Blast Furnace Control using Slag Viscosities and Liquidus Temperatures with Phase Equilibria Calculations

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Abstract – The viscosity of molten slag in blast furnace operations is an important process variable, influencing the rate and location of reactions inside the furnace, as well as the ease with which the final slag could be removed from the furnace. To optimize these processes, it is necessary to estimate the slag viscosity and liquidus temperatures as accurately as possible, and know the target values. Models to predict viscosities and liquidus temperatures, incorporating phase equilibrium calculations, have been developed and validated with measured data. Various diagrams have been developed to illustrate how the properties vary with composition and temperature, including pseudo-ternary diagrams of SiO_2-MgO-CaO-Al_2O_3 for viscosity and liquidus temperature. Models were also applied and methodologies developed to analyse historical blast furnace operations and determine target values.

INTRODUCTION

Physicochemical properties of slag, such as viscosity, are important process variables of the blast furnace process, and of significance when studying the process and applying the knowledge in management and optimization. Slag viscosity is a transport property that relates to the reaction kinetics and the degree of reduction of the final slag[1]. Slag viscosity also determines the slag–metal separation efficiency, and subsequently the metal yield and impurity removal capacity. In operation, the slag viscosity is indicative of the ease with which slag could be tapped from the furnace, and therefore relates to the energy requirement and profitability of the process.

The ability to predict the slag viscosity and liquidus temperature has the potential to optimize the analysis and decision-making control of blast furnaces, replacing the use of rules of thumb pertaining to slag compositions. Efforts have been made in the past to measure and model viscosities for different slag systems, of which the results of many can be found in published literature[1,2]. Some of these models correlate the effect of the different components very well,

but often do not consider the effect of the adjusted liquid slag composition, and precipitation of solids at lower temperatures.

In the typical slag system of interest, an increase in basicity not only leads to a lower liquid viscosity due to broken silicate bonds, but also increases the likelihood of solids precipitation, thereby increasing the viscosity. Phase equilibrium calculations are used to firstly determine the liquidus temperature, and then to estimate the amount (if any) of precipitated solids. For multiphase slags, the predicted amount of solids is to be used to adjust the viscosity predicted by the liquid viscosity model.

To optimize the process using the developed model, the first step would be to apply the models to historical data to generate a performance baseline and determine targets for improvement. In this work, the aim was to develop liquidus temperature and effective-viscosity models and application methodologies, to illustrate how they could be used for a blast furnace, to monitor performance and drive optimization.

METHODOLOGY

Overview of slag viscosity and liquidus temperature

Molten slag can be classified as a Newtonian fluid with the shear viscosity being independent of the shear rate, and therefore named dynamic viscosity[1]. Viscosity is largely influenced by bonding and the degree of polymerisation[2], with SiO_2 and Al_2O_3 contributing to higher viscosities with their highly covalent bonds[1]. In contrast, monoxides such as CaO and MgO exhibit ionic behaviour, leading to the destruction of silicate chains and lowering the viscosity[2]. These arguments only hold for the liquid slag-phase system, and in the multiphase system, an increase in monoxides leads to higher activities of solid phases and possible solids precipitation, which would increase the effective (observed) viscosity.

It should also be noted that, in the typical operation where it is possible to alter the slag composition, changes in the composition would have opposing effects. For example, the achievement of lower viscosity at higher basicities will likely be associated with the adverse effect of increased liquidus temperature. In addition to the effects on physicochemical properties, the slag basicity also influences the sulphur (and to some extent the phosphorus) removal capacity of the slag, and the silicon content of the hot metal; higher basicities lead to higher sulphur values in the slag and lower silicon values in the metal.

Metallurgical system

The models developed are based on phase equilibrium calculations, using thermodynamic data set up for specific metallurgical systems. To follow is the definition of the metallurgical system for the material of interest, considering the typical operating region of compositions and temperatures of blast furnace slags.

The viscosity and liquidus temperature models developed were configured and optimized specifically for blast furnace type slag. These slags contain predominantly SiO_2, Al_2O_3, MgO, and CaO, with smaller amounts of FeO, MnO, TiO_2, Na_2O, K_2O, and S that also influence the physicochemical properties. The temperature of the slag also influences the phase equilibria and subsequently the values of physicochemical properties.

Table I lists typical ranges and values for the chemical composition and temperature of blast furnace slag. The slag temperature values are estimates based on the assumption that this type of slag is approximately 100°C hotter inside the furnace than the tapped metal temperature. These values listed here can vary greatly between plants due to variances in the feed material.

Table I: Ranges and typical values of component mass percentages and temperatures of blast furnace type slags

Property	Range (mass %, or °C)	Typical Value (mass %, or °C)
CaO	30.0 – 45.0%	40.0%
MgO	3.0 – 18.0%	8.0%
Al_2O_3	8.0 – 17.0%	12.0%
SiO_2	30.0 – 47.0%	36.0%
S	0.2 – 2.5%	1.0%
K_2O	0.2 – 1.9%	0.6%
Na_2O	0.1 – 0.6%	0.4%
TiO_2	0.4 – 2.5%	0.7%
FeO	0.1 – 0.6%	0.3%
MnO	0.1 – 1.8%	0.3%
B4 = (CaO+MgO)/(Al_2O_3+SiO_2)	0.7 – 1.2	1.0
Slag Temperature	1520 - 1640°C	1590°C

Phase diagrams from literature were studied to obtain an indication of the stable phases under equilibrium conditions for the blast furnace type slag. One such source of ternary diagrams is the Slag Atlas[3] in which ternary phase diagrams can be found for the SiO_2-CaO-MgO-Al_2O_3 system with 10% and 15% Al_2O_3 respectively.

