2010b) and used in this study. In this coke analogue, the porosity, mineralogy and mineral phase dispersion have been controlled and characterized.

In this paper, results of the flow of molten slag through coke channels are presented and discussed.

EXPERIMENTAL

An experimental setup has been developed to simulate the flow of slag through the inter-particle voids of a coke bed. The main aim was to investigate the flow of slag of a known mass under its own weight through a narrow channel. In this experiment, slag was melted in a funnel shaped cavity that had been cut in a coke analogue sample (see Fig. 2 for a schematic of the experimental set up). The channel was of a fixed length and the channel diameter controlled. This channel approximates the inter-particle void necks of a coke bed. The analogue and the slag were heated to 1500°C in a high purity argon atmosphere in a resistance furnace. The sample (coke analogue and slag) was held at this temperature for 30 minutes then cooled to room temperature at 5°C/min. Once cooled, the sample was removed for inspection. Whether the slag flowed through the channel and any interaction/reaction the slag had with the analogue were characterized using optical and electro-optical techniques. In the cases where the slag did not flow through the channel, the depth of slag penetration through the channel was also measured. This procedure was carried out for a number of slag and coke analogue compositions and different channel diameters.

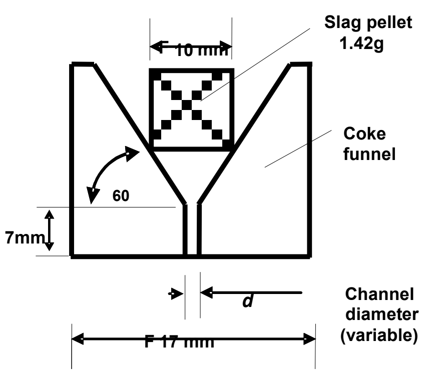

Fig. 2a: Schematic presentation of the flow of slag through a coke channel

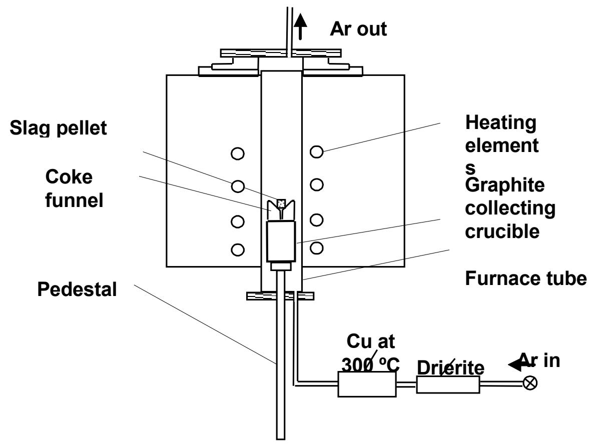

Fig. 2b: Schematic diagram of the apparatus for the dripping experiment

Materials preparation

The slag was prepared by mixing appropriate amounts of laboratory grade reagents to produce a slag of composition listed in Tab. 1. The estimated viscosities using the Riboud model (Riboud et al., 1981) and the surface tensions using the National Physical Laboratory slag model (Mills, 1991) are given. This mixture was then melted, quenched, crushed and pressed into pellets of approximately 1.42g. These pellets were then sintered at 800°C for 2 hours. The slag compositions chosen were based on what might be expected in the lower zone of a blast furnace and to exclude the effects of oxidation of the coke analogue by FeO. The slag compositions were confirmed by XRF.

Tab. 1: Slag compositions, viscosity and surface tension used in this study

Slag	Initial slag composition in Mass pct					Viscosity at 1500ºC, Pa·s (calculated (Riboud et al., 1981))	Surface tension at 1500ºC, N/m (calculated (Mills, 1991))
	CaO	SiO_2	Al_2O_3	MgO	$\frac{CaO}{SiO_2}$ $\frac{CaO}{SiO_2}$		
1	34.3	43.3	12.8	8.4	0.8	0.61	0.47
2	40.7	37.4	12.5	8.8	1.1	0.26	0.49
3	44.0	34.5	12.2	8.8	1.3	0.18	0.50
4	46.7	31.0	12.9	8.5	1.5	0.13	0.52

The coke analogue was prepared using laboratory grade crystalline and amorphous carbon forms mixed with the required weight percentage of mineral matter using a carbonaceous binding material, then pressed and fired. Full details of the coke analogue preparation procedure can be found elsewhere (Chapman, 2008; Longbottom et al., 2011) . Details of the coke analogue used in this study are given in Tab. 2. Coke analogue funnels were machined to the dimensions shown in Fig. 2a, then the exit channels were drilled through to the required diameters in 0.5 mm increments. The channel diameters were verified by optical microscopy. The range of channel diameters tested was from 1.5 mm to the maximum value of 5.0 mm.

