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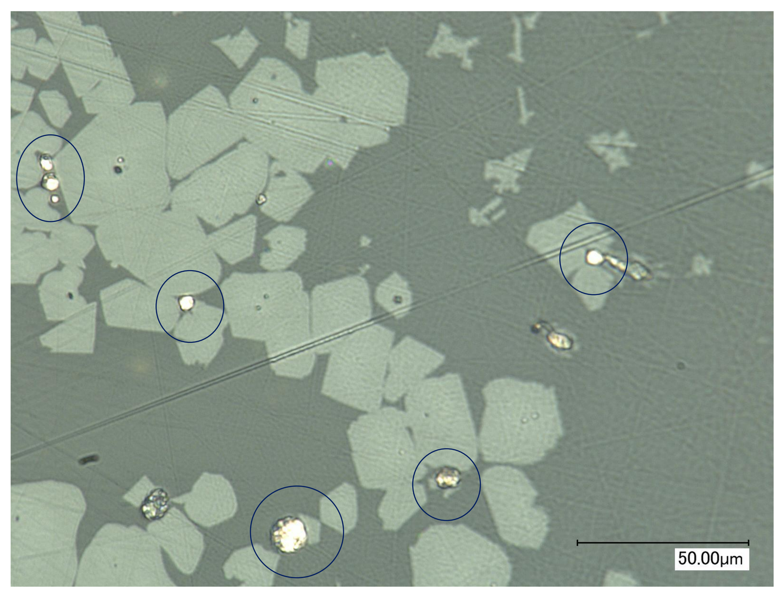
The influence of interfacial energy and particle characteristics on the attachment of metallic droplets to solid particles in liquid slags

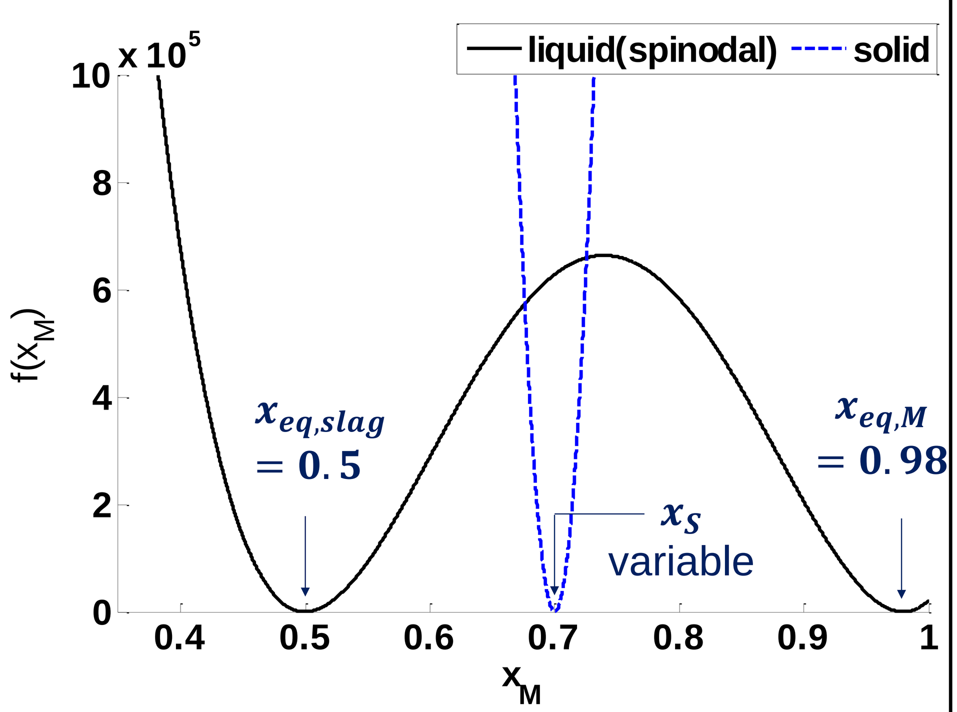
Introduction

Metallic droplets attached to solid particles in slags lead to production losses in pyrometallurgy.



