

Inclusion Agglomeration - a Key Issue Contributing to 'Inclusion Engineering' in Advanced Steels

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# Inclusion agglomeration - a key issue contributing to 'Inclusion Engineering' in advanced steels

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### Abstract

Better understanding of the agglomeration behavior of non-metallic inclusions in the steelmaking process is of vital importance to control the cleanliness of the steel product. Less inclusions in the steel ingot are always desired by the steelmakers to reduce the risk of engineering problem, e.g. nozzle clogging and improve the final product's mechanical properties. On the other hand, non-agglomerated inclusions with fine size can be utilized in different processes to optimize the microstructure and improve the final property. Both aspects correspond to the concept of 'Inclusion Engineering'. In this work, the motion and agglomeration behaviors of inclusions at the interfaces between steel/gas and steel/slag are overviewed, and previous results in the open literature including the in-situ characterization as well as theoretical development are summarized. Besides, the force balance model describing the inclusion motion (passing/remaining/oscillation) at the steel/slag interface has also been briefly overviewed. The present authors' work is also highlighted. This work aims to provide a mini overview of the agglomeration behaviors of inclusions at different stages and provide a way forward to the future research of this important phenomenon contributing to 'Inclusion Engineering' in steels.

Key words: Non-metallic inclusion, agglomeration, steels, interfacial phenomenon, inclusion engineering

### **1. INTRODUCTION**

Nature and quantity of the non-metallic inclusions formed dictate much of the refining process and if they are not removed profoundly and completely, especially for the case of large size inclusions, they will affect the physical properties of the solidified steels and alloys, and lead to the degradation of mechanical properties of final products. Normally, steelmakers aim to produce clean steel with less inclusions in order to reduce engineering problems, e.g. nozzle clogging [1, 2]. The concept of 'inclusion engineering' has been applied in the field of ferrous process metallurgy, which deals with the control of the amount, morphology, size distribution, and composition of non-metallic inclusions formed in liquid metal during refining and solidification. Agglomeration of non-metallic inclusions is one of the key issues for clean steel production. Specifically, argon bubbling is considered an important method for removing non-metallic inclusions from liquid steel, because the flotation of inclusions is enhanced by adherence to bubbles. Moreover, argon bubbling can create turbulent eddy flow from bath stirring, which improves inclusion agglomeration. Therefore, the agglomeration behavior of non-metallic inclusions at the steel/Ar interface needs to be well understood and controlled. Besides, the inclusion motion behaviors at the steel/slag interfaces, e.g. agglomeration, detachment, draining the interface, etc. is another scientific issue which is not so clear so far. Last but not the least, the inclusion motion and agglomeration behavior in the bulk steel may not be exactly the same as its behaviors at different interfaces, especially for the liquid phase inclusions, since the mechanisms may not be the same, which needs to be considered critically. This work provides a mini-survey of the above-mentioned theoretical issues, and will contribute to guide the further study of inclusion agglomeration in steels.

### 2. INCLUSION AGGLOMERTION AT STEEL/AR & STEEL/SLAG INTERFACES

Argon bubbling is considered as an important method for removing non-metallic inclusions from liquid steel. This is due to the flotation of inclusion particles could be enhanced by the adherence to bubbles. Moreover, argon bubbling can create a turbulent eddy flow from bath stirring, which could improve the potency of inclusion agglomeration. According to this, the agglomeration behavior of non-metallic inclusions at the steel/Ar interface needs to be understood and controlled, aiming for the clean steel production. High temperature confocal laser scanning microscopy (HT-CLSM) is one option to create the steel and Ar interface in the gold coated chamber, and observe the inclusion agglomeration behaviors in real time, to simulate the motion behaviors of inclusions during the Ar bubbling process. Besides the experimental study, a capillary force model considering the interaction energy between two particles has been established to estimate the inclusion agglomeration potency. This model was originally derived from colloid chemistry field, and has been subsequently applied in process metallurgy [4-11]. Key equations are summarized in Eqs. [1] to [3], different research activities in previous studies are summarized in Table I.

