

Introduction

The objective of this work is to ascertain the capability of COMSOL Multiphysics® in addressing complex nonlinear problems of columnar solidification, with adequate precision to unveil the appearance of a macrosegregation instability that manifests itself in the form of oblique channels of alternating liquid fraction and solute concentration.

This numerical exercise is performed in response to the call for contributions to a benchmark problem for 2D columnar solidification of binary alloys [1]. The call provides a set of physical equations to be solved numerically with the objective of verifying the agreement of the numerical solutions obtained by different contributors [2, 3].

The proposed solidification problem – with equations, boundary conditions and parameters – is described in detail in the “Problem statement” section, followed by the “Numerical methods” section. New modelling results, obtained with COMSOL Multiphysics®, are presented and discussed in the “Simulation results and discussion” section, where they are compared with other numerical works.

Problem statement

The modelled configuration is a 2D ingot casting problem. The heat is extracted from side walls of a rectangular mold initially filled with a stagnant liquid binary metal at a uniform temperature and composition. To have the possibility to compare numerical results with available experimental data, a geometry like in the Hebditch and Hunt experiment [4] is retained, see Figure 1.

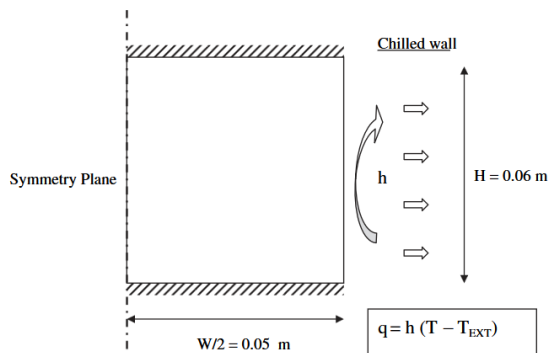


Figure 1. Problem geometry and boundary conditions, reproduced from [1].

The solidification of a binary alloy is modelled in a 2D rectangular cavity of height $H = 0.06$ m and width $W = 0.1$ m. The initial alloy temperature T_0 is equal to the liquidus temperature T_{liq} . A uniform initial solute mass fraction C_0 is imposed. The

solidification starts at time $t = 0$ by cooling right wall of the enclosure through an external cooling fluid at temperature T_{ext} and a heat transfer coefficient h , while the symmetry condition is imposed on the left side of the computational domain, see Figure 1. The top and bottom walls are thermally insulated. No-slip conditions at the cavity walls are assumed.

Hypotheses

The mathematical model for this benchmark problem, as proposed in the call [1], is minimal, i.e. it has been simplified to the greatest possible extent and includes the following hypotheses:

- Laminar flow and constant viscosity μ_l .
- Solute diffusion in the liquid and solid at the macroscopic scale is neglected.
- Constant density $\rho_s = \rho_l = \rho_0$ except in the buoyancy term of the momentum conservation equation, where ρ linearly depends on temperature T and solute mass fraction in a liquid phase ω_l (Boussinesq approximation):

$$\rho = \rho_0(1 - \beta_T[T - T_0] - \beta_C[C_l - C_0])$$
The direct consequence of constant density is the equivalence of mass and volume fractions:
- Saturated mixture: $g_l + g_s = 1$.
- Local thermal equilibrium, $T_l = T_s = T$, with perfect solute diffusion in both phases (lever rule).
- Constant thermal conductivity: $k_s = k_l = k$.
- Constant heat capacity: $c_{p,s} = c_{p,l} = c_p$ resulting in the following simplified expressions of specific enthalpies:

$$h_s = c_p T, \quad h_l = c_p T + L,$$

$$\langle h \rangle = \omega_l h_l + \omega_s h_s = c_p T + g_l L$$

- The solid phase is assumed motionless: $u_s = 0$.
- The mushy zone is an isotropic porous medium whose permeability κ is defined by the conventional Kozeny-Carman relation: $\kappa = \lambda_2^2 g_l^3 / 180(1 - g_l)^2$, although slightly different from that presented in [1].

