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ANALYSIS OF BLOATED DROPLET THEORY USING STEELMAKING PROCESS MODELS

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ABSTRACT

There is significant evidence that droplets generated in steelmaking "bloat" due to the inability of gas generated from the decarburisation reaction to escape from the surface of liquid metal droplets. A model to describe this behavior was developed by Brooks, Subagyo, Coley and Pan based on their own experimental work and calculations and previous studies by Fruehan and co-workers. This approach has been successfully incorporated into an overall process model of oxygen steelmaking. A unique feature of this model is an evaluation of the decarburization kinetics of individual metal droplets in the emulsion and comparing this to the overall kinetics of oxygen steelmaking. The model suggests that the droplets become bloated and remain in the emulsion for long periods (30+ seconds). This paper evaluates the effects of droplet size and volume fraction on the bloating behavior of droplets and critically examines the repercussions of the new theory on plant design and operation.

Introduction

One of the main goals of the oxygen steelmaking process is to effectively reduce the carbon concentration of the liquid iron. It is understood that most carbon removal reactions occur in the emulsion phase via a reaction between the metal droplets and slag phase.(Meyer *et al.*, 1968, Price, 1974) An improved understanding of decarburization reaction and the factors controlling the overall rate should provide better control of the process and increase the productivity. In the literature, there is a limited knowledge on how to relate the carbon removal rate within the droplets to the overall kinetics of the process under full scale operating conditions.

A computer based model which incorporates the bloated droplet theory under dynamic conditions was developed to evaluate its influence on the overall kinetics of the process. The model focused on the decarburization reaction in different reaction zones to predict the carbon content of liquid steel throughout the blow. The system includes 17 sub-models and 2 reaction zones. Two reaction zones, namely, the emulsion and impact zone are considered to investigate the kinetics and mechanism of carbon removal reactions since it is well known that these reactions take place via direct oxygen absorption at the impact area and FeO reduction in the emulsion phase. The reaction zones are linked to each other by material streams which are slag constitutes and liquid metal droplets. The input mass flows, process conditions and calculation sub-models to be considered for each reaction zone are hot metal, scrap and flux charges, hot metal,

scrap and slag compositions, oxygen blowing conditions, lance height, gas flow rates, temperature of the bath, the slag and the impact zones, flux dissolution, scrap melting, ejected metal droplets behaviour such as droplet generation rate, droplet size, residence time in the emulsion, decarburization rates in the emulsion and impact zones and are illustrated in Figure 1.