Tab. 2: Coke Analogue compositions used in this study

Designation	Mineral type	Minerals Content, Mass %
COKAN	No minerals	0.0%
COKAN-CA1(4.4)	$CaO.Al_2O_3$	4.4%
COKAN-CA2(4.4)	$CaO.2Al_2O_3$	4.4%
COKAN-CA6(4.4)	$CaO.6Al_2O_3$	4.4%
COKAN-CA6(12)	$CaO.6Al_2O_3$	12.0%

RESULTS AND DISCUSSION

For the slag and coke analogue compositions utilized in the current work, it was found that slags flowed through coke channels of diameters of either 4.4 mm or 5.0 mm as illustrated in Fig. 3. In the case of channel diameters less than these values, the slag penetrated the channel to different depths but did not flow out through the channel. The flow of Slag 2 through COKAN-CA1(4.4) and of Slag 4 through COKAN-CA1(4.4) experiments were repeated 2 times and 3 times, respectively, and the minimum channel diameter was confirmed each time. These values were in agreement with Husslage who found that similar slags were blocked when passing through coke beds with average pore neck size below 5.38 mm (Husslage, 2004).

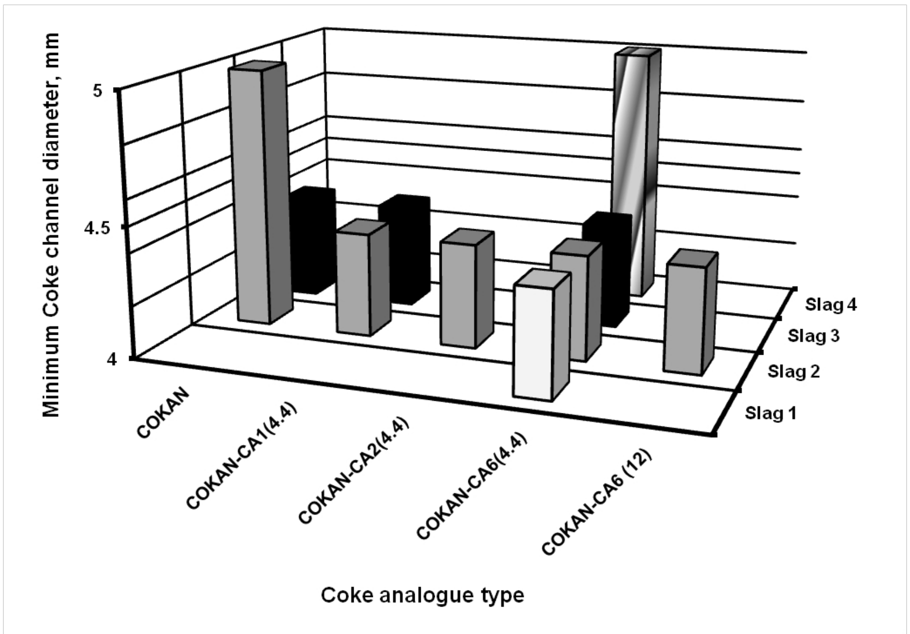

Fig. 3: Minimum coke channel diameters that allowed slag to flow through coke for different combinations of slag and coke types

In the cases where slag did not flow out, penetration depth through the channel varied between 0.5 mm and 7 mm for the different combinations of slag and coke types used.

Observations showed that for funnels of the same coke and slag combinations, greater slag penetration through the channel occurred with increasing channel diameter. Also, for a given coke type and channel diameter, there was greater slag penetration into the channel at lower surface tension. Further study is required to confirm and characterize the relationship between slag properties and the depth of channel penetration.

The interface between slag and coke was examined by SEM and mapped using EDS for elements Ca, Si, Al, and Mg. A typical micrograph and SEM-EDS analysis for elements Ca and Si at the interface is shown in Fig. 4. Patches of traces of Si are seen in the matrix of COKAN-CA1 which is attributed to cross contamination originated from the slag during sample grinding and polishing. It was found that slag had only penetrated the surface open pores of the coke analogue. There was little evidence of reaction or deeper penetration of the slag into the coke.