$$\Delta W = -\pi \gamma \sum_{k=1}^{2} (Q_k h_k - Q_{k\infty} h_{k\infty}) \left( 1 + O(q^2 R_k^2) \right)$$
[1]

$$F = \frac{d(\Delta W)}{dL}$$
[2]

$$F = \frac{2\pi\gamma Q_1 Q_2 (1 - q^2 L^2)}{L} \quad (r_k \ll L)$$
[3]

In this series equations, W is the capillary interaction energy between two spherical inclusions with a separation distance L.  $Q_k$  and  $Q_{k\infty}$  are the effective capillary charges for the separation distance L and infinity, k equals to 1, 2. Meanwhile,  $h_k$  and  $h_{k\infty}$  represent height differences of the meniscus at the separation distance of L and infinity.  $\gamma$  is the surface tension of the liquid steel, O(x) is the zero function of the approximation, a revision has been made in Refs. [6, 7], and q is the ratio of the density of inclusion divided by that of steel. Capillary force (F) can be further calculated by the deviation of  $\Delta W$  with each distance (L). Schematic illustration of this model is seen in Fig.1 (a), details of the model description can be seen in Refs. [4-7]. Fig.1 (b) presents a typical result to estimate the agglomeration potency of different types of inclusions.

Table I.	Summary of theoretical	I research of inclusion	agalomeration at	the steel/Ar and a	staal/slag interfaces
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Name [Ref.]	Inclusion type	Steel grade (interface)	Key research issues
Nakajima &	Al <sub>2</sub> O <sub>3</sub> -CaO-	6Cr Al-Si killed steel	Model firstly applied in metallurgy, distinguish L/S
Mizoguchi [4]	MgO/SiO <sub>2</sub>	(Steel/Ar)	inclusion.
Wikström et al.	CaO-Al <sub>2</sub> O <sub>3</sub>	High C steel	Apply the model to predict inclusion motion at
[5]	CAS slag	(Steel/Slag)	steel/slag interface.
Mu et al. [6, 7]	Al <sub>2</sub> O <sub>3</sub> ,	Inclusion-engineered	Revised model, quantitative to predict different
Mu & Xuan [8]	Ti-Al-O	steel (Low C) (Steel/Ar)	inclusions considering physical properties.
Qiu et al. [9-11]	$Ce_2O_3$	20Cr stainless steel	Sub-particle model considering arbitrarily shape
		(Steel/Ar)	inclusions considering planar/undulating contact
			lines. Prediction of meniscus profile is also provided.



Figure 1. (a) Schematic illustration of capillary force model of inclusion agglomeration at the steel/Ar interface [6], as well as the quantitative comparison results [7].

For this type of research, the capacity of model prediction has been extensively validated by the HT-

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CLSM data of Al<sub>2</sub>O<sub>3</sub> [6], Ti-Al-O [8], CaO-Al<sub>2</sub>O<sub>3</sub> [5], semi-solid oxide (CAS) [4], and Ce<sub>2</sub>O<sub>3</sub> [9], a close agreement shows the model reliability. Nakajima and Mizoguchi [4] applied this model to the steelmaking field, and made the first attempt to quantitatively predict the agglomeration/repulsion behaviours of different phases (solid, liquid, semi-solid) of inclusions in pairs. The model has been continuously revised by considering different types of inclusions by using the physical properties (contact angle, surface tension, interfacial energy, etc.) [6-8], and arbitrary shapes of inclusions by treating the particles as capillary 'charges' with planar contact lines or as the capillary 'multipoles' with undulating contact lines through the analogy with 2D electrostatics [9-11]. In addition, this capillary force model could also be used to predict the driving force during the bending of chain aggregate inclusions. There are still various issues have not been fixed and could be considered in the future work. For instance, different types inclusion behaviors in other advanced steel grades, e.g. 3<sup>rd</sup> AHSS grade including medium Mn steels, tool steels, could be considered. The current research mainly focuses on the steel/Ar interface, comprehensive studies considering various slag systems could also be investigated. Furthermore, connecting the current physical model with the thermodynamic database considering the evolution of thermo-physical properties during the metal and slag interactions could be another direction.