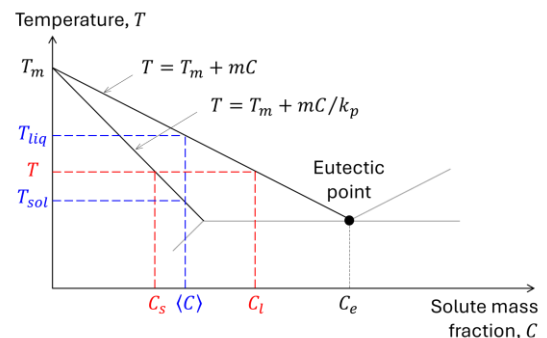


Figure 2. Simplified phase diagram.

In accordance with the lever rule, the following local relations are true, see Figure 2:

$$\begin{aligned} C_s &= k_p C_l \\ \langle C \rangle &= \omega_l C_l + \omega_s C_s = (g_l + k_p[1 - g_l])C_l \\ T &= T_m + mC_l, \quad T = T_m + mC_s/k_p \\ T_{liq} &= T_m + m\langle C \rangle, \quad T_{sol} = T_m + m\langle C \rangle/k_p \\ g_l &= \begin{cases} 0, & T < T_{sol} \\ \frac{k_p(T - T_{sol})}{k_p(T - T_{sol}) + (T_{liq} - T)}, & T_{sol} \leq T \leq T_{liq} \\ 1, & T > T_{liq} \end{cases} \end{aligned}$$

Although g_l above is conveniently expressed in terms of temperature T and Favre averaged solute mass fraction $\langle C \rangle$, this formula fails, from a mathematical point of view, in two limiting cases: 1) when $k_p = 1$ implying $T_{liq} = T_{sol}$; and 2) when $\langle C \rangle = 0$ implying $T_{liq} = T_{sol} = T_m$. In both cases, for $T = T_{liq} = T_{sol}$, the result is indeterminate of type (0/0). A more robust way to compute g_l is to define it as function of Favre averaged specific enthalpy $\langle h \rangle$ and Favre averaged solute mass fraction $\langle C \rangle$. To do so, one must solve the following quadratic equation and take greater root as the physical solution:

$$\begin{aligned} ag_l^2 + bg_l + c &= 0 \quad \text{for} \quad \langle h \rangle_{sol} \leq \langle h \rangle \leq \langle h \rangle_{liq} \\ g_l &= \frac{-b + \sqrt{D}}{2a}, \quad D = b^2 - 4ac \end{aligned}$$

where

$$\begin{aligned} a &= L(1 - k_p), \quad b = Lk_p + (1 - k_p)(c_p T_m - \langle h \rangle), \\ c &= k_p(c_p T_{sol} - \langle h \rangle), \\ \langle h \rangle_{sol} &= c_p T_{sol}, \quad \langle h \rangle_{liq} = c_p T_{liq} + L \end{aligned}$$

The final expression for g_l as function of $\langle h \rangle$ and $\langle C \rangle$ is

$$g_l = \begin{cases} 0, & \langle h \rangle < \langle h \rangle_{sol} \\ \frac{-b + \sqrt{D}}{2a}, & \langle h \rangle_{sol} \leq \langle h \rangle \leq \langle h \rangle_{liq}, a \neq 0 \\ -c/b, & \langle h \rangle_{sol} \leq \langle h \rangle \leq \langle h \rangle_{liq}, a = 0 \\ 1, & \langle h \rangle > \langle h \rangle_{liq} \end{cases}$$

This formula provides a well-defined value for g_l even in the previous limiting cases where $T_{liq} = T_{sol}$:

$$g_l = (\langle h \rangle - c_p T_{sol})/L \quad \text{if} \quad T = T_{liq} = T_{sol}.$$