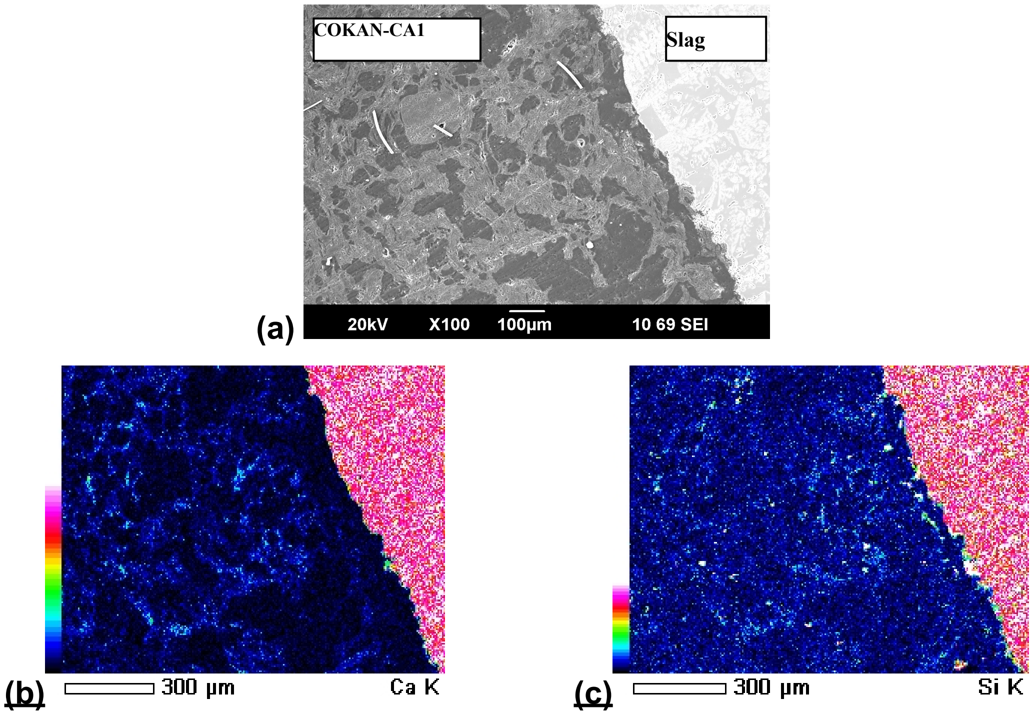

Fig. 4: Micrograph of SEM (a), and SEM-EDS mapping for Calcium (b) and Silicon (c) at the interface between slag 3 and COKAN-CA1(4.4)

Simple force balance model

There are six major forces that may act on liquid flowing through a packed bed in general as described by Fukutake and Rajakumar (1982). These forces are gravitational, inertial, viscous, surface, solid-liquid interface and that exerted by the gas flowing through the bed. However, a simplified force balance model describing the forces acting on a liquid phase statically suspended at the top of a pore neck can be built using Fukutake's description by eliminating the forces related to motion and gas drag, leaving only gravity and capillary (solid-liquid interfacial) forces. Such a force balance was used by Husslage (2004), as given in Eqn. 1:

$$\rho g h = -\frac{4\sigma \cos\theta}{d} \qquad (1)$$

where, ρ is slag density, *g* is gravitational acceleration, *σ* is the surface tension of the slag, *θ* is the contact angle between slag and coke analogue, *d* is the channel diameter and *h* is the molten slag head over the opening tip (about 12 mm in the experimental setup used). This equation is based on the force balance between the weight of the liquid slag and the capillary repulsion force between slag and coke as illustrated in Fig. 5. The bigger the channel diameter, the smaller is the repulsion force that prevents the slag from flowing through the channel. At a certain channel diameter, the liquid slag hydrostatic pressure overcomes the capillary pressure and the slag starts to flow through the channel. This process is analogous to the case of applying the Young (1805) equation to the process of infiltration of liquids into porous media where the porosity is assumed cylindrical and open (Eustathopoulos et al., 1999).

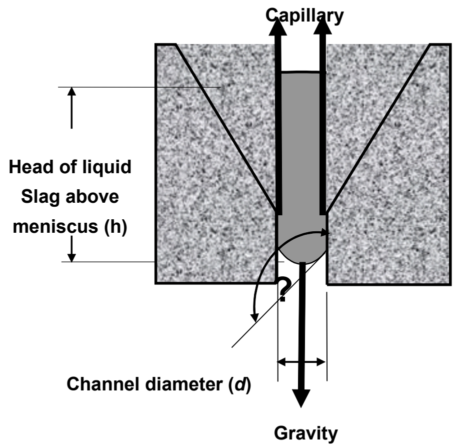

Fig. 5: Schematic illustration of the simplified force balance acting on statically suspended liquid slag above a coke channel.