According to these diagrams, the following phases are expected at the given conditions:
- Melilite, $(Ca)_2[Mg,Fe^{2+},Fe^{3+},Al]\{Fe^{3+},Al,Si\}_2O_7$
- Pyroxene, Al_2CaO_6Si
- Merwinite, $Ca_3MgSi_2O_8$
- Dicalcium Silicate, $(Mg, Mn, Fe, Ca)_2SiO_4$
- Periclase, MgO
- Monticellite, $CaOMgOSiO_2$
- Pseudowollastonite, $CaSiO_3$

- Rankinite, $Ca_3Si_2O_7$
- Spinel, $MgAl_2O_4$
- Wollastonite, $CaSiO_3$
- Forsterite, Mg_2SiO_4

In the models developed, FactSage software[4] was used to perform phase equilibrium calculations, using the contained thermodynamic data mostly from the FToxid and FACT53 databases, as per the metallurgical system defined above. The metallurgical system was configured in the software, selecting pure condensed and solution phases representing these phases from the available FactSage data. Additional phases were also selected to allow for equilibrium calculations incorporating the minor components of FeO, MnO, TiO_2, Na_2O, K_2O, and S.

Effective viscosity
The effective viscosity is the liquid viscosity of the slag, adapted to consider the precipitated solids from the liquid slag. In single-phase molten slag systems, the viscosity can be estimated using relationships between slag composition and temperature, and here the Urbain formalism has been used, as it is well-documented[2]. Preceding the calculation of the liquid viscosity, the model uses FactSage to calculate phase equilibrium through Gibbs free energy minimization at the specified temperature. The liquid viscosity is calculated using the predicted composition of the slag liquid phase, possibly different from the bulk slag composition when the slag temperature is below the liquidus temperature of the slag.

The phase equilibrium calculation results are subsequently used to determine the volume percentage of all the solid phases forming. This is used to correct the liquid viscosity for possible solids by using the Roscoe equation[5]:

$$\eta_{eff} = \eta_{liq}(1 - 1.35\phi)^{-2.5}$$ [1]

Where η_{liq} is the liquid viscosity (Poise), η_{eff} the effective viscosity (Poise), and ϕ is the volume fraction of solids predicted in the molten slag.

Liquidus temperature
The liquidus temperature model also uses phase equilibrium calculations, through Gibbs free energy minimization in the FactSage software. For a specified slag composition, the target temperature calculation is performed to determine the temperature with the liquid slag phase as precipitation target. The temperature is subsequently determined where only masses of liquid slag components are predicted, and where at least one other solid phase has an activity of one but still zero mass. This indicates the point at which precipitation is expected to start should the slag be cooled.

Temperature for target viscosity
The viscosity model was used to estimate the slag temperature required for a specified viscosity of a slag composition. This could be applied to determine

the temperature range over which a given slag can be tapped without difficulty, or to compare the slags based on their energy requirements.

The effective viscosity model, as defined above, is executed iteratively, finding the temperature for which the target viscosity is predicted. The phase equilibrium and liquid viscosity is calculated at each point, to obtain the effective viscosity for each iteration with a guessed temperature. For typical blast furnace slag compositions and higher target viscosities, the resulting temperature is likely to yield a multiphase slag, where the solids contribute significantly to the increase in viscosity.

Smelting process optimization

In the normal slag operating region, changes in the basicity would have opposing effects on the slag viscosity and liquidus temperature, while also affecting the other controlled variables. Considering all control objectives, a global optimum has to be found in which costs are minimized while all the other controlled variables fall within their limits. The viscosity and liquidus temperature models are therefore components of a global optimizing model, and cannot be considered in isolation to determine the manipulated variables being the feed recipe and energy requirements.

The control objectives for optimisation of the blast furnace process include:
- *Minimize cost.* For smelting and reduction of the available ore/sinter, the least amount of other feed materials such as coke must be used. This requires a low liquidus temperature and viscosity to minimize the energy requirements.
- *Slag chemical composition requirements.* There might be limitations imposed on components in the slag, such as requirements for use in downstream processes.
- *Low slag liquidus temperature.* The slag liquidus temperature needs to be sufficiently low to prevent the slag to from solidifying too easily during tapping, but not so low as to impede physical separation of slag and metal in the trough.
- *Below target viscosity.* The chemical composition of the slag needs to be optimized for the viscosity to be as low as possible at the operating temperature, to decrease the energy requirement to operate with a tappable slag. The lower the slag viscosity, the better the slag–metal separation is as well.
- *Minimize slag volume.* Energy is lost as 'sensible heat' in the slag, which needs to be minimized. Material handling and waste disposal costs also need to be minimized.
- *Sulphur, silicon, and alkali removal capacity.* The chemical composition and temperature of the slag influences the capacity to remove these components in the slag, and control their distribution to the metal or the gas, or the accumulation thereof in the process (alkalis).

MODEL RESULTS

Model validation using measured data

Despite the primary purpose of this work being the development of qualitative models, the viscosity model was applied to published data on blast furnace slag viscosity measurements, to quantify to some extent the error in the model. Information was obtained from literature[6,7] where viscosities were measured of typical blast furnace type slags with varying basicities and Al_2O_3 contents.

The correlation between the effective viscosities predicted by the model and the viscosities measured[6,7] are illustrated in Figure 1. The overall correlation between the predicted and measured viscosities is better at lower viscosities relating to higher temperatures, but deteriorates at higher viscosities.
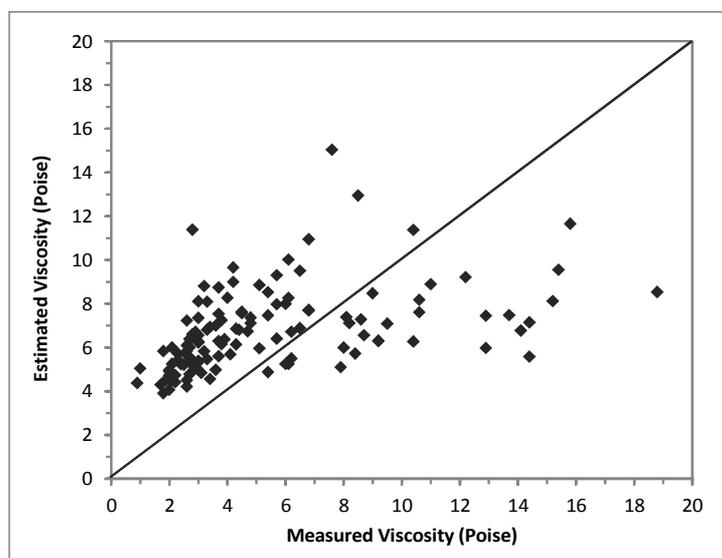
Figure 1: Correlation plot of the predicted effective viscosities versus the measured viscosities from literature[6,7]

Effect of solids on viscosity

A plot of the Roscoe equation[5] in Figure 2 illustrates the strong exponential effect that solids in the slag have on the viscosity, and the importance of having to consider it in the model. The value on the y-axis is the factor by which the liquid viscosity is multiplied to yield the effective viscosity figure.