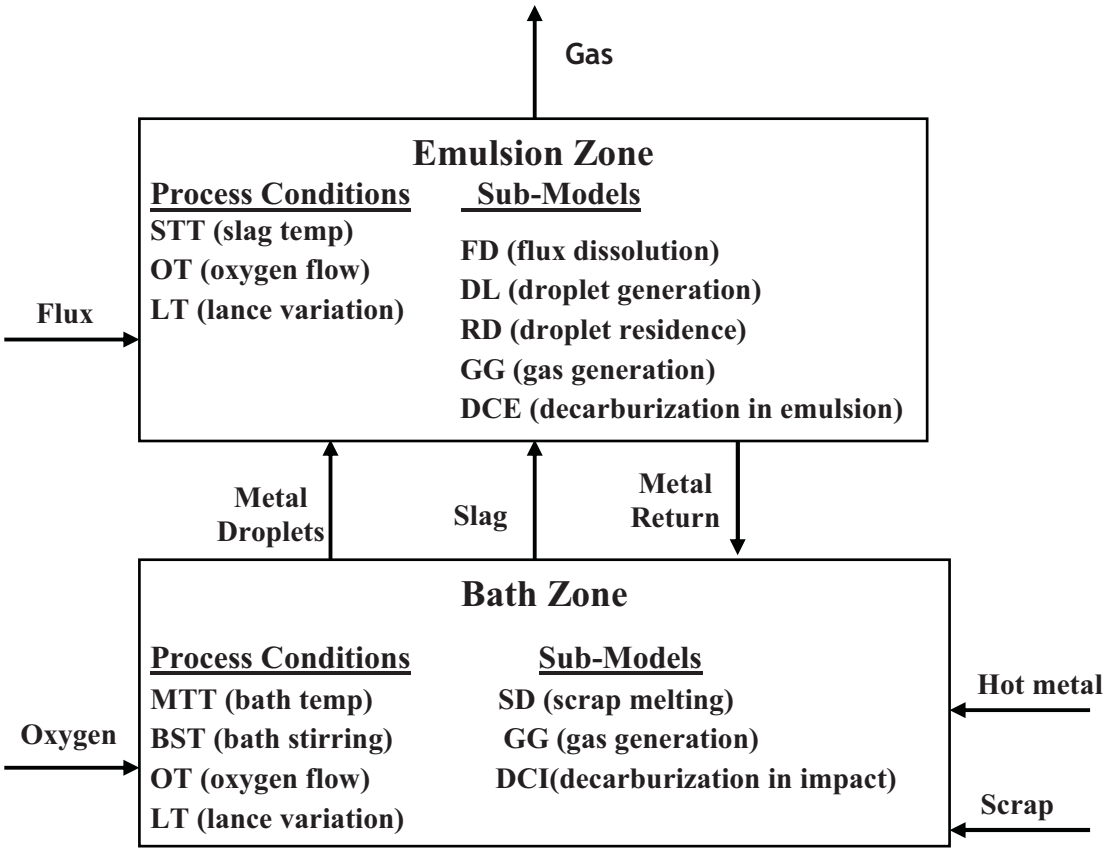

Fig. 1: The schematic description of the system

Once all the variables deemed to be important to the each sub-model have been identified, inter-relationships among them are developed. These sub-models were modelled individually in which they include the calculation procedure, assumptions and boundary conditions to represent each process variable considered. In the following step, the sub models were linked to each other dynamically to be used as input data or boundary condition and sub-models formed the global model of the oxygen steelmaking process. Most of the sub-models such as flux dissolution, droplet generation, scrap melting, decarburization reactions in the emulsion and at the impact zone that form the global model and the global model itself have been validated against industrial data in the open literature to investigate the feasibility of the application of the developed models. The results have been published elsewhere.(Dogan *et al.*, 2009a, 2009b, Dogan *et al.*, 2011a, Dogan *et al.*, 2011b, Dogan *et al.*, 2011c) This model is the first attempt to determine the role of emulsion quantitatively based on the bloated droplet theory, using a theoretical model under full scale operating conditions. In this paper the effects of droplet size and gas fraction in the emulsion on bloating behaviour of droplets will be discussed.

Basis of the model

In the development of the global model there are two sub-models generated to calculate the decarburization rates in the emulsion phase namely, decarburization rate in the emulsion and droplet residence models. The theoretical treatment suggested by Brooks *et* al.(2009) is applied in the decarburization in the emulsion model. Using this approach, total decarburization rate in the emulsion zone can be obtained from the summation of decarburization rates of individual metal droplets as a function of droplets volume due to the bloating behavior of droplets. The decarburization rate can be calculated using;

$$\frac{dW_C}{dt} = \frac{\sum_{i=1}^{m} \frac{m_i}{100}\left(C_i^{t+\Delta t} - C_i^{t}\right)}{\Delta t} \tag{1}$$

Here i represents the ejected time of metal droplets, m_i is the weight of a single droplet and C is the carbon concentration.

It is known that the carbon content of metal droplet ejected from the bath is lower than carbon content of the liquid metal. However, there is no calculation technique available to predict the initial carbon concentration of metal droplets. Therefore, it was assumed that carbon content of the metal droplet was equal to the bulk carbon content of the liquid metal. Bulk carbon content was calculated using mass balance, which includes scrap melting, the decarburization reactions in the emulsion and impact zones as given in Equation (2).

$$W_b^t \frac{mass\%\ C_b^t}{100} = W_b^{t-\Delta-} \frac{mass\%\ C_b^{t-\Delta-}}{100} + W_{Sc}^t \frac{mass\%\ C_{sc}}{100} - \left(\frac{dW_C}{dt}\right)_{em} \Delta t - \left(\frac{dW_C}{dt}\right)_{im} \Delta t \tag{2}$$

where W_b is the mass of metal in the bath, W_{sc} is the mass of scrap melted in the bath. The subscripts *em* and *im* refer to emulsion and impact, respectively. It should be noted that the amount of carbon removed via emulsion represents the amount of carbon removed via metal droplets returning to the liquid bath. The metal droplets suspended in the emulsion phase have no impact on the overall mass balance of carbon in the bath.

