Influence of rigid body motion on the attachment of metallic droplets to solid particles in liquid slags – a phase field study

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Abstract

Metallic droplets can remain attached to solid particles within liquid slags. This will result in production losses in several pyrometallurgical industries. This study shows the extension of a recently developed phase field model to include movement of the solid particles in the liquid slag in a system considering the attachment of liquid metal droplets to solid particles in slags. The influence of this movement on the wetting of the metal droplets to the solid particles in the slag and on the resulting microstructures is investigated as a function of the velocity of the particles. For all wetting regimes, the apparent contact angle in the final microstructures was clearly larger than without particle movement. For the amount of metal attached to the particle, a clear trade-off was found between the speed of motion of the solid particle and the wetting regime.

Keywords

Phase field model; Metallic droplets; Spinodal decomposition; Random initialization method; Rigid body motion

1 Introduction

Nowadays, a higher demand for a lot of metals exists, but the quantity and purity of the ores decreases. The amount of scrap, on the other hand, increases and thus, recycling becomes

more important (Hagelüken 2006). Besides recycling, it is also necessary to improve and optimize existing processes in extractive and recycling metallurgy. One of the main difficulties in the overall plant efficiency, after the final phase separation step, in both primary and secondary metal production are metal losses in slags. The separation between the slag and matte/metal phase after sedimentation is never perfect, resulting in the loss of valuable metals in the slag and/or the need for reprocessing of this slag phase. Metal/matte losses in slags are a widely occurring problem in different metal industries including industrial Cu smelters (Yannopoulos 1970), Pb reduction melting furnaces (Calvo and Ballester 1986a; b), ferrochrome smelters (Nkohla 2006).

Generally, metal losses in slags can be subdivided into two types: chemical losses and physical or mechanical losses (Yannopoulos 1970). Chemical losses refer to the dissolved copper in the slag and are thermodynamically determined (Nagamori 1974; Toguri and Santander 1972; Wang et al. 1974; Yazawa 1974). Mechanical losses are entrained copper droplets which do not settle and do not end up in the underlying matte or metal phase. In general, there are four main reasons for mechanical entrainment of metals in slag (Choo and Toguri 1993; Ip and

Toguri 1992):

- i) entrainment due to furnace charging or slag tapping;
- ii) precipitating metal from slag due to temperature fluctuations or chemical reactions;
- iii) dispersion of matte or metal into the slag phase due to gas bubbles, originating from reactions, when these bubbles cross the slag-matte/metal interface;
- iv) matte/metal droplets are attached to solid particles in the slag phase. The first three reasons have been extensively studied in literature.

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The fourth reason, however, has only recently drawn attention [\(De Wilde et al. 2015, 2016b\)](#)~~(De Wilde et al. online, 2015)~~, ~~in a more f~~undamental research way was executed to obtain knowledge on the mechanisms responsible for this interaction, as an increased understanding of the fundamental mechanisms governing these losses could help to further improve production efficiencies.

The interfacial energies played an important role in this interaction, according to the first experiments (De Wilde et al. 2015). In a next stage, two possible reaction paths for the origin of the attachment of the metal droplets to the solid particles were proposed: a non-reactive and a reactive pathway. (De Wilde et al. 2016b; a; Bellemans et al. 2017) In the non-reactive pathway, the metal droplet and spinel particle can get attached to each other due to the significant mixing taking place during pyrometallurgical operations. For the reactive origin, a reaction scheme was proposed, where the spinel solids form simultaneously with the copper droplets or form around the copper droplets, depending on the local conditions of the system. The interplay between the solid particles and metal droplets can be investigated by a combination of experiments and modelling to increase its understanding. The phase field method was already used to model various systems considering microstructural evolution, e.g. during solidification (Heulens et al. 2011; Karma and Rappel 1998), solid-state phase transformations (Chen 2002) and solid-state sintering (Wang 2006).

A phase field model was already developed in previous work (Bellemans et al. 2015b; a) to simulate growing and evolving metal droplets in a liquid oxide phase with solid particles dispersed in the slag. Several observations could be made with this model:

- Four wetting regimes of the metal on the solid particle can be distinguished: no wettability, low wettability, high wettability and full wettability (Bellemans et al.

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2015b). It was also shown that the contact angles measured from the simulated microstructures agree well with the contact angles expected from Young's law.

- Comparison to an experiment with a synthetic slag system (De Wilde et al. 2015) shows good agreement of the experimental system with the low wettability regime (Bellemans et al. 2015a).
- Two initialization methods (corresponding to two possible origins proposed based on experiments: reactive versus non-reactive) were compared: a spinodal initialization and a random initialization (Bellemans et al. 2016).

However, in real slags, convection occurs, resulting in the movement of the particles with respect to the two fluids, which is not considered so far in the model. An experimental study of the effect of the movement of the particles on the sticking behaviour of metallic droplets would be intricate. Therefore, in this study, the phase field model is extended to take into account rigid body motion of the solid particles in the liquid slag. The simulations can indicate possible microstructural effects induced by the relative movement of the particles in the slag.