The model was applied using a typical blast furnace slag composition (Table I) at different temperatures. Figure 3 illustrates the difference between the liquid viscosity and the effective viscosity. At lower temperatures, the viscosity model incorporating phase equilibria predicts significantly higher viscosities due to the effect of the solids precipitating from the slag phase.

Figure 4 illustrates viscosity of the typical blast furnace slag composition where the basicity (($CaO+MgO$)/SiO_2) is varied and viscosity calculated at 1500°C and 1600°C respectively. In both cases, it can be seen how the viscosity decreases with basicity as the SiO_2 decreases. At 1500°C and higher basicities, the viscosity is predicted to increase slightly, due to the effect of solids

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precipitating. This indicates how the effect of the solids could counteract the effect of the lower SiO_2 and cause viscosity to increase.
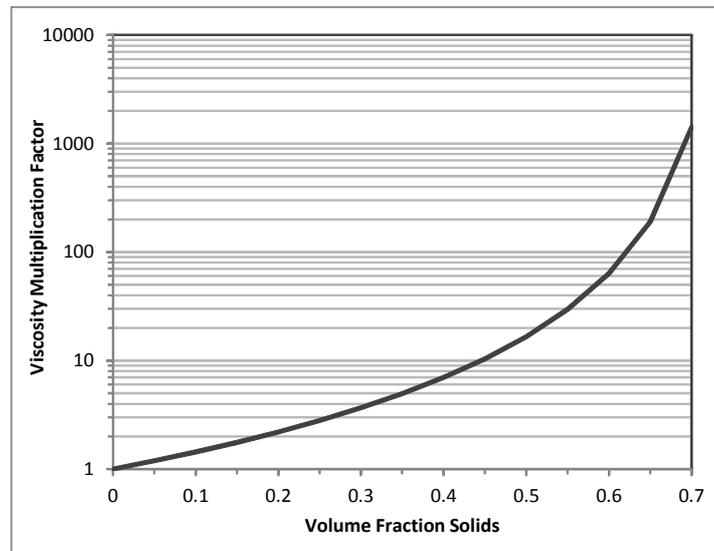
Figure 2: The exponential influence of solids on the viscosity predicted by the Roscoe equation[5]
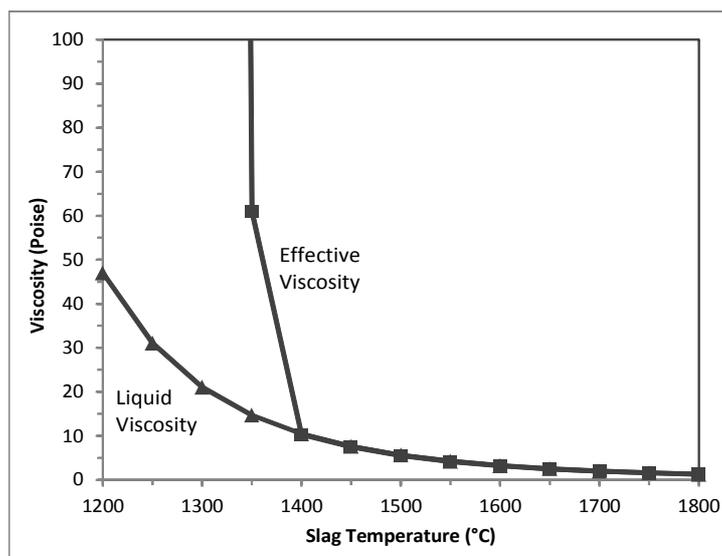
Figure 3: Viscosities calculated for typical blast furnace slag at different temperatures, illustrating the effect of precipitating solids on viscosity
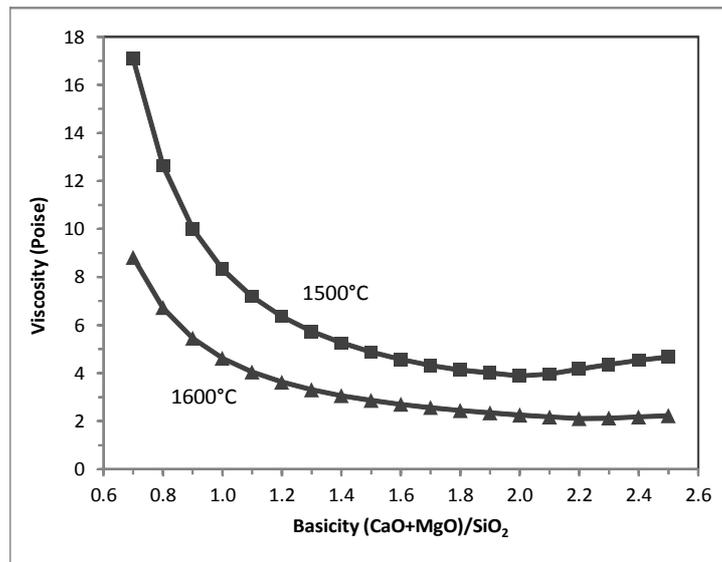
Figure 4: Effective viscosities of typical blast furnace slag with varying basicities ((CaO+MgO)/SiO2), illustrating the effect on viscosity of solids precipitating at higher basicities

Effect of solids on viscosity

As a visualisation aid, ternary diagrams were generated to illustrate the calculated properties over the complete range of possible compositions. The liquidus temperature ternary diagram is shown in Figure 5 for the quaternary system with 12.5% Al_2O_3. The increase of the liquidus temperature with increasing basicity can be observed.