The value of initial carbon content of metal droplets generated was fed as an input data to the Droplet Residence (RD) Model. In this sub-model the amount of carbon removed within the droplets was calculated based on a simple surface renewal model of carbon diffusions, developed by Brooks *et* al.(2005) The selection of this model can be found in an earlier article by the authors.(Dogan *et al.*, 2010) It is important to note that the Brooks *et* al.(2005) model used empirical data from the Molloseau & Fruehan study (2002) for some of their parameters in their model, so in effect, “fitted” their model to the data, and they only claimed that this model is useful for global kinetic calculations and not necessarily the “correct” kinetic model. In this model, effective mass transfer coefficient, k_{eff} was calculated on the basis of Higbie’s penetration theory (1935) This theory was employed by Brooks *et* al.(2005) to predict the mass transfer coefficient of carbon to the interface based on the residence time of the dense and bloated droplets. The relationship is;

$$k_{eff} = 2\sqrt{\frac{D_C u_d}{\pi D_p}} \tag{3}$$

where D_C is the diffusivity of carbon and u_d is the overall velocity of droplet.

The generated droplets, whose residence time is smaller than defined time-step, are returning from the emulsion phase. If the residence time was larger than the time-step, decarburization of droplets with that particular residence time was added to calculate the rate of overall decarburization in the emulsion phase as per Equation (1).

The residence time of the droplets was predicted based on the dynamics of the motion of droplets in the slag-gas-metal emulsion phase developed by Brooks *et* al.(2005). In the proposed model, force balance was made based on the ballistic motion of a single droplet at vertical and horizontal coordinates.

The following assumptions have been made in the model based on the industrial data available for a 200-ton oxygen steelmaking furnace. The operational conditions used for these calculations were taken from the industrial data reported by Cicutti *et* al.(2002, 2000) The outcome of other refining reactions such as FeO concentration was entered as known variables.

1) The effect of size distribution was not included and the diameter of a metal droplet was assumed to be 2 mm since the mean value ranges from 1 to 2 mm reported by Price (1974) And this value is valid for the industrial data taken from Cicutti *et* al.(2000) They also stated that drop size varies from 0.23 mm to 3.35 mm.

2) Subagyo *et* al.(Brooks et al., 2005, Subagyo & Brooks, 2005) proposed that the ejection angle of bloated droplets is a minor effect on the residence time calculations since the motion of the droplets is dominated by buoyancy. In this study, the effect of ejection angle was evaluated.

3) Diffusivity of carbon in liquid iron is $2\text{x}10^{-9}$ m^2/s at 1600°C.(Guthrie, 1989) Diffusivity of carbon will then be determined as a function of slag temperature and viscosity of slag-metal-gas emulsion based on the Stokes-Einstein and Eyring equations for various temperatures. It was assumed that slag temperature increases linearly and was 100°C higher than bath temperature.

4) For a slag-metal-gas emulsion, the motion of metal droplets is influenced by the gas bubbles trapped in the gas phase. The metal droplets are treated as dispersed phase in a slag-gas continuum. The average density and viscosity of the slag-gas continuum was calculated by the following equations.(Brooks et al., 2005, Subagyo & Brooks, 2002)

$$\rho_{sg} = \rho_g \phi_g + \rho_s (1 - \phi_g) \qquad \mu_{sg} = \frac{2}{3} \frac{\mu_s}{(1 - \phi_g^{1/3})} \frac{(\rho_{sg} - \rho_g)}{(\rho_s - \rho_g)} \tag{4}$$

Here ϕ_g refers to the volume fraction of the gas in the emulsion and can be given by;(Brooks et al., 2005)

$$\phi_g = \frac{V_s}{V_g + V_m + V_s} \tag{5}$$

Initially, the density and viscosity of slag was calculated as a function of slag composition and temperature. These values were entered as an input data in the droplet residence model. The average density and viscosity of slag-gas continuum were calculated as a function of gas volume to predict the velocity of the droplets at z and r directions.

5) The initial velocity of a droplet was estimated based on the conservation of energy relationship proposed by Subagyo *et* al.(2005) The relationship suggests that the kinetic energy of blown gas is used to generate and eject the droplets and it is valid if the all produced droplets are in spherical shape.

6) The equilibrium concentrations of carbon and iron oxide were determined by well established thermodynamics relationships using Henry's Law and Raoult's Law. (Turkdogan, 1996) The data for activity coefficients were taken from the literature. (Ban-ya, 1993, Sigworth & Elliott, 1974)

7) Ito and Fruehan(1989a, 1989b) reported that gas fraction in the slag-metal-gas emulsion varies between 0.7 and 0.9. Average value of this range, 0.8 was used in the calculations of slag-gas continuum in this study.