Numerical calculation of the solution of the Navier-Stokes equations for fluid flow is computationally much more expensive than the numerical solution of the current phase field equations. Furthermore, our main interest is to understand possible effects on the wetting behaviour coming from a relative movement of the particles with respect to the metal droplets, and not so much to predict the slag flow. Therefore, instead of solving the complex Navier-Stokes equations, an equation to move the phase field profiles representing the particle in space is added to the model, resulting in a displacement with a certain velocity of the particles with respect to the slag and the metal droplets. The influence of this rigid body motion on the wetting regimes, as determined in a previous study (Bellemans et al. 2015b), as well as on the resulting microstructures is then investigated from simulations based on this

model. The extension of the model is first discussed in section 2. However, the reader is referred to (Bellemans et al. 2015b) for the detailed description of the model. Section 3 describes the simulation parameters, the numerical implementation and the initialization methods. Finally, the simulation results are presented and discussed in section 4.

2 Model

The model considers a hypothetical binary oxide-metal (O-M) system. The phase field variables describe a microstructure with:

- a liquid ($\phi = 0$) metal phase with certain mole fraction of M $x_M = x_{eq,LM}$
- a liquid ($\phi = 0$) oxide phase with $x_M = x_{eq,LO}$
- a solid ($\phi = 1$) with a constant composition ($x_M = x_s$) and which does not react with the liquid phases

Other specific characteristics of the model can be found in (Bellemans et al. 2015b; a, 2016).

The evolution of the microstructure, and thus of the variables ϕ and x_M , is described by the following differential equations, which represent essentially minimization of the Gibbs free energy of the system:

$$\frac{\partial x_M}{\partial t} = \nabla \cdot [M \nabla [(1 - h(\phi))f'_{Liquid}(x_M) + h(\phi)f'_{Solid}(x_M) - \kappa_{x_M} \nabla^2 x_M]] \quad (1)$$

$$\frac{\partial \phi}{\partial t} = -L \frac{\delta F}{\delta \phi} = -L [W g'(\phi) + h'(\phi)(f_{Solid} - f_{Liquid}) - \kappa_\phi \nabla^2 \phi] \quad (2)$$

M ($\text{m}^5/(\text{Js})$) and L ($\text{m}^3/(\text{Js})$) are kinetic coefficients, which are related to the interdiffusion coefficient D (m^2/s) of the liquid ($D=A_{sp}M$) and the atomic velocity over an interface, respectively. κ_{x_M} and κ_ϕ are the coefficients in front of the gradient terms and can be linked to the interface energies and interface widths for the liquid-liquid and solid-liquid interfaces, respectively. $h(\phi)$ is a function to interpolate between the bulk free energies of the liquid and solid phases, i.e. f_{Liquid} and f_{Solid} , respectively. The former is described by a polynomial

of the fourth order for which A_{sp} represents the curvature. The latter is described by a parabolic equation of which the position of the minimum is described by x_s (constant composition of the solid particle) and A_s (curvature of the free energy). $g(\phi)$ is a double well function, which ensures the co-existence of both the solid and liquid phases. Here, W represents the depth of the double well. This parameter and the curvatures of the bulk free energy densities are also linked to the interface widths and energies. More details regarding the meaning of all parameters can be found in (Bellemans et al. 2015b; a, 2016).

This study considers an extension of the existing phase field model to include rigid body motion with a velocity (m_x, m_y) of the solid particles with respect to the liquid. For this, an equation is added to move the phase field profiles in space, resulting in the displacement of the particle with respect to the slag:

$$\frac{\partial \phi}{\partial t} = -\nabla \cdot [m_x h(\phi), m_y h(\phi)] \quad (3)$$

This equation represents the movement of the particle, where m_x and m_y determine the speed at which this happens. The equation is very similar to the equations used in the level-set method (Osher and Fedkiw 2001; Osher and Sethian 1988; Sethian and Smereka 2003) and moves the ϕ -profile, after which the x_M adjusts itself to the new ϕ -profile.

3 Simulation set-up

3.1 Model parameters

The parameter values in (Bellemans et al. 2015b; a, 2016) were used again in this study. The initial study of the influence of the interfacial energies showed that different wetting regimes can be reached by changing the parameter x_s , i.e. the constant composition of the solid particle (Bellemans et al. 2015b). Changing x_s resulted in a variation of the solid-liquid interface energies and thus in the wetting regime, as represented by Young's equation

$$\gamma_{S,LO} = \gamma_{S,LM} + \gamma_{LO,LM} \cos \theta \quad (4)$$

The following x_s -values were investigated in our previous study: 0.50; 0.60; 0.70; 0.74; 0.80 and 0.90. The corresponding interfacial energies and contact angle predicted by Young's equation (4) are tabulated in [Table 1](#).

x_s	$\gamma_{S,LO}$ (N/m)	$\gamma_{S,LM}$ (N/m)	$\gamma_{LO,LM}$ (N/m)	$\theta_{\text{predicted}}$ (°)	Wetting regime	θ_{measured} (°)
0.5	1.2500	2.8140	0.903	/	Non-wetting	164.20±5.22
0.6	1.2641	2.0260	0.903	147.54	Low wettability	148.05±9.21
0.7	1.3631	1.5604	0.903	102.62	Low wettability	104.31±8.90
0.74	1.4455	1.4455	0.903	90.00	Low/high wettability limit	87.09±2.73
0.8	1.6318	1.3325	0.903	70.64	High wettability	full wetting
0.9	2.1551	1.2572	0.903	6.09	High wettability	full wetting