The calculated effective viscosity for the quaternary system with 12.5% Al_2O_3 at 1600°C is illustrated in Figure 6. This shows that the viscosity increases at higher SiO_2 contents, but also at lower SiO_2 contents below the liquidus temperature. For example, compositions having an MgO content of 10% would have viscosities above 10 Poise at SiO_2 contents higher than 53%, but also with SiO_2 contents below 12%.

The volume fraction of solids predicted for this system is illustrated in Figure 7. The liquidus line is indicated by the line for the lowest volume fraction of solids calculated, and the values increase with decreasing amounts of SiO_2.

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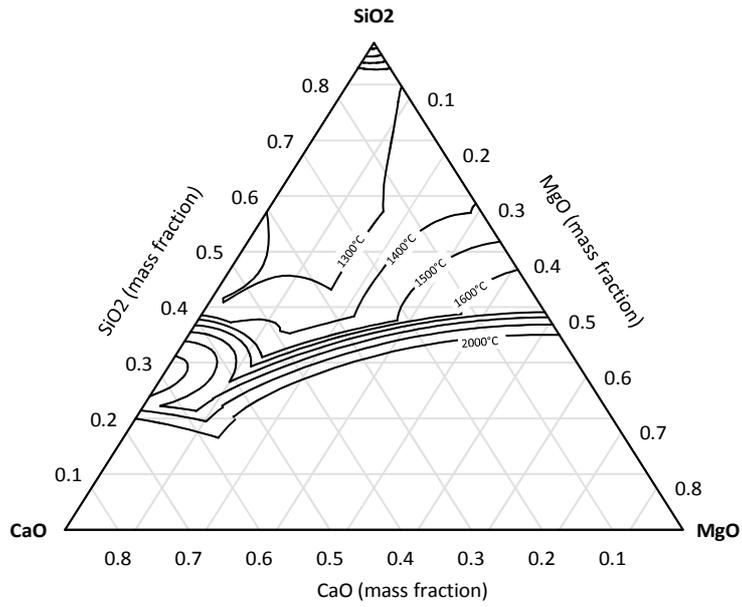
Figure 5: Liquidus temperature ternary diagram of the SiO_2-CaO-MgO-Al_2O_3 system, with 12.5% Al_2O_3
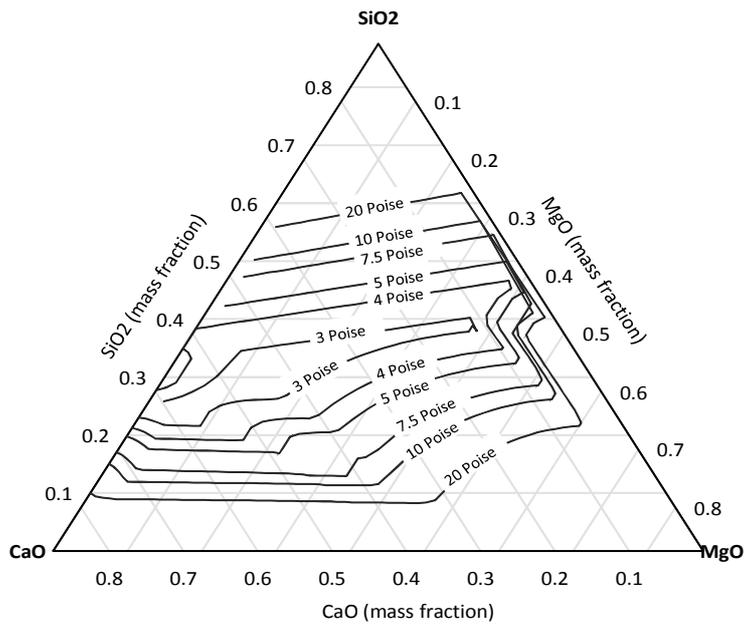
Figure 6: Effective viscosity ternary diagram of the SiO_2-CaO-MgO-Al_2O_3 system, with 12.5% Al_2O_3
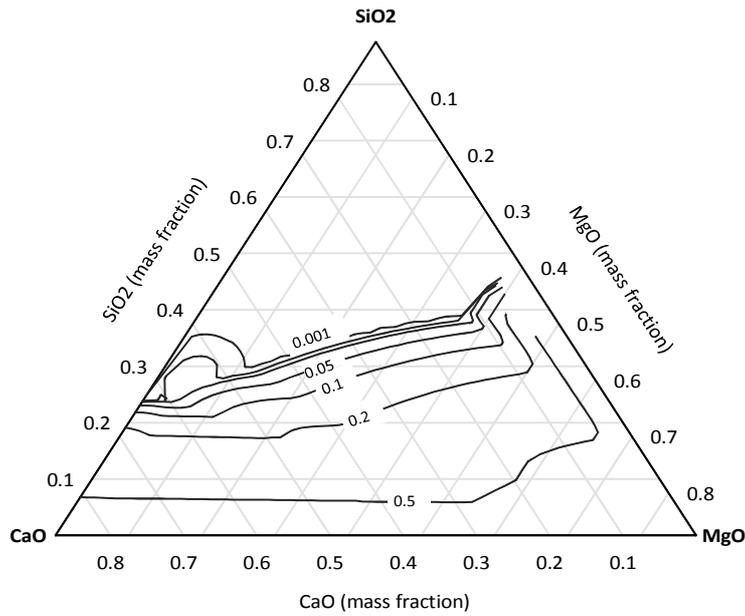
Figure 7: Volume fraction of solids ternary diagram of the SiO_2-CaO-MgO-Al_2O_3 system, with 12.5% Al_2O_3