8) Slag foam height was assumed to be constant and it was equal to 2 m. In oxygen steelmaking furnace, the foam height might reach to the mouth of the furnace, particularly during the main blowing period. An assessment of this assumption will be discussed in the results section.

9) The time step for droplet residence model was selected based on previous work by Brooks *et* al.(2005), that demonstrated that time steps below millisecond was required to ensure that the predictions did not significantly differ with different time steps. Accordingly, 0.0001s was selected which is sufficient enough for the numerical accuracy and for computational time. The global model was developed using a time step of 10 s based on the model predictions compared to the industrial data.

10) There are several studies (Gare & Hazeldean, 1981, Gaye & Riboud, 1977, Sun & Zhang, 2005) available in the literature that focus on the effects of silicon, manganese, phosphorus, and sulphur in metal droplets reacting with an oxidizing slag, on the mechanism of decarburization reaction. It was found that these impurities have a retarding effect on the reaction kinetics. Sun and Zhang (2005) found that this effect was lower for low concentrations of manganese and silicon. Based on current knowledge, it is difficult to estimate the concentrations of impurities in metal droplets and incorporate them into the model development. Additionally, the industrial data used in this study has no information about impurities within the metal droplets. For simplicity, it was assumed that droplets only contain carbon and the effects of other impurities were not included.

Results and Discussion

Residence Time

Figure 2 illustrates the evolution of the residence time of droplets with a diameter of 2 mm as a function of the carbon concentration of the bath, as predicted by the proposed model. In the early part of the blow the residence time of droplets is around 45 s. Towards the end of the blow it decreases to 0.4 s. As seen, the residence time of droplets is much higher in the presence of high carbon concentrations. Towards the end of the blow the residence time is low due to weak decarburization rates. This may imply that the metal droplets are "bloated" with CO gas, generated during the active decarburization period and then become less dense and spend longer time in the emulsion.(Min & Fruehan, 1992) However, towards the end of the blow the metal droplets maintain their original density due to slow decarburization.

Significant differences in droplet residence time are also due to the physical properties of slag as a function of the gas volume fraction. For example, the gas hold up of 80% in the emulsion increases the viscosity of the slag-gas continuum by two times the viscosity of the slag, and decreases the density of the slag-gas continuum by four times the density of the slag. Model results of residence time, assuming a gas volume fraction of 0.8 with respect to the density and the viscosity of slag-gas continuum, are illustrated in Figure 3.

The residence times of droplets decrease as the density of the slag-gas continuum decreases towards the end of the blow. On the other hand, it is expected to observe a higher residence time because the viscosity of the slag-gas continuum decreases. As the carbon content of droplets decreases the droplets are not bloated and have a short residence time. This demonstrates that the carbon content of liquid iron has a predominant role on the residence time of droplets.

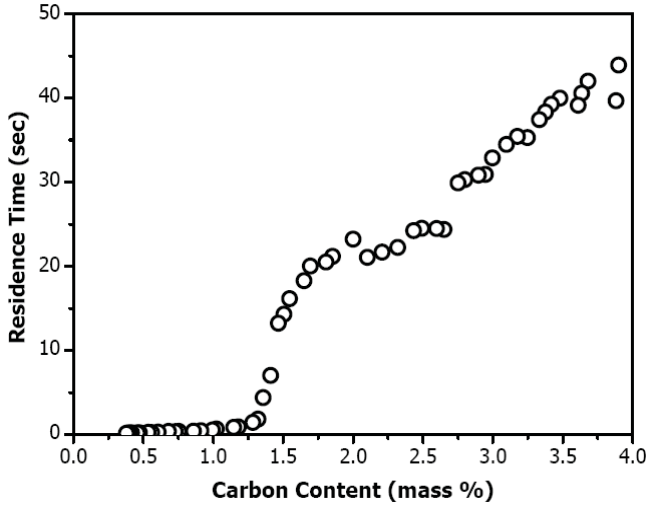

Fig. 2: Residence times of droplets as a function of initial carbon content in the metal droplets

