METAL DROPLET ENTRAINMENT BY SOLID PARTICLES IN SLAGS: A PHASE FIELD - EXPERIMENTAL APPROACH

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Introduction

Various pyrometallurgical industries encounter production losses due to the mechanical attachment of metal droplets to solids in liquid slags. The attached metal cannot settle, decreasing the yield of the phase separation. This results in inadequate sedimentation and eventually production losses in *e.g.* industrial Cu smelters[1], Pb reduction smelting furnaces[2,3], *etc.* In this study, simulations and experiments are combined to obtain a systematic understanding of the role of different parameters involved. The performed experiments were used both as input for the simulations and to validate the simulation results.

Phase Field Simulations

A binary phase-field model simulates the attachment of liquid metal droplets to solid particles in liquid slags with a non-reactive solid particle. The influence of several parameters on the attachment of the metal droplets to the solids was investigated: the interfacial energies[4], the particle morphology[5], initialisation method[4,6], solid particle movement[7] and the speed of the solid particle movement[8]. Some of these parameters will be discussed in more detail below.

Depending on the interfacial energies[4], four regimes were observed: no wettability of the metal on the particle, low wettability, high wettability and full wetting. In the case of low wettability, the amount of attached metal does not increase with an increasing perimeter per area[5], even though the number of available positions for the metal increases. For high wettability, the expected increase in attached metal for an increasing perimeter per area is observed. Moreover, it became clear that several small particles yield more attached metal than one large particle. Furthermore, not only the perimeter per area, but also the available space for the droplet to grow is an important factor.

The model was also extended to consider realistic microstructures[6] based on actual micrographs. The origin of the attachment was investigated by comparing the two initialisation methods for the metal droplets. One initialisation corresponds to a case where the droplets are formed by a chemical reaction with both the droplets and the spinel solids involved. The other initialisation corresponds to a situation where the droplets and particles are formed independently and are then mixed in the slag. Based on previous research, both methods were considered for two wettability regimes: no and low wettability. In the non-wetting case, the spinodal initialisation gave microstructures corresponding best with the experiments, but in the low wettability case, the simulations of both initialisations correspond well with the experimental system.

Afterwards, it was investigated how rigid body motion of the solid particle influences the attachment[7]. Regarding the amount of attached metal, no actual trends could be observed. A major observation, however, is the fact that the apparent contact angle of the metal is larger when rigid body motion is present, which corresponds to a lower apparent wettability.

The study on the influence of the speed of the rigid body motion[8] showed that 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 were 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.

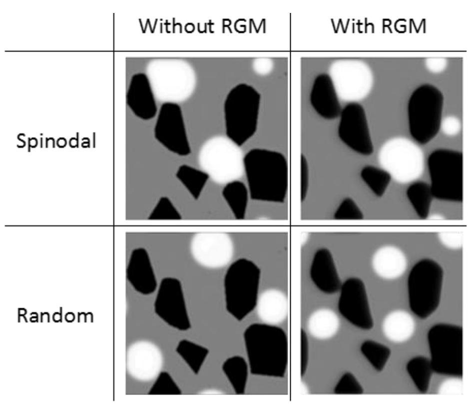

Figure 1: Comparison of simulations with spinodal (top) and random (bottom) initialisation in the absence (left) or presence (right) of rigid body motion (RGM) for a realistic solid microstructure in an extreme low wetting case

Figure 1 illustrates simulations based on real micrographs[8] with the two initialisation methods and absence and presence of rigid body motion.

The simulations for the spinodal initialisation correspond better with the experimentally observed micrographs.

Experiments

The simulations were compared to micrographs obtained from a smelting experiment in the Fe-Si-Al-O system with Cu-Ag droplets. During the experiment, the system was first oxidised so that the copper could dissolve in the slag. This was followed by reducing the system to lower the copper solubility in the slag in turn and favour precipitation of Cu-droplets, either attached to solid particles or on their own.