Influence of basicity, Al₂O₃, and FeO

Effective viscosities were calculated for the typical blast furnace type slag (Table I) with varying basicity ((CaO+MgO)/SiO_2) at 1500°C, for different amounts of FeO and Al_2O_3. The results calculated for FeO equal to 0% are illustrated in Figure 8, and for FeO equal to 5% in Figure 9. The results indicate a lowering of the viscosity with increased FeO content, explained by FeO acting as a silicate bond breaker. At higher basicities, the MgO and CaO have the same effect, up to the point where solids start to precipitate and the viscosity starts increasing. Al_2O_3 has an increasing effect on viscosity, below the liquidus temperature, which appears to be somewhat elevated for higher Al_2O_3 concentrations.
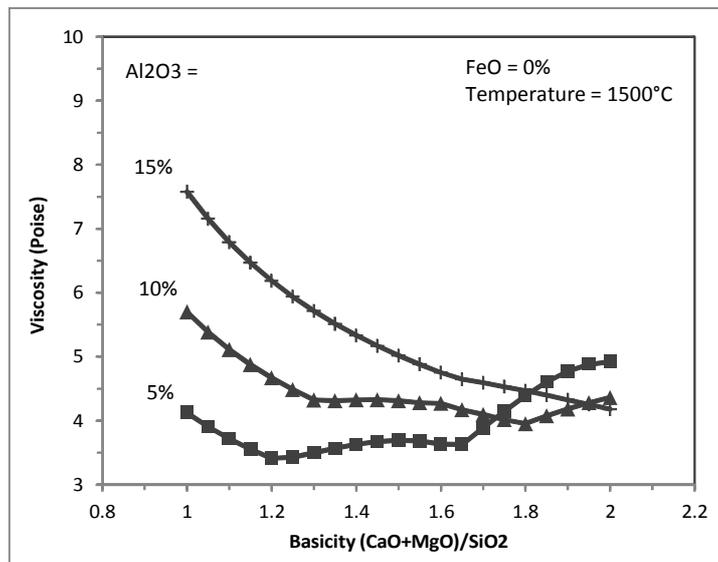
Figure 8: Effective viscosity as a function of basicity ((CaO+MgO)/SiO_2) at 1500°C for FeO = 0%, and Al_2O_3 = 5%, 10%, and 15%

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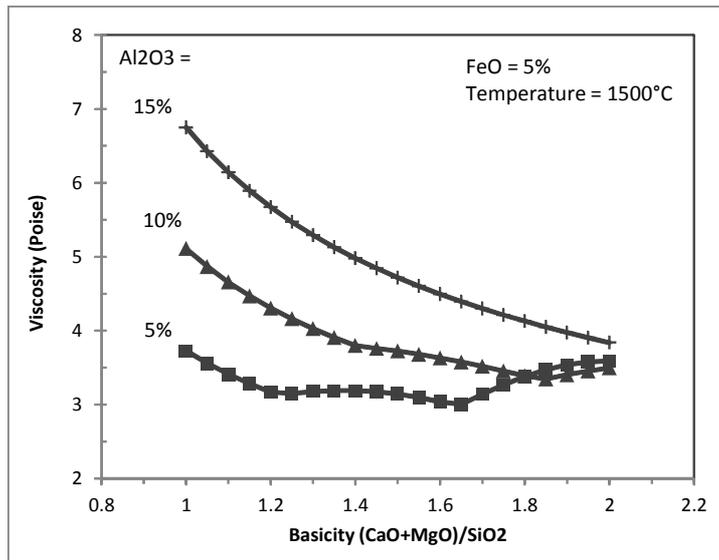
Figure 9: Effective viscosity as a function of basicity ($(CaO+MgO)/SiO_2$) at 1500°C for FeO = 5%, and Al_2O_3 = 5%, 10%, and 15%

Influence of temperature, FeO, and Al_2O_3

Effective viscosities were calculated for the typical blast furnace type slag (Table I) with a C/S ratio (weight percentage ratio of CaO/SiO_2 in slag) of 1.5 at varying temperature, for different amounts of FeO and Al_2O_3. The results calculated for FeO equal to 0% are illustrated in Figure 10, and FeO equal to 5% in Figure 11. In both cases, the results illustrate lower viscosities for lower Al_2O_3 concentrations. The results also illustrate that the compositions with lower Al_2O_3 contents have higher liquidus temperatures, as viscosities increased significantly at higher temperatures (compared to other compositions) due to the formation of solids.
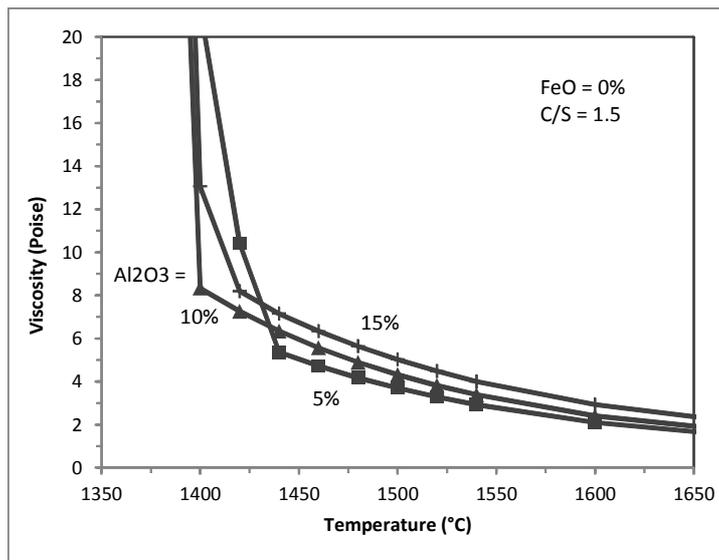
Figure 10: Effective viscosity as a function of temperature for FeO = 0%, C/S = 1.5, and Al_2O_3 = 5%, 10%, and 15%
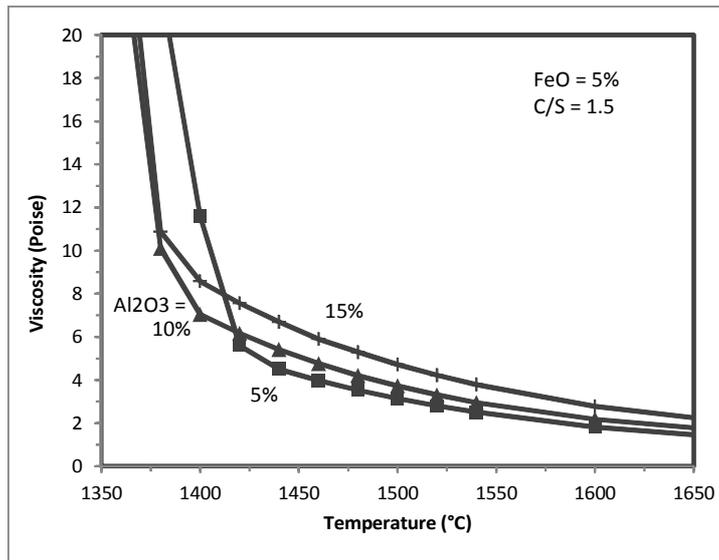
Figure 11: Effective viscosity as a function of temperature for FeO = 5%, C/S = 1.5, and Al_2O_3 = 5%, 10%, and 15%