TABLE 1 THE VARIATION OF x_s RESULTS IN A VARIATION OF THE INTERFACIAL SOLID-LIQUID ENERGIES AND CONSEQUENTLY OF THE PREDICTED CONTACT ANGLE. THE COLUMN ON THE RIGHT SHOW THE MEASURED CONTACT ANGLES FROM PREVIOUS SIMULATIONS (BELLEMANS ET AL. 2015B) WITH $x_i = 0.63$ WITHOUT RIGID BODY MOTION OF THE SOLID PARTICLE

The model was extended to consider rigid body motion of the solid particles with a certain velocity m , which was kept small in the first part of this study: the solid particle moved $1 \Delta x$ per 10 s ($1.265 \cdot 10^{-8}$ m/s) in the x-direction (the velocity in the y-direction was 0 m/s). To apply the rigid body motion equation, very small time steps are needed, for which a sub-cycling implementation was used (i.e. for every phase field time step, 100 rigid body motion time steps passed).

Afterwards, the solid particle velocity m , was varied: the solid particle moved with either $5 \Delta x$ per 10 s ($6.32 \cdot 10^{-8}$ m/s) or $10 \Delta x$ per 10 s ($1.26 \cdot 10^{-7}$ m/s). As a reference, the characteristic diffusion distance is $\sqrt{Dt}$ (Depover et al. 2016) and can be considered as a measure for the distance the metal can diffuse during the simulation. In this case, i.e. for 100 s, this distance equals $6.32 \cdot 10^{-5}$ m, which results in an average speed of $6.32 \cdot 10^{-7}$ m/s.

3.2 Numerical implementation

The evolution equations describe the change of the variables in time. This change is calculated with the semi-implicit Fourier spectral method (Chen and Shen 1998). The model was

implemented in Matlab (*Matlab 7.12.0 (R2011a)* 2011). One simulation for system with size $[256 \times 256 \times 1]$ and 10^6 time steps, typically took 6 to 7 days to be completed on an Intel® Core™ i7-3610QM CPU @ 2.30GHz processor with 8Gb RAM.

To record a movement of the solid particle, very small time steps are required, but to reduce the computational time, this equation is solved using a different time step than the one used to solve the other evolution Eqs. (2) and (3): for every phase field time step, 100 rigid body motion time steps passed.

During the first simulations, it became clear that this implementation indeed resulted in a solid particle shifting, in the present case, towards the right-hand side of the system. However, the diffusive particle-liquid interface was not shifted along with the particle.

Resulting in numerical artefacts in the microstructures such as the one illustrated in Fig. 1.

Fig. 1 Illustration of loss of correct diffuseness for a simulation with spinodal initialization, $x_1 = 0.63$ and $x_2 = 0.70$ (low wetting regime) with a zoomed-in inset

On the right side of the particle, a sharp edge is formed, whereas on the left side of the particle, the diffuse remains of the particle do not move along with the rest of the particle. This is a clear anomaly as opposed to the simulations without rigid body motion. As mentioned previously, during the first 1000 time steps, a larger kinetic coefficient of $L_{initial} = 10^{-7} \text{ m}^3/(\text{J s})$ for the phase field evolution was used, in contrast to $L = 10^{-30} \text{ m}^3/(\text{J s})$ in the rest of the simulation, to allow the boundaries of the particle to become diffuse. To obtain a

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correct diffuse interface profile at all particle interfaces, L was taken equal to $L_{initial}$ every 100th time step over the complete simulation. This results in the **microstructure image as shown in**

Fig. 2.

FIG. 2 MICROSTRUCTURE WHEN $L_{initial}$ IS ALSO USED EVERY 100th TIME STEP TO OBTAIN CORRECT DIFFUSENESS OF THE INTERFACES FOR A SIMULATION WITH SPINODAL INITIALIZATION, $x_i = 0.63$ AND $x_e = 0.70$ (LOW WETTING REGIME).

The diffuseness at the edges is better, i.e. the right edge is no longer sharp and on the left edge, the diffuse remains of the particle from previous time steps are no longer present. It is, however, clear that the corners of the rectangular particle become more blunted by this action. The change in area for the simulation in which L is taken equal to $L_{initial}$ every 100th time step is 1.51 % (= 100 * (5000 - A_{final}) / 5000, with A obtained by summing ϕ over all grid points and multiplying with $(\Delta x)^2$), whereas the change in area is 0.12 % if $L_{initial}$ is only used in the first 1000 time steps. As an area reduction of 1% probably has negligible influence on the results, we chose to implement the abovementioned use of $L_{initial}$ in every 100th time step. However, after every simulation, special care was taken to investigate the change in area of the solid particle to assure that it did not shrink too much. The change in area of the solid particle (over the complete simulation) is expressed as the relative error (= 100 * | $A_{with\ extra\ L_{initial}} - A_{without}$ | / $A_{without}$) in **Table 2** for the simulations with the spinodal initialization and $x_i = 0.605$; 0.63 and 0.65, respectively, and for all the x_s -values. The error is mostly of the order of 1% and thus relatively low.