# 3. INCLUSION MOTION BEHAVIOR AT STEEL/SLAG INTERFACE

To efficiently remove inclusions during steelmaking, the floated inclusions will pass through the steel/slag interface and dissolve in the liquid slag. In this case, the motion of inclusion particles at the steel/slag interface is the key procedures. Nakajima and Okamura [13] developed a theoretical model to evaluate the inclusion displacement at the steel-slag interface, the physical properties, e.g. inclusion and slag viscosity were considered. Based on this model, Strandh and co-workers investigated the solid [14] and liquid [15] inclusion motion (passing/remaining/oscillation) at the steel-slag interfaces. Subsequently, Liu and co-workers [16] considered the influence of Reynolds number into Nakajima model to investigate the influence of inclusion size, interfacial tension, slag viscosity, etc.

Very recently, Xuan and co-workers [17] proposed a revised mathematical model which assumed that the deformable thin film could exist if the liquid inclusion/bubble approached to the steel-slag interfaces. All the parameters in this model were derived from a vacuum degassing (VD) trial from industry. The predicted critical size of liquid inclusion for detaching is validated by means of experimental analysis of inclusion size density. The new model could also point out that the thin-film drainage stage was the main stage of inclusion detachment which is not considered in the previous models [13-16]. It is found that the terminal velocity of inclusion has a vital role in the inclusion detachment process, and a small slag surface tension is favourable to aid in the liquid inclusion detachment, according to Xuan's model. The schematic illustration of each model describing thin film drainage, steel droplet formation when the gas bubble rupture at the steel-slag interface, as well as the film rupture, where the steel/slag interface is simplified as flat is shown in Figure 2. For the future work, a comprehensive model incorporating the inclusion motion at the steel/slag interface as well as its dissolution in the slag is needed for considering the whole process.



Figure 2. Schematic illustration of (a) film drainage, (b) bubble, and (c) film rupture models in Ref. [17].

# 4. INCLUSION AGGLOMERATION AT THE BULK OF STEEL

Besides the inclusion motion and agglomeration at the steel/Ar and steel/slag interfaces, the inclusion agglomeration in the bulk of steel is also a vital topic, dominant agglomeration force during cluster formation by inclusions include van der Waals Force and cavity bridge force due to the un-wetting behavior [18, 19]. A comparison between agglomeration force at the steel/Ar interface as well as in the

bulk has been proposed [20], the force exerted on the inclusions in the steel bulk a few magnitudes higher than that at the steel/Ar interface. Detailed comparison will be performed in the future work. In addition, advanced techniques like X-ray CT [21] could be useful for detecting the real morphology of agglomerated inclusions in the steel bulk.

### **5. SUMMARY**

A mini-review of different theoretical models to predict inclusion motion and agglomeration behaviors at different interfaces have been made. This work will shed light on the future development of theoretical studies of inclusion engineering towards the clean steel production.