Influence of temperature, FeO, and C/S

Effective viscosities were calculated for the typical blast furnace type slag (Table I) with Al_2O_3 equal to 10% at varying temperature, for different amounts of FeO and C/S. The results calculated for FeO equal to 0% are illustrated in Figure 12, and for FeO equal to 5% in Figure 13. As with varying the Al_2O_3 contents, the viscosities are lower and the liquidus temperatures higher for the compositions with increased C/S values.
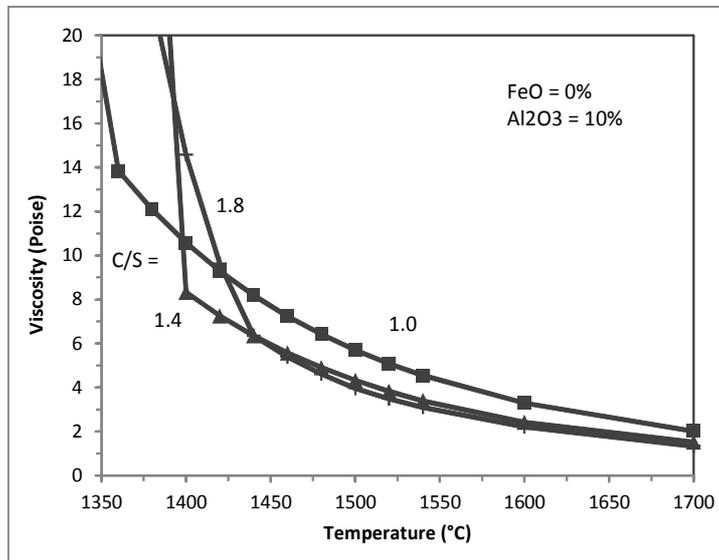
Figure 12: Effective viscosity as a function of temperature for FeO = 0%, Al_2O_3 = 10%, and C/S = 1, 1.4, and 1.8

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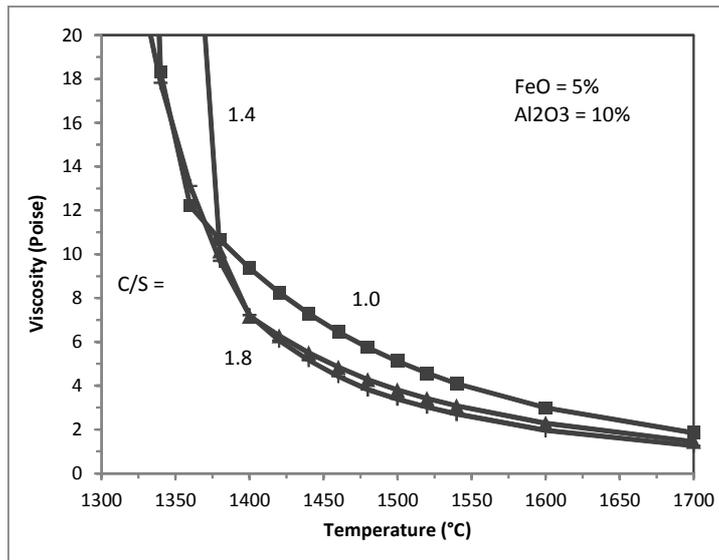
Figure 13: Effective viscosity as a function of temperature for FeO = 5%, Al_2O_3 = 10%, and C/S = 1, 1.4, and 1.8

The predicted volume fraction of solids as a function of temperature is illustrated in Figure 14, as calculated for the typical slag composition with FeO equal to 5%, Al_2O_3 equal to 10%, and varying values of C/S. This illustrates how the volume fraction of solids steadily increases at the liquidus temperature, up to a point where it starts to increase sharply. The liquidus temperature and range is higher for higher C/S values, as the viscosity of these compositions starts increasing, and peaks at higher temperatures.
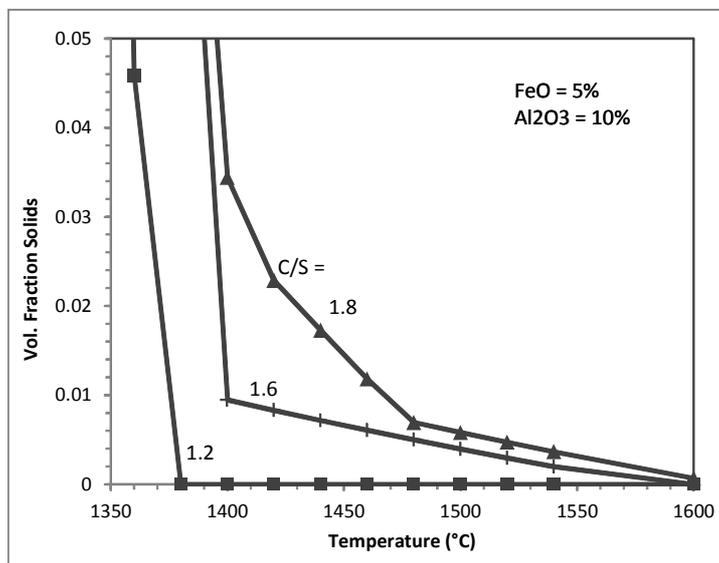
Figure 14: Volume fraction of solids as a function of temperature for FeO = 5%, Al_2O_3 = 10%, and C/S = 1.2, 1.4, and 1.8