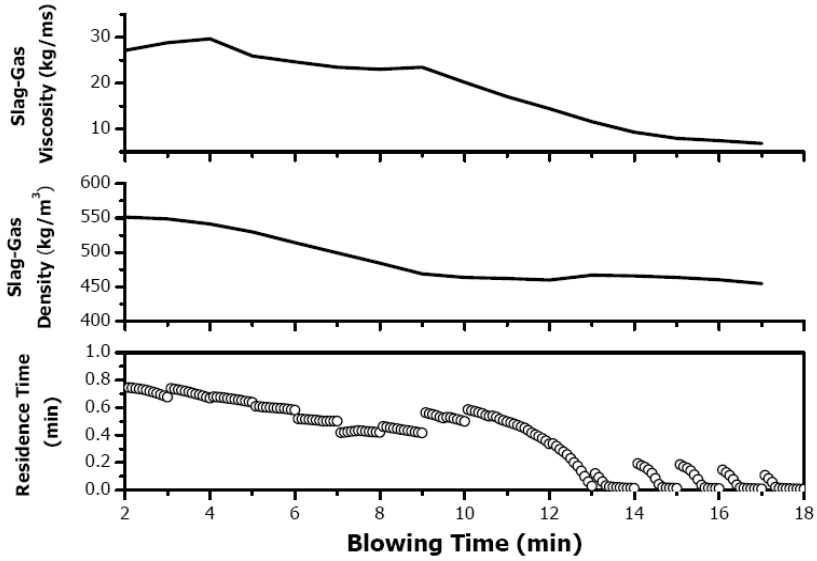

Fig. 3: Evolution of droplets residence time with respect to physical properties of slag-gas continuum during the blow

Effect of Gas Fraction on Residence Time

Figure 4 shows the residence time of bloated droplets in the slag-metal-gas emulsion predicted for different volume fractions of gas hold up. It can be seen from these results that the greater the hold up of gas in the slag, the shorter the predicted residence time of the droplets. As the gas volume fraction increases from 0.7 to 0.9, the residence time of droplets is predicted to decrease from 45 s to 0.9 s. This variation is due to a change in the physical properties of the slag-gas continuum. Particularly, the variation in viscosity has a greater impact on the residence time of the droplets. The residence time of droplets will decrease from 51 s to 0.6 s as the viscosity of the slag-gas continuum increases from 26.02 kg/ms to 28.175 kg/ms, if the volume fraction of gas increases from 0.7 to 0.9.

A further instance of this is that the density of the slag-gas continuum varies between 450 and 550 kg/m^3 for a gas fraction of 0.8. This range will decrease approximately 250-270 kg/m^3 for a gas fraction of 0.9. As the gas hold up increases the density decreases and the viscosity increases, which in turn, influences the velocity of the droplets and thereby the trajectory of the droplets. As a result, the droplets return to the bath in a shorter time. This implies that the amount of gas in the emulsion is crucial to accurately predict the residence of droplets.

The presence of gas in the emulsion depends on the bloating behavior of the droplets as well as gas generated from the impact zone. The amount of gas generated in the emulsion was calculated as a function of the gas generated within the metal droplets and in the impact zone. The change in the gas fraction is given in Figure 4. As seen from the results presented, the gas fraction is relatively smaller than those reported in the literature. Figure 4 and 5 show that the fraction of gas is important to predict the residence time of metal droplets. However, it is very difficult to estimate this process variable due to the lack of data in the literature. Therefore, the gas volume faction will be considered as constant and equal to 0.8 in this study.