The amount and size of copper droplets and spinel particles increase during the complete experiment, as illustrated in Figure 2, which is in agreement with the expectations from the suggested mechanism during the reductive part, but during the oxidation part, this is not expected. Because the system was left to rest before the start of the oxidative part of the experiment, it is possible that the disturbance of the underlying alloy layer by blowing of the gas through the slag phase, introduces small metal droplets into the slag phase during the oxidative step.

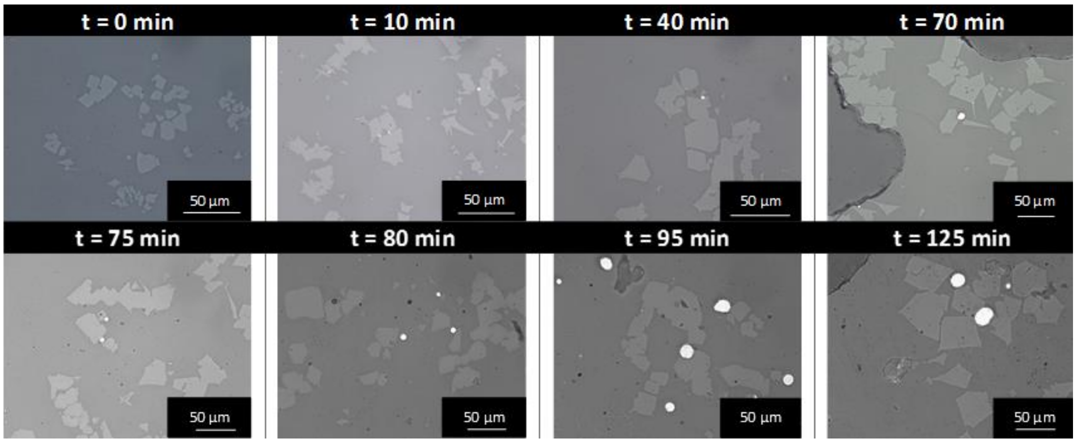

Figure 2: Optical images from the samples taken during the high-temperature oxidation (upper part) and reduction part (lower part) of the experiment

During the oxidative part of the experiment, iron oxide has a tendency to shift towards Fe_2O_3 and the metallic copper is oxidised. During the reductive part of the experiment, a lack of oxygen becomes apparent and because copper is more noble than iron, Cu_2O acts as oxygen donor for the oxidation of iron oxide, thereby precipitating metallic copper: $2FeO + Cu_2O \leftrightarrow 2Cu^0 + Fe_2O_3$. The resulting increase of Fe_2O_3 leads to the formation of magnetite spinel particles by reaction with FeO. In the experiment, silver was added to the copper alloy as a trace element to get more insights into the origin of the attachment. The fact that silver is present in the attached copper droplets indicates that the origin of the attachment is not purely reactive. It is possible that

small Cu-Ag droplets were introduced in the slag during the first oxidative blow. Afterwards, these droplets act as nucleation sites for a simultaneous reduction of copper oxides into metallic copper and the oxidation of slag oxides into more stable spinel structures. Consequently, the spinel solids grow at the side of the Cu-Ag droplets, which are in turn enriched with Cu and grow, leading to copper droplets attached to spinel solids within the slag phase.

Conclusions

The experiments correspond better to the experiments with spinodal initialisation, which shows that the droplet-particle interaction could result from a reactive origin. Moreover, experimentally, the actual 'attachment' of the droplets seems to be a non-equilibrium phenomenon, which is in correspondence with the results for the spinodal initialisation, as in that case the droplets are first attached to the solid as they are formed simultaneously and are afterwards also detached/dissolved. Explicit implementation of a model of the formation of the solid particles within a liquid slag on the side of or together with a metal droplet in a multicomponent multiphase system will allow to simulate and study the reactive origin for the attachment in more detail.

References

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