Experimental micrograph of Cu droplets attached to solid particles (E. De Wilde et al. unpublished data)

Input parameters

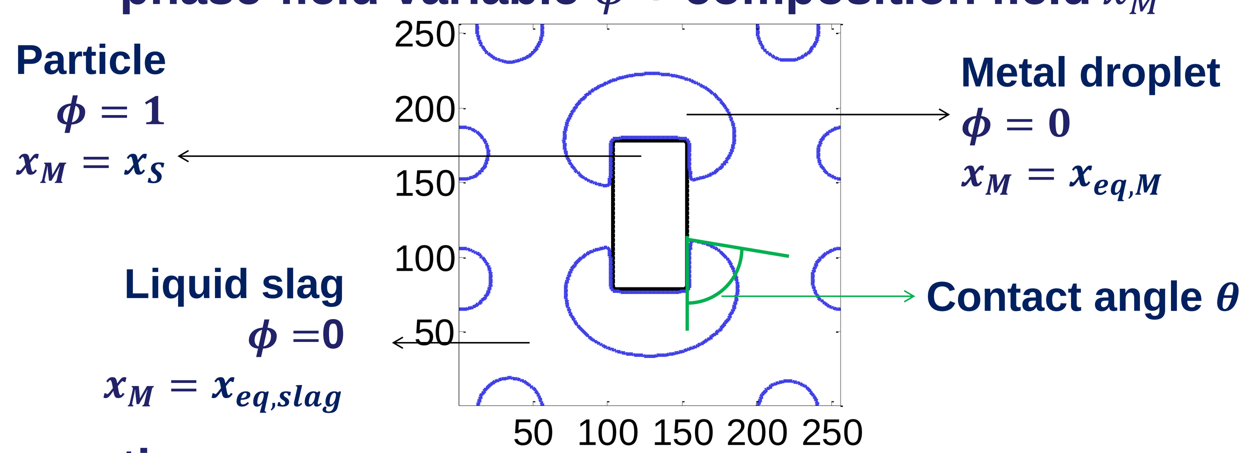


Symbol	Description	Value(s)
Δx	Grid spacing	$4/\sqrt{10}\,10^{-7}$ m
W	Depth of the double well function	$15 \cdot 10^6$ J/m³
κ_ϕ	Gradient energy coefficient for the solid-liquid interfaces	$15/8 \cdot 10^6$ J/m
L	Kinetic coefficient for the evolution of ϕ	10^{-30} m³/(J s)
κ_{x_M}	Gradient energy coefficient for the liquid-liquid interfaces	$6 \cdot 10^{-6}$ J/m
M	Mobility coefficient of the metal	10^{-19} m⁵/(J s)

Model

Phase field model for solid-liquid binary system (O-M) with spinodal decomposition in the liquid

phase-field variable ϕ + composition field x_M



Assumptions:

- Particle does not react
- Constant molar volume V_m in all phases
- Particles present before droplets
- Constant T and P
- No convection

$$F = \int \left[f_{S-L,interf}(\phi) + (1-h(\phi)) f_{Liquid}(x_M) + h(\phi) f_{Solid}(x_M) + f_{\nabla x_M} \right] dV$$

$$= \int \left[W g(\phi) + \frac{\kappa}{2}(\nabla\phi)^2 + (1-h(\phi))\frac{A_{Sp}}{2}(x_M - x_{eq,0})^2 (x_M - x_{eq,1})^2 + h(\phi)\frac{A_S}{2}(x_M - x_S)^2 + \frac{\kappa_{x_M}}{2}(\nabla x_M)^2 \right] dV$$

With $h(\phi) = \phi^3(10 - 15\phi + 6\phi^2)$

Interfacial widths and energies:

$$\gamma_{L,L} = \frac{1}{6}\sqrt{\kappa_{x_M} A_{Sp}}\left(x_{eq,1} - x_{eq,0}\right)^3$$

$$\gamma_{S,L} = \frac{1}{3\sqrt{2}}\sqrt{W\kappa} + \gamma_{S,L}^{\nabla x_M}$$

With $\gamma_{S,L}^{\nabla x_M} \approx \frac{1}{6}\sqrt{\kappa_{x_M} 0.5 (A_{sp} + A_s)}\left(\left|x_{eq,k} - x_S\right|\right)^3$