THE ANALYSIS OF HISTORICAL DATA

Operating region

The viscosity and liquidus temperature models were applied to historical blast furnace data to illustrate the analysis of historical performance and to determine improvement targets. The slag compositions considered (normalised to average 12.5% Al_2O_3) are illustrated on the ternary liquidus diagram, shown in
Figure **15**. This indicates that the relatively wide variation in compositions are all expected to be fully liquid at final slag temperatures above 1600°C.
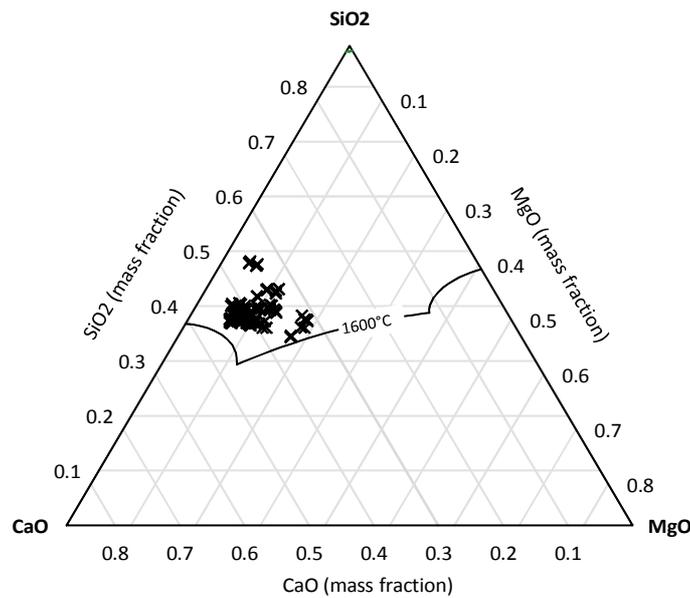
Figure 15: Liquidus ternary diagram for the SiO_2-CaO-MgO-Al_2O_3 system, with 12.5% Al_2O_3, showing the historical blast furnace slag compositions considered

Liquidus temperatures

Al_2O_3 in the slag lowers the liquidus temperature, and a sharp gradient is predicted in the liquidus temperatures towards lower SiO_2 values. The liquidus temperatures had to be determined for compositions individually to confirm operation in the single-phase region. Figure 16 shows a histogram of the predicted liquidus temperatures.

An average liquidus temperature of 1380°C was predicted, with all the values below the typical metal temperature of 1450 to 1480°C. The final slag in the furnace is generally assumed to be 100°C hotter than the metal tapping temperature measured, indicating operation in the fully liquid region for all slags considered.
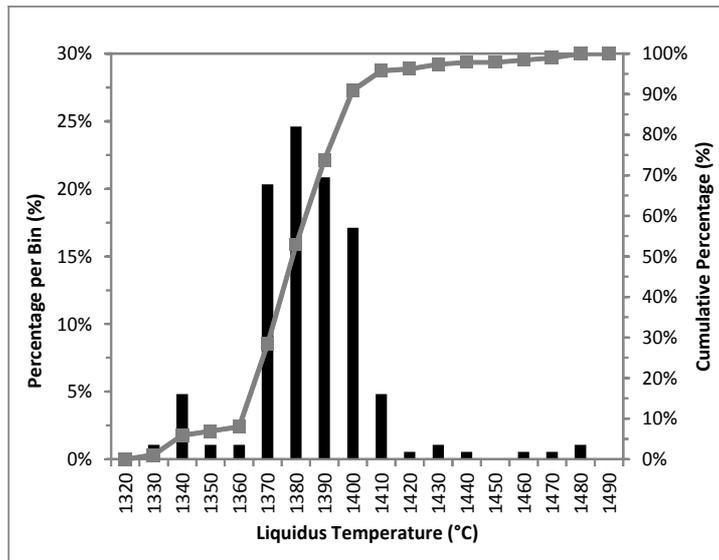
Figure 16: Histogram of predicted slag liquidus temperatures for historical blast furnace slag compositions

Metal tapping temperatures

The metal tapping temperature recorded with each slag composition is considered in relation to the predicted slag liquidus temperatures. Figure 17 shows a histogram of the difference between tapped metal temperatures and predicted slag liquidus temperatures. On average, the tapped metal temperatures were 105°C higher than the slag liquidus temperatures predicted, with the distribution skewed to the right. This average figure is high and a fairly conservative target metal-tapping temperature could be approximated from the slag composition by subtracting 105°C from the calculated slag liquidus temperature.
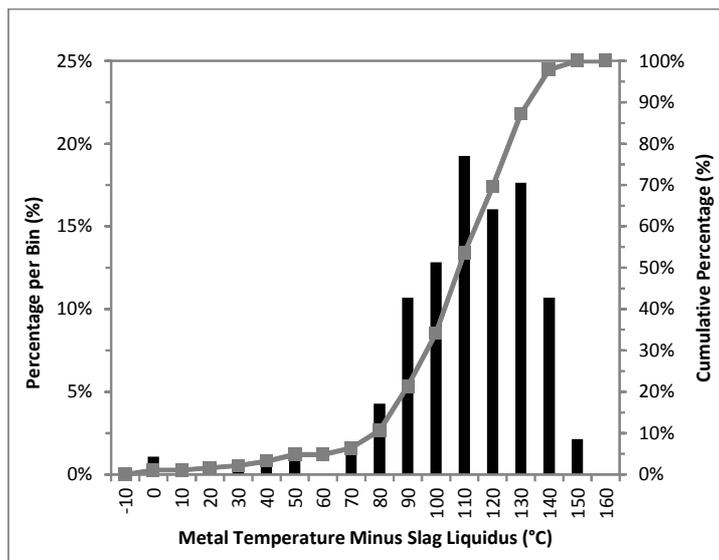
Figure 17: Histogram of the differences between the metal tapped temperature and predicted slag liquidus temperature, for historical blast furnace data

Effective viscosity

In the single-phase region, the slag viscosity is inversely proportional to the slag basicity, while the slag liquidus temperature is directly proportional. Even though the final slags considered were all in the single-phase region, the viscosities have to be calculated to determine the spread of predicted values. This was done using the developed viscosity model, assuming a final slag temperature of 100°C higher than the metal tapping temperature, with the histogram of predicted values in Figure 18.