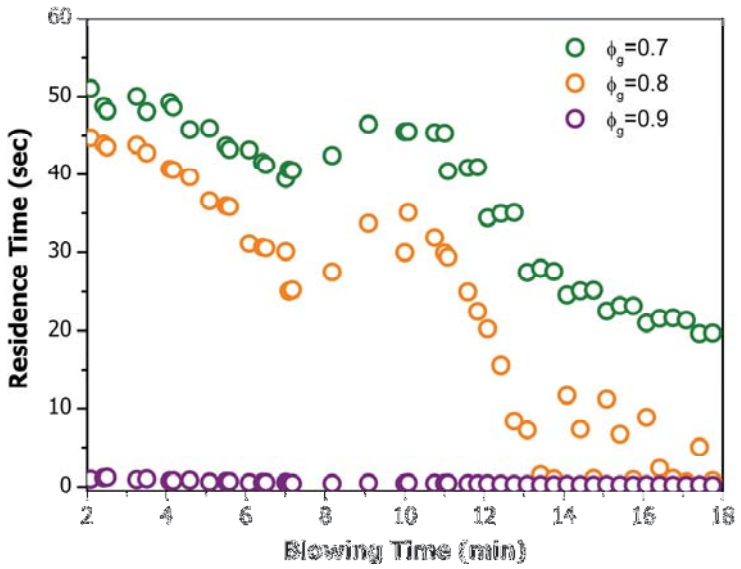

Fig. 4: Residence time of the droplets as a function of gas fraction

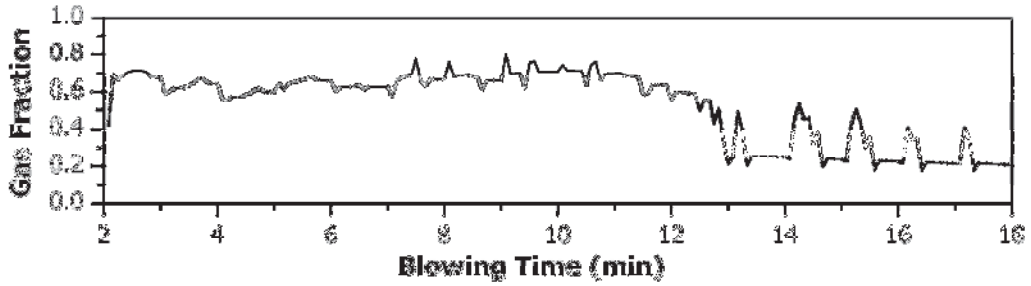

Fig. 5: Gas fraction in the emulsion during the blow

Effects of Droplet Size on Residence Time

Figure 6 illustrates the change in the size of a droplet ejected at a 60° angle from the bath predicted by the global model as a function of residence time under various operating conditions. Table 1 lists the selected operating conditions such as variations in the lance height and FeO concentrations of the slag taken from industrial data by Cicutti *et al*.(2000) at various blowing periods, to investigate the effects of different process conditions on the behavior of metal droplets.

Tab. 1: Measured FeO concentration and lance variations taken from the industrial data(Cicutti et al., 2000) at different blowing period

	mass % FeO	Lance height	Ejection time from start of the blow	
			min	sec
Early blow	31.3	2.5 m	3	00
Main blow	17.5	2.2 m	8	00
End blow	23.5	1.8 m	15	00

There is a significant increase in diameter (three times larger than initial diameter) of a droplet due to its bloating behavior regardless of the blowing period. However, this increase in size decreases towards the end of the blow. This is most likely due to a decrease in the carbon concentration because the decarburization rate decreases with a decrease in the carbon concentration of the droplets. This indicates that maximum decarburization will be achieved initially followed by an eventual decrease in the reaction rate. The carbon content of the droplet and concentration of FeO are important in determining changes in the size of the droplet. However, the effect of variations in FeO content in the slag is not clear in this study. Therefore, a more robust understanding of how FeO varies during the blow will be important to improve the models described in this study because the FeO values used in our calculation come directly from industrial data.

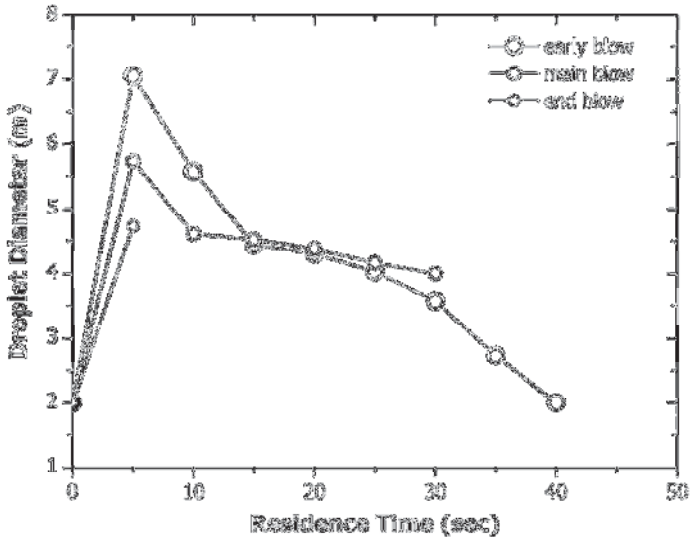

Fig. 6: Change in diameter of droplets ejected in a 60-deg angle at different times predicted by the model

Additionally, slag formation has an influence on the prediction of droplet residence times. However, it is unlikely the current study will investigate any further because the model does not include slag formation. The effect of slag formation on the droplets residence time is also worthy of further study.

Figure 7 illustrates the model's predicted residence time of bloated droplets ejected at a 60˚ angle with respect to different droplet sizes in various ejection periods. The droplet residence time varies significantly with respect to the operating conditions as well as droplet size. In an early blow, as droplet size increases, the residence time increases. It is most likely that the metal droplets contain more carbon and they require a longer time for the decarburization reaction. The droplets with an initial diameter of 0.25 mm are predicted to return quickly due to fast decarburization in the early and main blowing period.