Using characteristic values of the surface tension and density of the slags used in this study at the experimental temperature, and the contact angle between slag and coke from literature, the channel diameter *d* from Eqn. 1 can be calculated. See Tab. 1 and 3 for the values used. Values obtained ranged from 2.6 to 5.4 mm corresponding to the minimum and maximum contact angles used. The critical *d* values measured in this study (Fig. 3) lie within this range.

Tab. 3: Characteristic values for slag properties and contact angle with coke at 1500°C that have been used in Eqn. 1 to calculate channel diameter

Sample	Slag density ρ, kg/m^3 (Mills, 1991)	Contact angle between slag and coke θ,° (Husslage, 2004)
Slag 1	2646	Between 100 and 140 on different industrial cokes and graphite
Slag 2	2675	
Slag 3	2686	
Slag 4	2697	

The simplified force balance assumes a static slag. However, once the slag starts to flow, viscous and other surface forces will be active, and the general Fukutake analysis applies. Both Fukutake's general description and the simplified force balance model (Eqn. 1) suggest that an increase in slag surface tension will increase the repulsion force and hence the required channel diameter for slag flow. However, an effect for slag viscosity would only be expected under Fukutake's flow conditions where slag velocity component is active, and not in the static experimental condition.

To test the effects of both slag surface tension and viscosity, the minimum channel diameter required to allow slag flow was plotted against these parameters in Figs. 6 and 7, respectively. The effect of slag surface tension was consistent with both Fukutake's general description and the simplified force balance model as the slag with the highest value of surface tension had the largest minimum channel diameter for slag flow. The slag viscosity effect, shown in Fig. 7, indicates that a slag with higher viscosity could flow through the smallest channel diameter. There is no fundamental basis for this and the result is taken to indicate that viscosity is not a significant parameter in the flow experiments. This is as expected from the static force balance conditions of the reported experiments (Eqn. 1).

The results for minimum channel diameter indicate that the simplified force model (Eqn. 1) provides a good description of the system and could be used as the basis of a predictive model for establishing minimum criteria for flow in a packed bed or other pore network.

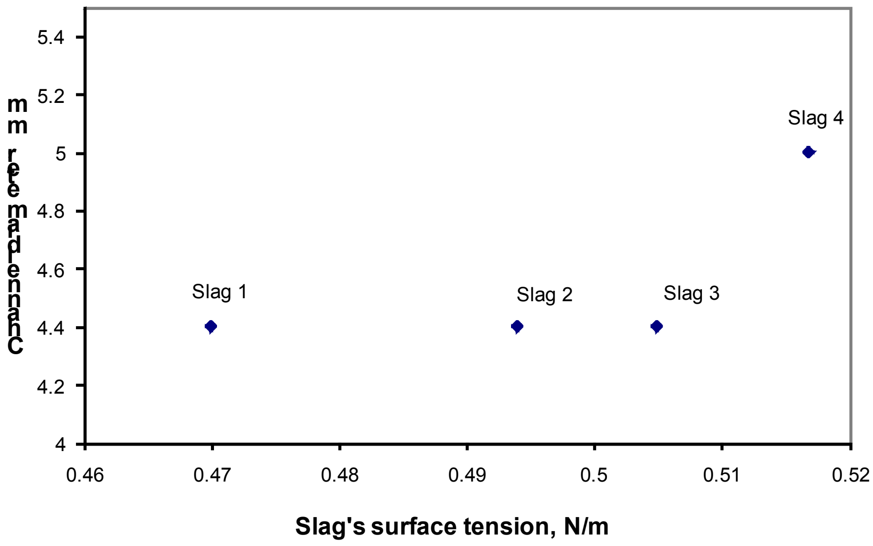

Fig. 6: Effect of slag surface tension on the minimum channel diameter for flow through COKAN-CA6(4.4)