x_S can be used **as a parameter** to vary the solid-liquid interfacial energies

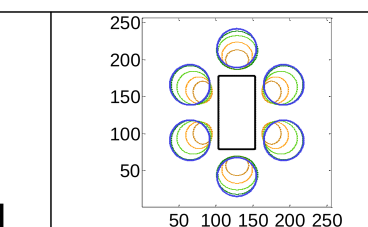
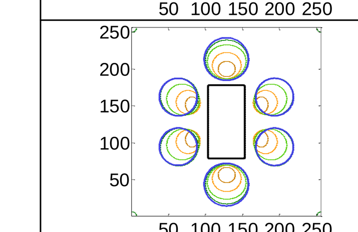
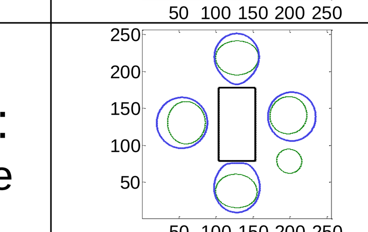
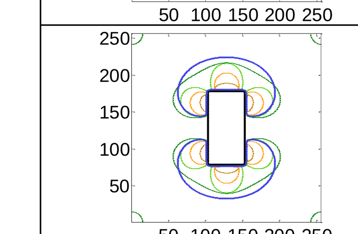
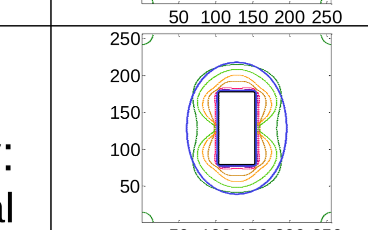
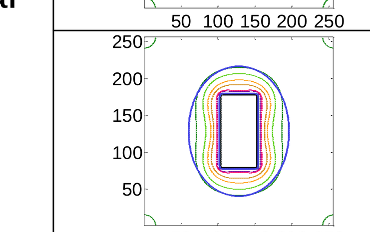
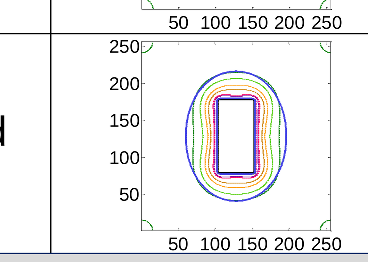
Evolution equations

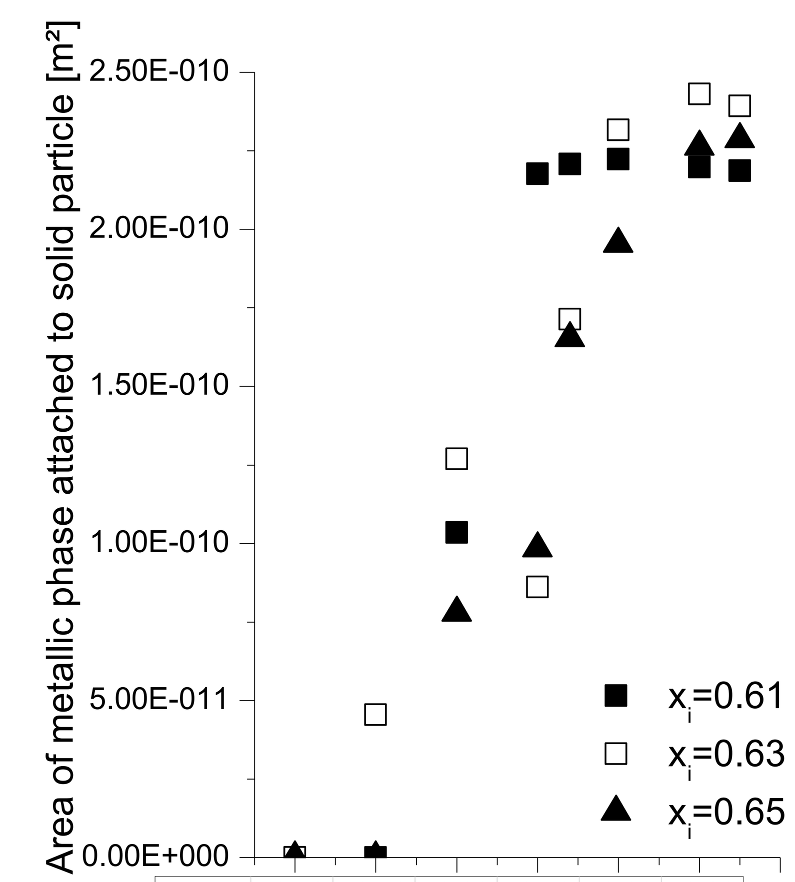
$$\frac{\partial x_M}{\partial t} = \nabla\left[M \nabla\left[(1-h(\phi)) f'_{Liquid}(x_M) + h(\phi) f'_{Solid}(x_M) - \kappa_{x_M}\nabla^2 x_M\right]\right]$$

$$\frac{\partial \phi}{\partial t} = -L\left[W g'(\phi) + h'(\phi)(f_{Solid} - f_{Liquid}) - \kappa\nabla^2\phi\right]$$

Results (after evolution of 100s = 10⁶ steps)