Conservation equations

Macroscopic conservation equations result from volume averaging of microscopic conservation equations. The following notation is adopted:

$$p = p_l, \quad \mathbf{u} = g_l \mathbf{u}_l.$$

Laminar *Brinkman equation* for momentum conservation in a porous medium, as defined in COMSOL Multiphysics®:

$$\begin{aligned} \frac{\rho_0}{g_l} \left(\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \frac{\mathbf{u}}{g_l} \right) &= \nabla \cdot [-p\mathbf{I} + \mathbf{K}] - \frac{\mu_l}{\kappa} \mathbf{u} + \rho \mathbf{g} \\ \mathbf{K} &= \frac{\mu}{g_l} [\nabla \mathbf{u} + (\nabla \mathbf{u})^T] \end{aligned}$$

Note that this equation is slightly different from the one proposed in [1]. In particular, the placement of the factor $1/g_l$ is different in the viscous stress term and in the convective term. Nevertheless, after thorough consideration, we believe that the Brinkman equation above is derived rigorously from the Navier-Stokes equations using the volume averaging technique and is therefore retained in the present work.

Mass conservation

$$\nabla \cdot \mathbf{u} = 0$$

Energy conservation

$$\begin{aligned} \rho \frac{\partial \langle h \rangle}{\partial t} + \rho \mathbf{u} \cdot \nabla h_l + \nabla \cdot \mathbf{q} &= 0, \quad T = \vartheta(\langle h \rangle, \langle C \rangle) \\ h_l &= c_p T + L, \quad \mathbf{q} = -k \nabla T \end{aligned}$$

Note that, although the convective term in the above equation differs, it is still equivalent to that in reference [1], which is expressed via the temperature gradient $\rho \mathbf{u} \cdot \nabla h_l = \rho c_p \mathbf{u} \cdot \nabla T$. The function $\vartheta(\langle h \rangle, \langle C \rangle)$ can be defined using the expressions for $\langle h \rangle$ and g_l :

$$\vartheta(\langle h \rangle, \langle C \rangle) = \frac{\langle h \rangle - L g_l(\langle h \rangle, \langle C \rangle)}{c_p}$$

Solute conservation

$$\frac{\partial \langle C \rangle}{\partial t} + \mathbf{u} \cdot \nabla C_l = 0, \quad C_l = \frac{\langle C \rangle}{g_l + k_p[1 - g_l]}$$

Problem parameters for two cases of Sn-Pb alloys are given in **Table 1**.

Table 1: Physical properties of Pb-18% Sn and Sn-10% Pb alloys. Reproduced from [1].

Property	Symbol	Units	Pb-18% Sn	Sn-10% Pb
Specific heat	c_p	J(kg K) ⁻¹	176	260
Thermal conductivity	k	W(m K) ⁻¹	17.9	55.0
Reference density	ρ_0	kg m ⁻³	9250	7000
Latent heat of fusion	L	J kg ⁻¹	3.76×10^4	6.1×10^4
Liquid dynamic viscosity	μ_l	Pa s	1.10×10^{-3}	1.0×10^{-3}
Liquid thermal expansion coefficient	β_T	K ⁻¹	1.16×10^{-4}	6.0×10^{-5}
Liquid solutal expansion coefficient	β_C	(wt%) ⁻¹	4.90×10^{-3}	-5.3×10^{-3}
Secondary dendrite arm spacing	λ_2	m	1.85×10^{-4}	65.0×10^{-6}
Melting point at $C = 0$	T_m	°C	327.5	232.0
Eutectic solute mass fraction	C_e	wt%	61.911	38.1
Equilibrium partition coefficient	k_p	—	0.310	0.0656
Liquidus slope	m	°C (wt%) ⁻¹	-2.334	-1.286
Nominal solute mass fraction	C_0	wt%	18.0	10.0
Initial temperature	$T_0 = T_{liq}$	°C	285.488	219.14
Heat transfer coefficient	h	W m ⁻² K ⁻¹	400	400
External temperature	T_{ext}	°C	25	25