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x_s	$x_i=0.605$	$x_i=0.63$	$x_i=0.65$
0.5	1.20	1.28	1.40
0.6	0.41	1.26	1.28
0.7	1.95	1.40	1.39
0.74	2.10	1.66	1.43
0.8	2.11	2.02	1.80
0.9	1.52	1.52	1.51

TABLE 2 RELATIVE ERROR [%] OF THE CHANGE IN AREA OF THE SOLID PARTICLE FOR THE DIFFERENT x_i, x_s -COMBINATIONS

3.3 Initialization

The model had the possibility to use two different initialization methods, which are described in detail in (Bellemans et al. 2015b, 2016):

- The spinodal initialization introduces metal droplets in the system by spinodal decomposition of an initially supersaturated ~~the~~ solution ($x_M = x_i$) into metal droplets ($x_M = x_{eq,LM}$) in a liquid oxide ($x_M = x_{eq,LO}$). x_i values of 0.605, 0.63 and 0.65 were considered for the simulations in this study. These supersaturations correspond to a volume fraction of metal of 0.219, 0.271 and 0.312, respectively.
- The random initialization randomly positions droplets ($x_M = x_{eq,LM}$) in an oxide liquid ($x_M = x_{eq,LO}$). The volume fractions of metal droplets in this study were 0.10 and 0.20, corresponding to x_i -values of 0.548 and 0.596, respectively.

3.4 Post-processing

The simulations data are processed both qualitatively and quantitatively. The former as greyscale plots of $x_M - \phi x_s$ to illustrate the microstructure. They are rescaled to [0 1] so that the values higher/lower than the maximum/minimum value are converted to the maximum/minimum, respectively. The greyscale images of the final time step (10^6) were used

to measure apparent contact angles ($^{\circ}$) with the image processing software ImageJ (Rasband 1997).

For a quantitative analysis, the different phases are defined as the connected domains where:

- $x_M - \phi x_S > 0.71$ for a metal droplet
- $0.5 < \phi$ for the particle.

The attachment of the droplet to the solid particle is investigated by extension of the two phases by 2 ~~or~~ and 3 grid point layers, [for the droplet and the solid particle](#), respectively. If the extended domains cover each other partially, the droplet is considered 'attached'. Dividing the total [area-volume](#) of attached metal by the total [area-volume](#) of metal in the system, yields the fraction of attached metal.

3.5 Accuracy

Repeating several simulations four times in previous work ($x_i = 0.63$; $x_s = 0.50$; 0.60 ; 0.70 ; 0.80 in a system with size ~~{256-x256-x1}~~) (Bellemans et al. 2015b), enabled the calculation of the standard deviation on the results, which is used for the error bars in the quantitative graphs in the results section.

4 Results and discussion

4.1 Microstructural influence of rigid body motion

The influence of the rigid body motion on the finally ~~obtained~~ microstructures [obtained](#) is illustrated in [Fig. 3](#) and [Fig. 4](#) for the spinodal and the random initialization, respectively ($x_s = 0.74$ is not shown but the behaviour is between that of $x_s = 0.70$ and $x_s = 0.80$).

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FIG. 3 THE INFLUENCE OF x_i AND x_s (WETTING REGIME) ON THE RESULTING MICROSTRUCTURES FOR SPINODAL INITIALIZATION

It should be noted that for $x_i = 0.605$ and $x_s = 0.60$, no spinodal decomposition took place, because the system can be considered as almost 'stable', as both compositions are very close to one another. The combination in this study yields an even more stable situation than for $x_s = 0.60$ and $x_i = 0.61$ in a previous study (Bellemans et al. 2015b), where it also took quite some time before spinodal decomposition occurred.

FIG. 4 THE INFLUENCE OF x_s AND f_v ON THE RESULTING MICROSTRUCTURES FOR RANDOM INITIALIZATION

In the no wetting regime, the influence of the rigid body motion seems to be twofold. As the solid particle moves, it can either have metal droplets already attached to it, due to interfacial energies, but can lose this when the droplet is attached to the side of the particle in the opposite way of the rigid body motion (i.e. here the left side of the solid particle), for example in the $x_l = 0.65$; $x_s = 0.50$ -case. However, it is also possible that a particle initially did not have

metal attached to it, but ‘meets’ a droplet on its way, which also happens in the $x_i = 0.65$; $x_s = 0.50$ -case.

From the low wettability regime onwards, the influence of the interfacial energies come into play and this results in the movement of the droplets towards the solid particle, as illustrated at the left side of the particle in the $f_v = 0.20$; $x_s = 0.60$ -case without rigid body motion. If the rigid body motion is then added, a solid particle without metal attached to it, moves towards a metal droplet, which in turn will also be slightly attracted to the solid particle due to the interfacial energies. If a solid particle has a metal droplet attached to it, no detachment from the particle was observed when the particle was moving away from the attached droplet, even though the attachment is not very strong.

At the higher wetting regimes, the influence of the interfacial energies becomes more important, i.e. attachment of metal to the solid particle is extremely favourable from an energetic point of view. This results in the droplet remaining attached to the solid particle.