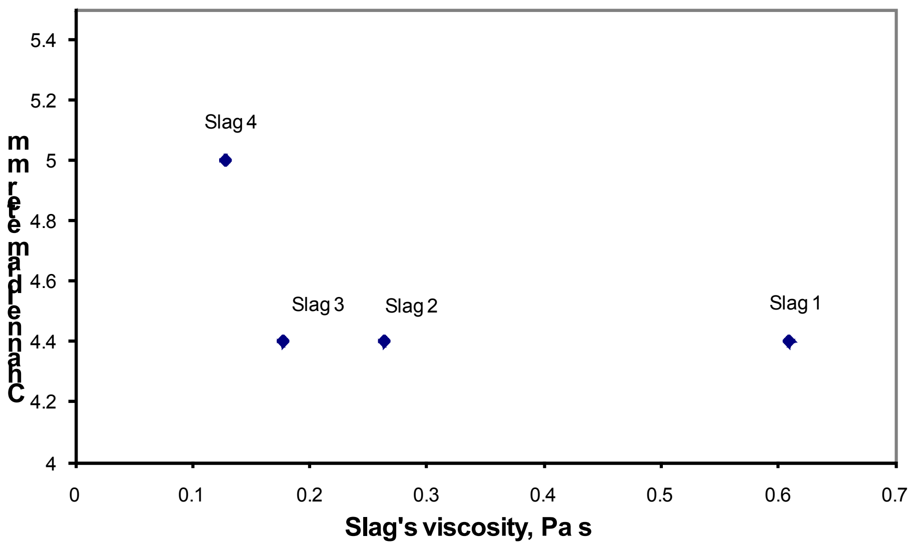

Fig. 7: Effect of slag viscosity on the minimum channel diameter for flow through COKAN-CA6(4.4)

The mineral matter content in coke can affect the interfacial properties with slag. To investigate this influence, the minimum channel diameter for flow of a single slag type (Slag 2) was determined for different mineral phases at total mineral phase contents of 0, 4.4 and 12.0 weight percentage in coke (Fig. 8). For this system, it was observed that mineral addition decreased the required diameter for the slag to flow. However, further investigations are required to clarify the influence of mineral matter type and amount.

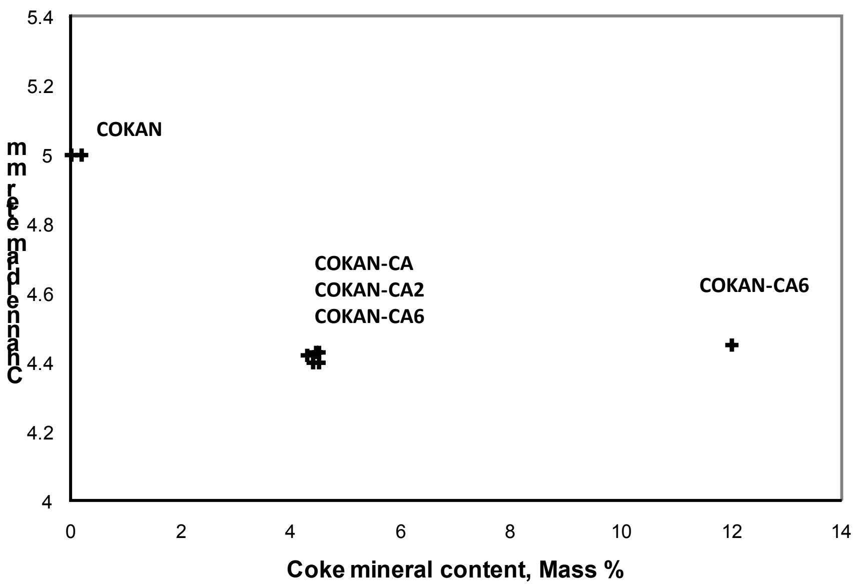

Fig. 8: Effect of mineral matter fraction in coke on the minimum channel diameter required for flow of Slag 2

CONCLUSIONS

Experiments simulating the passage of slag through a narrow pore neck between coke particles in the lower zone of an ironmaking blast furnace revealed that there is a minimum opening needed to allow slag to freely flow. In this work, the minimum opening, as represented by a cylindrical channel through synthetic coke (coke analogue), was either 4.4 or 5.0 mm for the combinations of slags and cokes examined. The results were in good agreement with previously reported figures of pore neck diameter that led to blockage of similar liquid slags. A simple force balance analysis based on gravity and capillary forces, where the slag surface tension plays a dominant role in the resistance to slag flow through the channel, was found to adequately describe the system. It is thought that this approach could form the basis of a predictive model for establishing flow conditions through a coke packed bed subject to further characterisation of the influence of coke and slag properties.

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BRIEF BIOGRAPHY OF PRESENTER

Hazem Labib came to the University of Wollongong as a PhD student after nearly twenty years of work in the steel industry. After graduating from Alexandria University, Egypt as a mechanical engineer, he worked as a design and quality engineer, then as a sales manager for Steel Products. In 1997, he obtained a Masters degree in materials science and engineering from Imperial College, London. He is a member of the PYROmetallurgical group at UOW and his research work is focused on Ironmaking.