Numerical methods

The problem is solved with the finite element software COMSOL Multiphysics® version 6.4. The following modules have been used to set up the model physics: the Brinkman Equations for laminar fluid dynamics in porous medium (variables $\mathbf{u}$ and p), the Stabilized Convection-Diffusion Equation for the transport of averaged solute mass fraction (variable $\langle C \rangle$), and the Weak Form PDE for enthalpy (variable $\langle h \rangle$), temperature (variable T) and solute mass fraction of liquid phase (variable C_l). The velocity field $\mathbf{u}$ is spatially discretized with quadratic Lagrange elements, while other fields are discretized with linear Lagrange elements. Time integration is performed with the Backward Differentiation Formula (BDF) of order 1. The computational domain is spatially discretized with a triangular mesh that consists of at least 13 400 finite elements resulting in 88 400 degrees of freedom.

Enthalpy method

The enthalpy method involves solving two equations: one for the averaged specific enthalpy $\langle h \rangle$ and one for the temperature T (as a function of $\langle h \rangle$ and $\langle C \rangle$). Following the recommendations of Feulvarch & Bergheau [5], the correct finite-element formulation of the problem requires that both the temperature and the enthalpy equations are solved in a weak sense to avoid spurious oscillations of the numerical solution. Same linear spatial discretization for $\langle h \rangle$ and T is recommended to model phase changes [5]. In the spirit of this method, the two solute transport equations, one for $\langle C \rangle$ and one for C_l are also solved simultaneously in a weak sense with same spatial discretization for both fields.

Simulation results and discussion

The present modelling results obtained using COMSOL Multiphysics® are compared with results from the literature obtained using different software. Mesh and time-step convergence are qualitatively assessed. The influence of the heat exchange coefficient on the appearance of the macro segregation channels is also briefly discussed.

First, let us assess the mesh convergence of the present numerical model. As one can see in Figure 3, refining the spatial resolution by increasing the total number of mesh elements from 13 394 to 120 612 results in a slight variation of the observable pattern of segregated channels. Although the overall appearance of the computational result remains unchanged, mesh refinement provides higher spatial resolution, not only revealing finer structures within the segregated channels but also changing their morphology and spacing. In agreement with the

work of Combeau et al. [2, 3, 6], we conclude that the local structure of the segregated channels is highly mesh-sensitive, whereas the overall behavior of the segregation instability is well captured even on a coarse mesh.

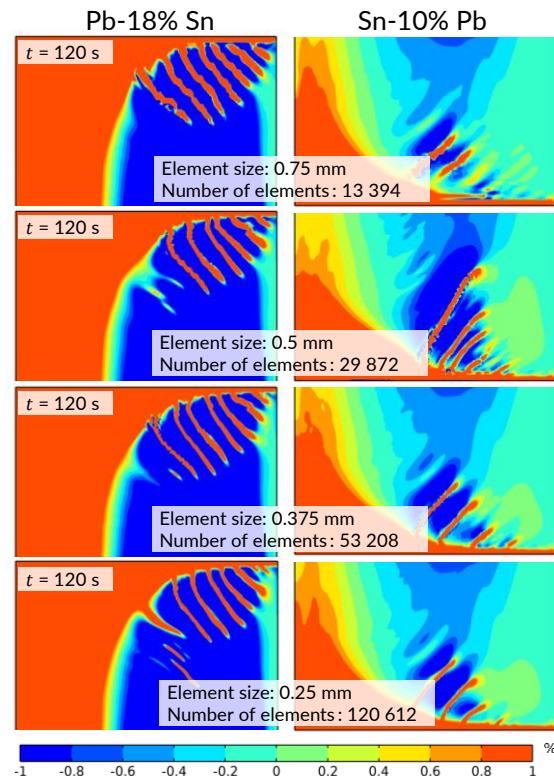


Figure 3. Influence of the numerical mesh resolution. The color field represents the averaged solute mass fraction $\langle C \rangle - C_0$ in %. Only the upper part of the computational domain is shown for Pb-18% Sn, and the lower part is shown for Sn-10% Pb.