4.2 Amount of attached metal

The influence of the rigid body motion on the area of attached metal is shown in the graphs in [Fig. 5](#). The simulation results (denoted ‘with’) are compared to the results from our previous study (Bellemans et al. 2015b), in which rigid body motion was not present (denoted ‘without’).

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FIG. 5 AREA ATTACHED METAL FOR SEVERAL x_i, x_s COMBINATIONS TO INVESTIGATE THE INFLUENCE OF RIGID BODY MOTION (ASSIGNED AS 'WITH')

For $x_i = 0.605$, more metal is attached to the solid particle when no rigid body motion is present in the system, except for $x_s = 0.60$. However, this finds its origin in the fact that in this study we investigated $x_i = 0.605$ instead of 0.61 as in the previous study (this $x_i = 0.605$ in turn was chosen to be able to compare to the other initialisation method with $f_V = 0.20$). Because $x_i = 0.605$ was investigated, less metal was present in the system and this also influences the amount of attached metal. This is also visible in the fact that the fraction of attached metal does not change when rigid body motion is applied to the system. Moreover, for $x_i = 0.605$ and $x_s = 0.60$, no spinodal decomposition took place, as observed previously.

For $x_i = 0.63$, no conclusive effect of the presence of rigid body motion of the solid particle on the amount of attached metal can be observed. For $x_i = 0.65$, except for the non and the full wetting case, the simulations with rigid body motion have less attached metal to the solid particle. The same comparison between presence and absence of rigid body motion was made for the random initialization, as shown in Fig. 6. In most cases, the amount of attached metal seems larger for the cases without rigid body motion.

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FIG. 6 AREA ATTACHED METAL IN SEVERAL fV, x_s COMBINATIONS TO INVESTIGATE THE INFLUENCE OF RIGID BODY MOTION (ASSIGNED AS 'WITH')

4.54.3 Contact angle

Another quantitative measure of the influence of the rigid body motion on the microstructure can be found in the apparent contact angle. A comparison of the mean contact angles for situations with and without rigid body motion are tabulated in [Table 3](#) and [Table 4](#), for the spinodal and random initializations, respectively.

x_s	$\theta_{\text{predicted}} (^{\circ})$	$\theta_{x_i=0.61} (^{\circ})$ without	$\theta_{x_i=0.61} (^{\circ})$ with	$\theta_{x_i=0.63} (^{\circ})$ without	$\theta_{x_i=0.63} (^{\circ})$ with	$\theta_{x_i=0.65} (^{\circ})$ without	$\theta_{x_i=0.65} (^{\circ})$ with
0.5	/	/	/	164.2±5.2	/	/	150.2±3.9
0.6	147.5	152.8±2.3	/	148.1±9.2	158.1±10.7	155.3±7.2	143.3±7.8
0.7	102.6	103.3±5.7	105.5±16.3	104.3±8.9	123.8±16.2	111.8±2.7	115.0±9.4
0.74	90.0	full wetting	73.5±2.8	87.1±2.7	114.1±27.3	85.4±5.7	112.1±18.3
0.8	70.6	full wetting	full wetting	full wetting	55.1±4.5	70.4±4.1	81.8±5.9
0.9	6.1	full wetting	full wetting	full wetting	full wetting	full wetting	full wetting

TABLE 3 COMPARISON OF THE PREDICTED AND MEASURED CONTACT ANGLES ($^{\circ}$) OF THE METALLIC DROPLETS ATTACHED TO THE SOLID PARTICLE FOR THE SPINODAL INITIALIZATION

x_s	$\theta_{\text{predicted}} (^{\circ})$	$\theta_{fV=0.1} (^{\circ})$ without	$\theta_{fV=0.1} (^{\circ})$ with	$\theta_{fV=0.2} (^{\circ})$ without	$\theta_{fV=0.2} (^{\circ})$ with
0.5	/	/	/	/	/
0.6	147.5	146.9±7.6	/	128.9±2.7	160.2±2.3
0.7	102.6	111.4±38.5	/	96.1±10.4	/
0.74	90.0	102.2±7.6	/	79.5±38.5	98.6±13.4
0.8	70.6	73.4±15.8	/	71.2±13.2	77.4±11.0
0.9	6.1	full wetting	full wetting	full wetting	full wetting

TABLE 4 COMPARISON OF THE PREDICTED AND MEASURED CONTACT ANGLES ($^{\circ}$) OF THE METALLIC DROPLETS ATTACHED TO THE SOLID PARTICLE FOR THE RANDOM INITIALIZATION

For $x_i = 0.605$ and $x_i = 0.63$, the full wetting regime is encountered at larger x_s -values (corresponding to higher wetting regimes) when rigid body motion is present. Moreover, in most of the cases, the apparent contact angle is larger when rigid body motion is present. Except for the $x_i = 0.65$; $x_s = 0.50$ -case, which was an illustration that the rigid body motion can increase the attachment of the metal to the solid particle in the no and low wetting cases. On the other hand, a larger apparent contact angle corresponds to a lower wettability. The reason for this can be that the particle ‘pushes’ its way through the liquid metal. It should be noted that this effect is very clear, even though the speed of movement of the solid particle is rather low.