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#### REFERENCES

- J.H. Park, L. Zhang. Kinetic modeling of non-metallic inclusions behavior in molten steel: A review. Metall. Mater. Trans. B, 51(2020), pp.2453-2482.
- [2] J.H. Park, Y. Kang. Inclusions in stainless steels-a review. steel research international, 88(2017), p.1700130 (25 pages).
- [3] L. Holappa, O. Wijk. Inclusion engineering. Treatise on Process Metallurgy, Elsevier Ltd. (2014), pp.347-372.
- [4] K. Nakajima, S. Mizoguchi. Capillary interaction between inclusion particles on the 16Cr stainless steel melt surface. Metall. Mater. Trans. B, 32(2001), pp. 629-641.
- [5] J. Wikström, K. Nakajima, H. Shibata, A. Tilliander, P. Jönsson. In situ studies of agglomeration between Al<sub>2</sub>O<sub>3</sub>-CaO inclusions at metal/gas, metal/slag interfaces and in slag. Iron. Steelmak., 35(2008), pp.589-599.
- [6] W. Mu, N. Dogan, K.S. Coley. Agglomeration of non-metallic inclusions at steel/Ar interface: In-situ observation experiments and model validation. Metall. Mater. Trans. B, 48(2017), pp. 2379-2388.
- [7] W. Mu, N. Dogan, K.S. Coley. Agglomeration of non-metallic inclusions at the steel/Ar interface: model application. Metall. Mater. Trans. B, 48(2017), pp. 2092-2103.
- [8] W. Mu, C. Xuan. Agglomeration mechanism of complex Ti-Al oxides in liquid ferrous alloys considering hightemperature interfacial phenomenon. Metall. Mater. Trans. B, 50(2019), pp. 2694-2705.
- [9] Z. Qiu, A. Malfliet, B. Blanpain, M. Guo. Capillary interaction between arbitrarily-shaped inclusions at the gas/steel interface. Metall. Mater. Trans. B, 53(2022), pp. 1894-1903.
- [10] Z. Qiu, A. Malfliet, B. Blanpain, M. Guo. Capillary interaction between micron-sized Ce<sub>2</sub>O<sub>3</sub> inclusions at the Ar gas/liquid steel interface Metall. Mater. Trans. B, 53(2022), pp. 1775-1791.
- [11] Z. Qiu, A. Malfliet, M. Guo, B. Blanpain. A sub-particle model for capillary interaction between arbitrarily shaped nonmetallic inclusions with an undulated contact line. Metall. Mater. Trans. B, 53(2022), in press, <u>https://doi.org/10.1007/s11663-022-02608-0</u>.
- [12] W. Mu, N. Dogan, K.S. Coley. In situ observation of deformation behavior of chain aggregate inclusions: a case study for Al<sub>2</sub>O<sub>3</sub> at a liquid steel/argon interface. J. Mater. Sci., 53(2018), pp.13203-13215.
- [13] K. Nakajima, K. Okamura. Inclusion Transfer Behavior Across Molten Steel-Slag Interface, Proc. of 4<sup>th</sup> Int. Conf. on Molten Slags and Fluxes, ISIJ, Tokyo, 1992, pp. 505-510.
- [14] J. Strandh, K. Nakajima, R. Eriksson, P. Jönsson. Solid inclusion transfer at a steel-slag interface with focus on tundish conditions. ISIJ Int., 45(2005), pp. 1597-1606.
- [15] J. Strandh, K. Nakajima, R. Eriksson, P. Jönsson. A mathematical model to study liquid inclusion behavior at the steel-slag interface. ISIJ Int., 45(2005), pp. 1838-1847.
- [16] C. Liu, S. Yang, J. Li, L. Zhu, X. Li. Motion behavior of nonmetallic inclusions at the interface of steel and slag. Part I: Model development, validation, and preliminary analysis. Metall. Mater. Trans. B, 47(2016), pp.1882-1892.
- [17] C. Xuan, E.S. Persson, R. Sevastopolev, M. Nzotta. Motion and detachment behaviors of liquid inclusion at molten steel-slag interfaces. Metall. Mater. Trans. B, 50(2019), pp. 1957-1973.
- [18] C. Xuan, A.V. Karasev, P.G. Jönsson, K. Nakajima. Attraction force estimations of Al<sub>2</sub>O<sub>3</sub> particle agglomerations in the melt. Steel Res. Int., 88(2017), p.1600090 (8 pages).
- [19] K. Sasai. Interaction between alumina inclusions in molten steel due to cavity bridge force. ISIJ Int., 56(2016), pp.1013-1022.
- [20] D. Kumar, M. Ferriera, P.C. Pistorius. Comparing agglomeration behaviour of inclusions observed on steelargon interface in confocal laser scanning microscope experiments and in bulk steel. The 3rd International Conference on Science and Technology of Ironmaking & Steelmaking, Kanpur, India, 2017, pp. 415-418.
- [21] D. Kumar, R. Cunningham, P.C. Pistorius. Use of X-ray microtomography to determine volume fraction and 3D morphology of inclusion clusters in steel, AISTech 2018, pp. 1493-1500.