In addition, the blowing conditions also influence the distribution of residence time for different size droplets. In the main blow, the residence time first increases with the initial diameter of the droplets and then decreases for droplets initially larger than 1 mm. A similar behavior was obtained for droplets initially larger than 0.5 mm towards the end of the blow. It can be concluded that maximum residence time shifts from larger droplets to smaller ones as the carbon concentration decreases.

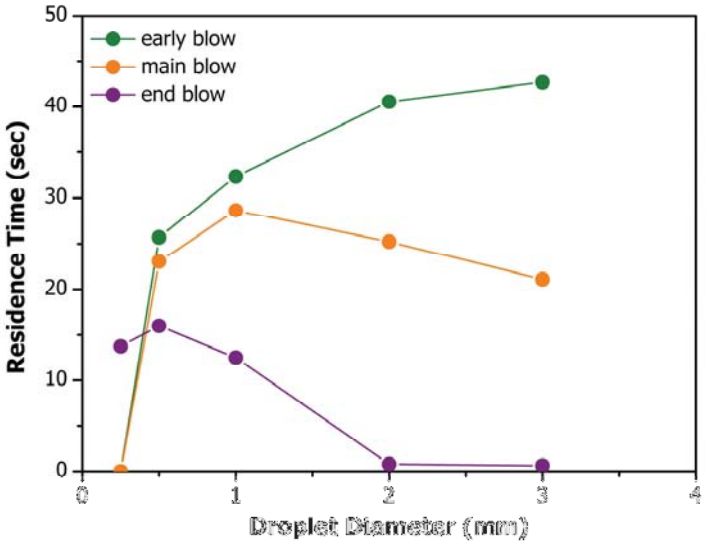

Fig. 7: Residence times predicted by the model for industrial data by Cicutti *et al*. as a function of droplet size at different blowing period

The trajectory of droplets with various droplet size ejected at a 60° angle at various ejection times was compared with respect to time in Figure 8. It shows that larger droplets are predicted to spend a longer time than smaller droplets in the emulsion, except at the end of the blow. In the early blow the droplets are thrown higher than those ejected during later stages of the blow. It can be noticed that the droplets with an initial diameter of 3 mm return to the bath immediately since the ejection velocity is more likely to become low towards the end of the blow. On the other hand, droplets with an initial diameter of 0.25 mm return to the bath zone directly, due to fast decarburization. This implies that the residence time of droplets decreases significantly, which in turn lowers the decarburization rates, particularly towards the end of a blow.

The model predicts that the metal droplets could not reach the top of the slag and the maximum height predicted varies as a function of the blowing period. The highest point in the z direction predicted is 0.55 m which is a relatively short distance from assumed height of the slag foam.