The fact that rigid body motion makes it appear that a lower wetting regime is present than expected based on the contact angles predicted by Young’s equation sheds a new light on the previous comparison of experimental observations in a PbO-FeO-CaO-SiO₂-Cu₂O-ZnO-Al₂O₃ system (De Wilde et al. 2016b)(De Wilde et al. online) to our simulations (Bellemans et al. 2015b; a). It could imply that the actual wetting regime is not a low-almost-no wetting regime, but, depending on the velocity ~~by~~with which the particles move in the liquid, it could as well correspond to a higher (but still low) wetting regime.

It is expected that the influence of the rigid body motion on the apparent contact angle disappears again once the rigid body motion is stopped. This was also checked with

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simulations without rigid body motion starting from the final situation (x_M and ϕ -values) of a simulation with rigid body motion. These simulations ran for 10^7 time steps and the measured apparent contact angles are tabulated in [Table 5](#).

x_s	$\theta_{\text{predicted}} (^{\circ})$	$\theta_{x_i=0.63} (^{\circ})$ without	$\theta_{x_i=0.63} (^{\circ})$ with	Steady state after with
0.5	/	164.2±5.2	/	/
0.6	147.5	148.1±9.2	158.1±10.7	144.4±12.8
0.7	102.6	104.3±8.9	123.8±16.2	108.0±4.4
0.74	90.0	87.1±2.7	114.1±27.3	100.3±13.0
0.8	70.6	full wetting	55.1±4.5	41.4±19.7
0.9	6.1	full wetting	full wetting	Full wetting

TABLE 5 COMPARISON OF THE PREDICTED AND MEASURED CONTACT ANGLES ($^{\circ}$) OF THE METALLIC DROPLETS ATTACHED TO THE SOLID PARTICLE FOR THE SPINODAL INITIALIZATION WITH $x_i = 0.63$ WITHOUT AND WITH RIGID BODY MOTION AND FOR A SIMULATION WITHOUT RIGID BODY MOTION, STARTING FROM THE FINAL SIMULATION STEP OF A RIGID BODY MOTION SIMULATION

It is observed that these ‘equilibration’ simulations result in contact angles that return towards the values corresponding to the motionless simulations, as expected. Only the $x_s = 0.80$ -case did not return to the full wetting situation where the solid is surrounded by a metallic droplet, as illustrated in [Fig. 7](#).

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FIG. 7 THE COMPARISON OF THE APPARENT CONTACT ANGLE BETWEEN THREE DIFFERENT CASES: (A) WITHOUT MOVEMENT OF THE SOLID PARTICLE; (B) WITH THE RIGID BODY MOTION OF THE SOLID PARTICLE; (C) AN ‘EQUILIBRATION’ SIMULATION WITHOUT MOVEMENT OF THE SOLID PARTICLE (STARTING FROM THE FINAL RESULT IN (B)).

4.84.4 Influence of the speed of the rigid body motion

The influence of the rigid body motion on the finally [obtained](#) microstructures [obtained](#) is illustrated in [Fig. 8](#).

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Wetting regime	speed	without	1Δx per 10s	5Δx per 10s	10Δx per 10s
	x_s

FIG. 8 THE INFLUENCE OF THE SPEED OF MOTION OF THE SOLID PARTICLE AND x_s ON THE RESULTING MICROSTRUCTURES FOR SPINODAL INITIALIZATION ($x_i = 0.63$); THE RESULTS WITHOUT RIGID BODY MOTION ARE DUPLICATED FROM A PREVIOUS PUBLICATION (BELLEMANS ET AL. 2015b) AND THE RESULTS FOR THE RELATIVELY SLOW MOVEMENT ($10\Delta x$ PER $10s$) ARE DUPLICATED FROM SECTION 4.1.

In the non-wetting regime, it is clear that the attachment of the metal to the solid particle depends on coincidence, i.e. whether or not a metal droplet is present on the path in which the solid particle moves. Thus, there is no influence of the solid particle speed. For the low, and high wetting regimes, however, the influence of the interfacial energies becomes larger, which means that the metal will try to remain attached to the solid particle, by moving along with it. However, if the solid moves too fast, for example in the $10\Delta x$ per $10s$ case, the metal liquid cannot keep up with the solid and the attachment disappears. Finally, in the full wetting case ($x_s = 0.90$), the interfacial energy attraction of the metal towards the solid is so high that the liquid follows the solid even at the highest velocity.

The influence of the speed of rigid body motion on the area of attached metal is shown in the graphs in [Fig. 9](#) for the different x_s -values, and thus for different wetting regimes.

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FIG. 9 AREA ATTACHED METAL FOR SEVERAL VALUES OF THE SPEED OF MOTION OF THE SOLID PARTICLES FOR DIFFERENT x_s -VALUES (OR WETTING REGIMES). FULL BLACK SYMBOLS REPRESENT THE SIMULATIONS WITHOUT RIGID-BODY MOTION; GREY FULL SYMBOLS REPRESENT THE SIMULATION WITH A SOLID PARTICLE VELOCITY OF $1 \Delta x / 10 \Delta t$; THE OPEN BLACK SYMBOLS REPRESENT THE SIMULATION WITH A SOLID PARTICLE VELOCITY OF $5 \Delta x / 10 \Delta t$ AND THE OPEN GREY SYMBOLS REPRESENT THE SIMULATION WITH A SOLID PARTICLE VELOCITY OF $10 \Delta x / 10 \Delta t$. THIS LEGEND IS ALSO SHOWN IN PART A OF THE FIGURE.