Influence of interfacial energies and initial supersaturation (x_i)

x_S	$\gamma_{S,LO}$ [N/m]	$\gamma_{S,LM}$ [N/m]	$\gamma_{LO,LM}$ [N/m]	Contact angle θ [°]	Wetting behaviour	Contour plots x_i = 0.61 (*)
0.4	1.2641	4.0093	0.903	/	**Non-wetting**: only oxide attached $\lvert\gamma_{SLO} - \gamma_{SLM}\rvert > \gamma_{LL}$ & $\gamma_{SLO} < \gamma_{SLM}$	
0.5	1.2500	2.8140	0.903	/		
0.6	1.2641	2.0260	0.903	147.54	**Low wettability**: predominantly oxide attached $\lvert\gamma_{SLO} - \gamma_{SLM}\rvert < \gamma_{LL}$ & $\gamma_{SLO} < \gamma_{SLM}$	
0.7	1.3631	1.5604	0.903	102.62		
0.8	1.6318	1.3325	0.903	70.64	**High wettability**: predominantly metal attached $\lvert\gamma_{SLO} - \gamma_{SLM}\rvert < \gamma_{LL}$ & $\gamma_{SLO} > \gamma_{SLM}$	
0.9	2.1551	1.2572	0.903	6.09		
0.95	2.5387	1.2504	0.903	/	**Full wetting**: only metal attached $\lvert\gamma_{SLO} - \gamma_{SLM}\rvert > \gamma_{LL}$ & $\gamma_{SLO} > \gamma_{SLM}$	

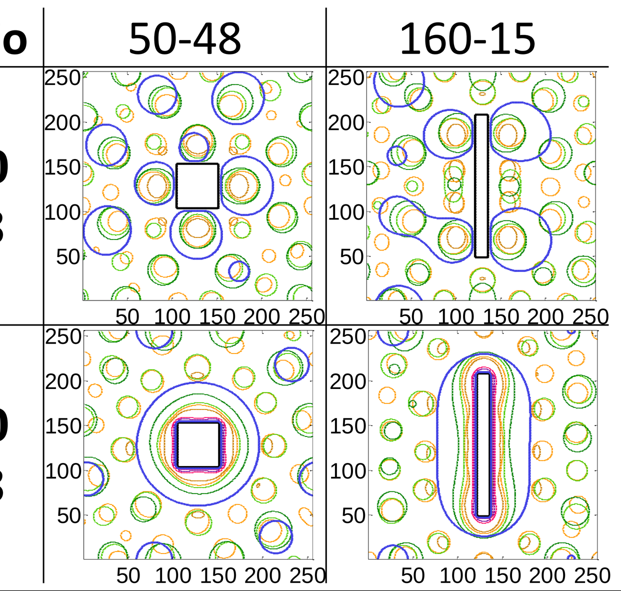


x_s	0.40	0.50	0.60	0.70	0.80	0.90
cos(θ)	< -1	< -1	-0.84	-0.22	0.33	0.99
θ (°)	/	/	**147.5**	**102.6**	**70.64**	**6.09**

Influence of particle morphology

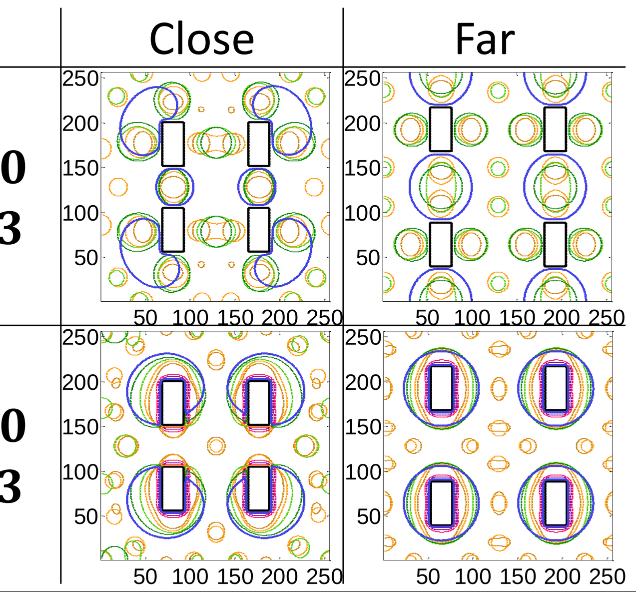
- Particle aspect ratio / perimeter
  - HIGH wettability: as expected: larger perimeter → more attached metal
  - LOW wettability: not as expected
- Proximity of particles
  - particles close together : metal attached on side not oriented to other particles
  - 1 metal droplet shared by several particles for LOW wettability

Aspect ratio: 50-48, 160-15



(*) legend
- particle
- 0.25 s
- 0.5 s
- 2.5 s
- 5 s
- 10 s
- 25 s
- 100 s

Close, Far



Conclusions

- Presence of particle → 'localized' spinodal decomposition → droplets nucleate locally around or even on sides of particle
- x_S-variation → change in interfacial energies, contact angles and wetting behaviour (4 regimes)
- Perimeter of the solid particles → number of available positions for metal to be attached to → only important in HIGH wettability regime
- Distance between particles → no influence on amount of metal attached + but droplets oriented away from other particles when too close to one another

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