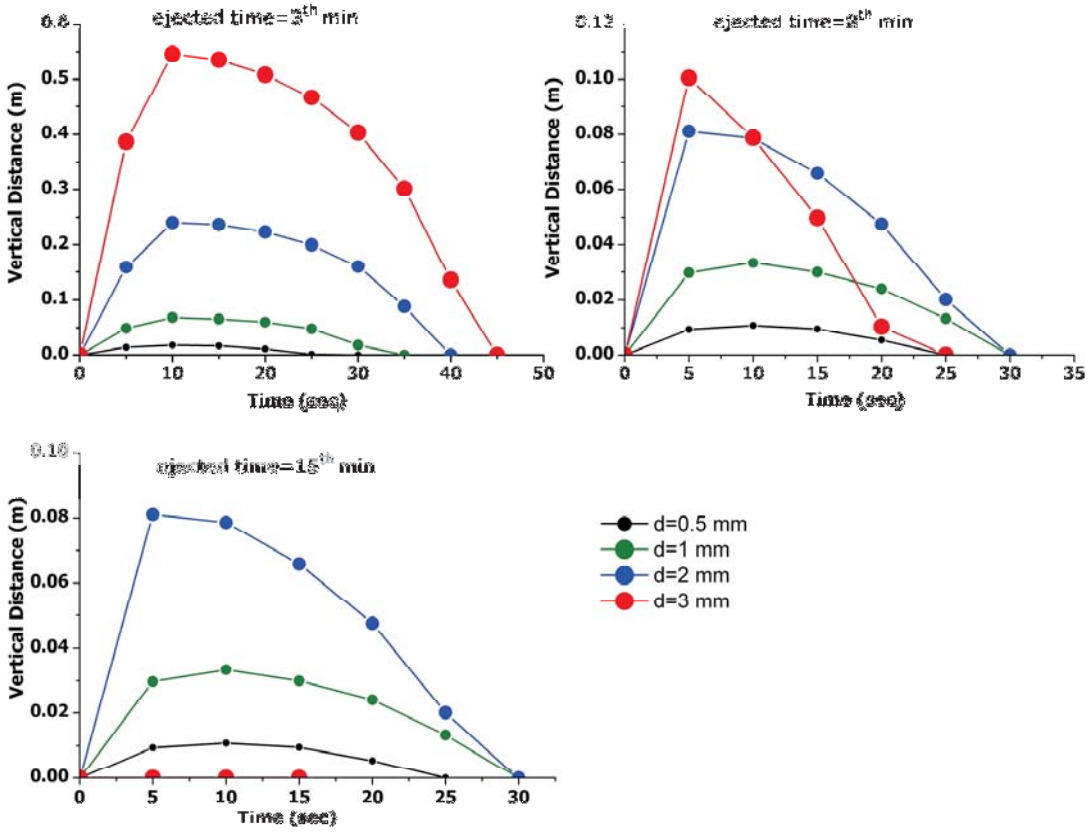

Fig. 8: Behavior of droplets ejected at different times predicted by the model

Industrial Applications

The global model proposed that the bloating behavior of droplets is crucial to understand the overall kinetics of the process because it enhances the decarburization rates in the emulsion. A global model coupled with a dense droplet assumption would not be able to predict the decarburization rates in the emulsion phase because they decreased from 300 to 50 kg/min for dense droplets. The author would expect that the decarburization rate via dense droplets would be slow which would in turn influence the overall reaction kinetics.

The bloating behavior of droplets was represented by measuring the residence time of droplets in the slag-metal-gas emulsion. By comparing the residence time predictions with the practical estimates shown in Table 2, it can be seen that the bloated droplet motion model provides reasonable predictions for the residence times of droplets in the slag-metal-gas emulsion. The difference may be due to the assumptions of the effect of gas fraction and drop size distribution. It should be noted that further industrial trials are required to verify these findings.Better understanding of the evolution of the gas in the slag-metal-gas emulsion and including the drop size distribution to the model will strengthen the predictions of the model significantly. Incorporating new theories into the process modelling of oxygen steelmaking should improve control and help in designing new technologies.

Tab. 2: A comparison of the global model using bloated droplet theory predictions with plant measurements/predictions, and a numerical model on the residence time of droplets in slag in top blown oxygen steelmaking

Investigators	Methods	Residence Time (s)
Schoop *et al*. (1978)	Indirect plant measurement from which the residence time was calculated based on the chemical analysis and kinetics model	~60
Price (1974)	Plant measurement with radioactive gold isotope tracer technique	120±30
Kozakevitch (1969)	Predictions based on carbon and phosphorus contents in metal droplet from plant measurement	60 to 120
Brooks *et al*. (2005)	Predictions using bloated droplet motion model on slag-metal-gas emulsions with 15 % FeO and gas volume fraction less than 85 %	20 to 80
Present work	Predictions using bloated droplet motion model on slag-metal-gas emulsions with 14-30 % FeO and gas volume fraction of 80 %	0.4 to 45

Conclusion

The influences of the bloating behaviour on the residence time of droplets were analysed in this study. The following conclusions can be drawn.

1. The proposed model provides information about variations in the residence time of ejected metal droplets. For bloated droplets, it is predicted that their residence time in emulsion is around 45 s during the blow. Towards the end of the blow, the residence time of droplets decreases to 0.4 s.

2. The carbon content of the metal and the volume fraction of gas in the emulsion have been shown to be of prime importance in determining the residence time of metal droplets. In general, an increase in the carbon content or decrease in the gas fraction increases the residence time of droplets. This model suggests that the residence times of metal droplets in the early blow have the highest values during the blow.

3. The maximum reaction rates in the emulsion can be achieved by the generation of smaller droplets.

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Brief Biography of Presenter

Neslihan Dogan is currently working at University of Wollongong as Associate Research Fellow. She finished her PhD in Chemical & Materials Engineering at the Swinburne University of Technology, studying modelling of oxygen steelmaking process. Neslihan's research is focused on improvements to oxygen steelmaking processes, whether using modelling or experimental techniques. The research is part of her PhD project.