It is clear that at low speed, the solid particle movement does not influence the amount of attached metal significantly. However, when the solid particle moves faster, a clearer influence becomes visible, depending on the wetting regime. For the non-wetting case ($x_s = 0.50$), there is no clear influence of the movement of the solid particle, it results in either a 'pick up' of droplets, thereby increasing the amount of attached metal, or it can lose an already attached metal droplet.

For the low wettability cases ($x_s = 0.60$ and 0.70), the amount of attached metal decreases as the speed of motion of the solid particle increases. At $x_s = 0.74$, with an expected contact angle of 90° , the velocities of x_5 and x_{10} clearly show the disappearance of attached metal to the solid particle. Also note that this detachment happens faster at a velocity of x_{10} .

For the high wetting regime ($x_s = 0.80$), the interfacial energies favour the attachment of the metal to the solid more, which is also seen in the fact that the detachments for both the x_5 and x_{10} velocity take place at a later time step than for $x_s = 0.74$.

Finally, at a value of $x_s = 0.9$, which corresponds to very high wetting, the attachment of the metal to the solid is energetically so favourable that the metal remains attached during the whole simulation, even during the very fast movement of the solid particle.

In general, we can conclude that, for low up to high wetting, the amount of attached metal will first increase (droplets do not detach and more droplets can attach as the particle moves) and then decrease (droplets detach) with increasing velocity of the particle. For increasing wettability and a given volume fraction of the metal droplets, the maximum amount of attached metal will be reached at a higher particle velocity.

Moreover, the apparent contact angle was measured and compared to the contact angles for situations without rigid body motion and the relatively slow movement in section 4.3. The results are shown in [Table 6](#) and [Fig. 10](#).

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x_s	$\theta_{\text{predicted}} (^{\circ})$	$\theta (^{\circ})$ without	$\theta (^{\circ})$ 1 Δx per 10s	$\theta (^{\circ})$ 5 Δx per 10s	$\theta (^{\circ})$ 10 Δx per 10s
0.5	/	164.2 $\pm$ 5.2	/	169.5 $\pm$ 4.5	172.9 $\pm$ 5.4
0.6	147.5	148.1 $\pm$ 9.2	158.1 $\pm$ 10.7	161.7 $\pm$ 3.7	No wetting
0.7	102.6	104.3 $\pm$ 8.9	123.8 $\pm$ 16.2	131.8 $\pm$ 0.2	No wetting
0.74	90.0	87.1 $\pm$ 2.7	114.1 $\pm$ 27.3	116.4 $\pm$ 1.2	No wetting
0.8	70.6	Full wetting	55.1 $\pm$ 4.5	75.8 $\pm$ 3.4	/
0.9	6.1	Full wetting	Full wetting	Full wetting	Full wetting

TABLE 6 COMPARISON OF THE PREDICTED AND MEASURED CONTACT ANGLES ($^{\circ}$) OF THE METALLIC DROPLETS ATTACHED TO THE SOLID PARTICLE FOR THE SPINODAL INITIALIZATION WITH $x_i = 0.63$ FOR SYSTEMS WITHOUT RIGID BODY MOTION AND WITH A VARIATION IN THE SPEED OF THAT MOTION

FIG. 10 APPARENT CONTACT ANGLES FOR SIMULATIONS OF SPINODAL DECOMPOSITION WITH $x_i = 0.63$ FOR SYSTEMS WITHOUT AND WITH VARIOUS SPEEDS OF RIGID BODY MOTION. THE CONTACT ANGLE PREDICTED BY YOUNG'S EQUATION IS ALSO PLOTTED AS A REFERENCE.

For a faster movement ($5\Delta x$ per 10s), the apparent contact angle increases. This difference increases for higher wetting regimes. The fastest speed of motion, however, resulted in either non-attached metal or full wetting. It should be noted that the simulation with $x_s = 0.80$ for the highest speed was unclear regarding the attachment of the metal to the solid. Nevertheless, it is clear that the actual wetting regime is higher than expected based on the measured contact angle when rigid body motion is present.

A limitation of this work is that we have only considered the movement of a single particle. A more quantitative statistical analysis of the effect considering a large number of particles would require a huge system size and huge computational power, as the system has to be sufficiently large to contain a statistically relevant number of particles. However, from the presented simulations we could already obtain a number of important insights useful for the interpretation of experimental micrographs.

5 Conclusion

This study investigated how rigid body motion of the solid particle with respect to the slag and metal droplets and its speed influences liquid metal droplet attachment to that solid particle in liquid slags with a phase field model. Important insights from the simulations are the following.