An average value of 3.7 Poise was predicted, with the distribution skewed to the left. The higher viscosity values are likely to indicate operational occurrences where slag was difficult to tap. This average value could be considered indicative of a slag having acceptable ease of tapping, based on the general ease of the period for which data was obtained.
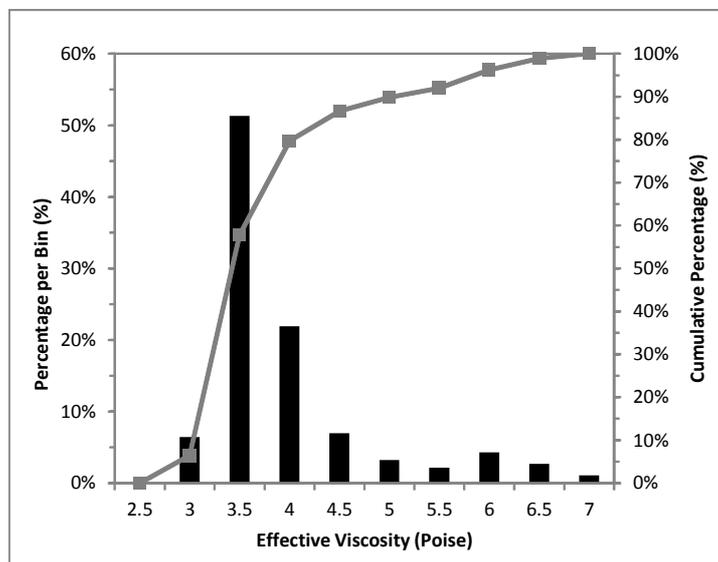
Figure 18: Histogram of predicted slag effective viscosities for historical blast furnace slag compositions

Combined analysis of results

The predicted slag viscosities (at 100°C above metal tapping temperature) were plotted versus the predicted slag temperatures in Figure 19, with the encircled region showing where both parameters are minimized. The highest variance occurs in the liquidus temperatures, due to variances in the slag composition and in the feed materials. Being skewed to the left, the process was probably managed with compositional changes that related to low liquidus temperatures.

There is less variance in the predicted slag viscosities, also skewed to the left. If temperature remained unchanged, the same effects causing lowered liquidus temperatures would have increased viscosity (decrease in basicity), and the distribution would not be skewed to the left. This indicates that the temperatures were managed and kept high enough to maintain low viscosity.

The results indicate that possible opportunities exist to control the slag chemical composition and energy input, to obtain less variability in the slag liquidus and metal temperatures. Lower variability in these variables should enable operation closer to overall optimized conditions, being lower metal temperatures (for lower energy demand and target viscosity) and limited slag liquidus temperatures (for ensuring tappability).

That said, it should be kept in mind that slag liquidus temperatures and viscosities cannot be controlled in isolation, and the other control objectives and the influence that compositional changes have on them should also be considered. It is important to also consider variables such as the capacity of impurities in the slag, and compare it with baseline values when slag compositions and temperature target values are.
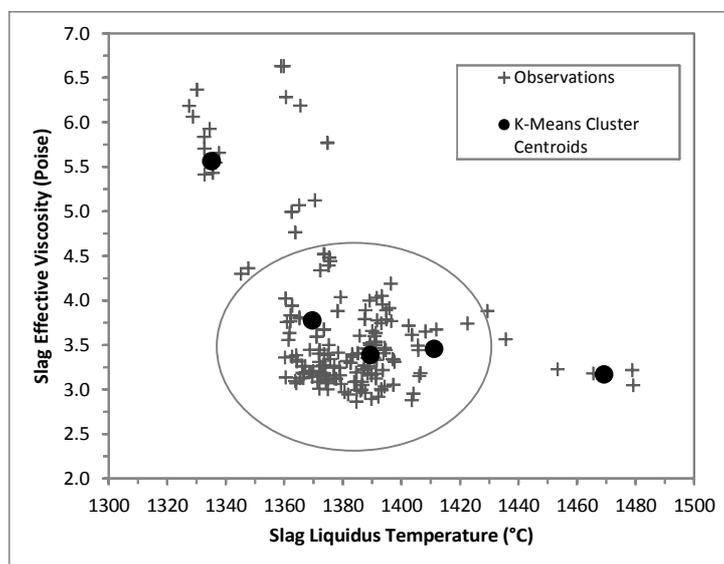
Figure 19: Predicted slag effective viscosity vs slag liquidus temperature for historical blast furnace data

CONCLUSIONS

Viscosity and liquidus temperature models incorporating phase equilibrium calculations were developed and applied to blast furnace type slags. The importance of considering phase equilibrium calculations was illustrated by calculating viscosities for varying temperatures, basicities, and amounts of FeO and Al_2O_3. It was observed how the viscosity model predicted increased viscosities at low basicities, due to the silicate bonds, but also at high basicities due to the effect of precipitating solids.

The models were applied to historical blast furnace data, as a first step towards analysing the historical performance based on the values of viscosity and liquidus temperatures, and also to derive possible target values should the models be actively applied to the control of the process. It was shown that, for the data used, and an assumed slag temperature elevation of 100°C above the metal temperature, that the average slag viscosity was 3.7 Poise. The

information obtained from the analysis and models developed could however not be used in isolation, but should be a vital component in an overall process model and optimization system.

ACKNOWLEDGEMENTS

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