1. There is a trade-off between the speed of the movement of the liquid metal on the one hand, which depends on the attraction of the metal towards the solid particle by interfacial energies and the speed of the movement of the solid particle, which we varied. For slow solid motion, the interfacial energies have the upper hand, but for larger velocities, the metal cannot necessarily keep up with the movement of the solid. Thus, the amount of metal will decrease for faster movement. However, this is only the case for the low and high wettability regimes. For the non-wetting case, metal

droplets coincidentally present on the path of the solid particle will get attached, but the solid particle movement can similarly decrease the amount of the attached metal by moving away from the metal droplet. On the other hand, for the very high (almost full) wetting case, the interfacial energy attraction of the metal to the solid is so large that even the largest velocity did not decrease the amount of attached metal.

2. For the considered particle velocities, the apparent contact angle of the metal is larger when rigid body motion is present, which corresponds to a lower apparent wettability.

This effect was very clear and may confuse the interpretation of experimental micrographs.

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Figures

FIG. 1 ILLUSTRATION OF LOSS OF CORRECT DIFFUSENESS FOR A SIMULATION WITH SPINODAL INITIALIZATION, $x_l = 0.63$ AND $x_s = 0.70$ (LOW WETTING REGIME) WITH A ZOOMED-IN INSET

FIG. 2 MICROSTRUCTURE WHEN L_{INITIAL} IS ALSO USED EVERY 100TH TIME STEP TO OBTAIN CORRECT DIFFUSENESS OF THE INTERFACES FOR A SIMULATION WITH SPINODAL INITIALIZATION, $x_l = 0.63$ AND $x_s = 0.70$ (LOW WETTING REGIME)

Wetting regime	x_l x_s		0.61 without	0.605 with	0.63 without	0.63 with	0.65 without	0.65 with
No wetting	0.50							
Low wettability	0.60

FIG. 3 THE INFLUENCE OF x_i AND x_s (WETTING REGIME) ON THE RESULTING MICROSTRUCTURES FOR SPINODAL INITIALIZATION

FIG. 4 THE INFLUENCE OF x_i AND x_s ON THE RESULTING MICROSTRUCTURES FOR RANDOM INITIALIZATION

FIG. 5 AREA ATTACHED METAL FOR SEVERAL X_i, X_s -COMBINATIONS TO INVESTIGATE THE INFLUENCE OF RIGID BODY MOTION (ASSIGNED AS 'WITH')

FIG. 6 AREA ATTACHED METAL IN SEVERAL f_v, X_s -COMBINATIONS TO INVESTIGATE THE INFLUENCE OF RIGID BODY MOTION (ASSIGNED AS 'WITH')

FIG. 7 THE COMPARISON OF THE APPARENT CONTACT ANGLE BETWEEN THREE DIFFERENT CASES: (A) WITHOUT MOVEMENT OF THE SOLID PARTICLE; (B) WITH THE RIGID BODY MOTION OF THE SOLID PARTICLE; (C) AN 'EQUILIBRATION' SIMULATION WITHOUT MOVEMENT OF THE SOLID PARTICLE (STARTING FROM THE FINAL RESULT IN (B)).

FIG. 8 THE INFLUENCE OF THE SPEED OF MOTION OF THE SOLID PARTICLE AND x_s ON THE RESULTING MICROSTRUCTURES FOR SPINODAL INITIALIZATION ($x_i = 0.63$); THE RESULTS WITHOUT RIGID BODY MOTION ARE DUPLICATED FROM A PREVIOUS PUBLICATION (BELLEMANS ET AL. 2015b) AND THE RESULTS FOR THE RELATIVELY SLOW MOVEMENT (1 Δx PER 10 s) ARE DUPLICATED FROM SECTION 4.1.

(a) $x_s = 0.50$ (non-wetting) (b) $x_s = 0.60$ (low wetting) (c) $x_s = 0.70$ (low wetting) (d) $x_s = 0.74$ (contact angle of 90°) (e) $x_s = 0.80$ (high wetting)(f) $x_s = 0.90$ (high wetting)

FIG. 9 AREA ATTACHED METAL FOR SEVERAL VALUES OF THE SPEED OF MOTION OF THE SOLID PARTICLES FOR DIFFERENT x_s -VALUES (OR WETTING REGIMES). FULL BLACK SYMBOLS REPRESENT THE SIMULATIONS WITHOUT RIGID BODY MOTION; GREY FULL SYMBOLS REPRESENT THE SIMULATION WITH A SOLID PARTICLE VELOCITY OF $1 \Delta x/10$ s; THE OPEN BLACK SYMBOLS REPRESENT THE SIMULATION WITH A SOLID PARTICLE VELOCITY OF $5 \Delta x/10$ s AND THE OPEN GREY SYMBOLS REPRESENT THE SIMULATION WITH A SOLID PARTICLE VELOCITY OF $10 \Delta x/10$ s. THIS LEGEND IS ALSO SHOWN IN PART A OF THE FIGURE.

FIG. 10 APPARENT CONTACT ANGLES FOR SIMULATIONS OF SPINODAL DECOMPOSITION WITH $x_i = 0.63$ FOR SYSTEMS WITHOUT AND WITH VARIOUS SPEEDS OF RIGID BODY MOTION. THE CONTACT ANGLE PREDICTED BY YOUNG'S EQUATION IS ALSO PLOTTED AS A REFERENCE.