Since COMSOL Multiphysics® may adjust the time step of the BDF solver based on the estimated numerical error, we can only restrict its maximum value. Thus, the influence of the maximum time step on the numerical solution is shown in Figure 4.

As one can observe, different time steps can lead to a slightly different local structure of segregated channels. As in the case of mesh convergence, a similar conclusion can be drawn for time-step convergence: the local structure of the segregated channels is highly sensitive to the time step, whereas the overall behavior of the instability remains similar for different time steps.

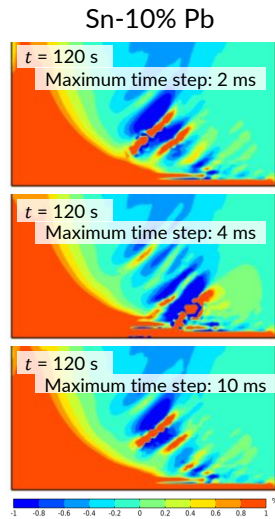


Figure 4. Influence of the maximum time step. The color field represents the averaged solute mass fraction $\langle C \rangle - C_0$ in %. Only the lower part of the computational domain is shown.

As can be seen from Figure 5, the appearance of segregated channels and the area they occupy are related to the cooling rate, which is controlled by the heat transfer coefficient h . Figure 5 shows the computational results for h ranging from 4 to 400 $W m^{-2} K^{-1}$.

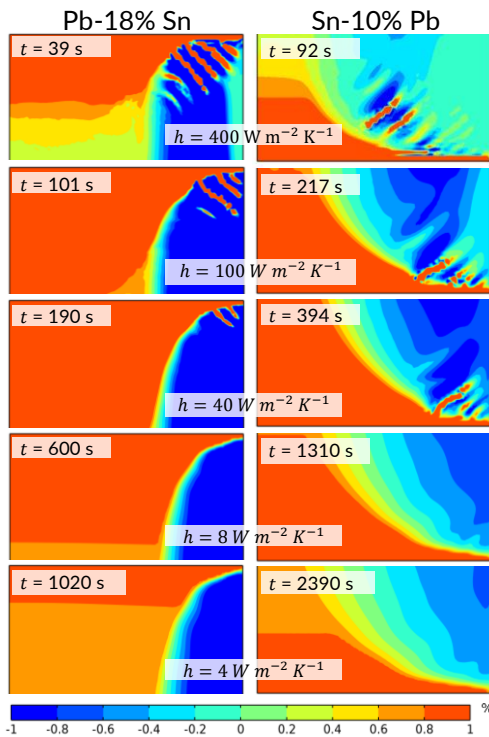


Figure 5. Influence of the heat transfer coefficient h . The color field represents the averaged solute mass fraction $\langle C \rangle - C_0$ in %. Only the upper part of the computational domain is shown for Pb-18% Sn, and the lower part is shown for Sn-10% Pb.

The results are compared at approximately the same position of the solidification front and therefore correspond to different time instances. As can be observed, the area occupied by the segregated channels decreases with decreasing cooling rate. Below a certain value of h between 8 and 40 $W m^{-2} K^{-1}$, the segregated channels cease to appear.

Finally, let us compare the solidification results obtained in the present work with the ones previously published by other researchers. Two alloys are considered: Pb-18% Sn (**case 1**) and Sn-10% Pb (**case 2**). In **case 1**, the comparison of the computational results by different research groups is presented in Figure 6. The final spatial distributions of the averaged solute mass fraction $\langle C \rangle$ are qualitatively similar. The differences appear to be more pronounced in the fine structure of the segregated channels, as the process of their formation is very sensitive to the numerical method.

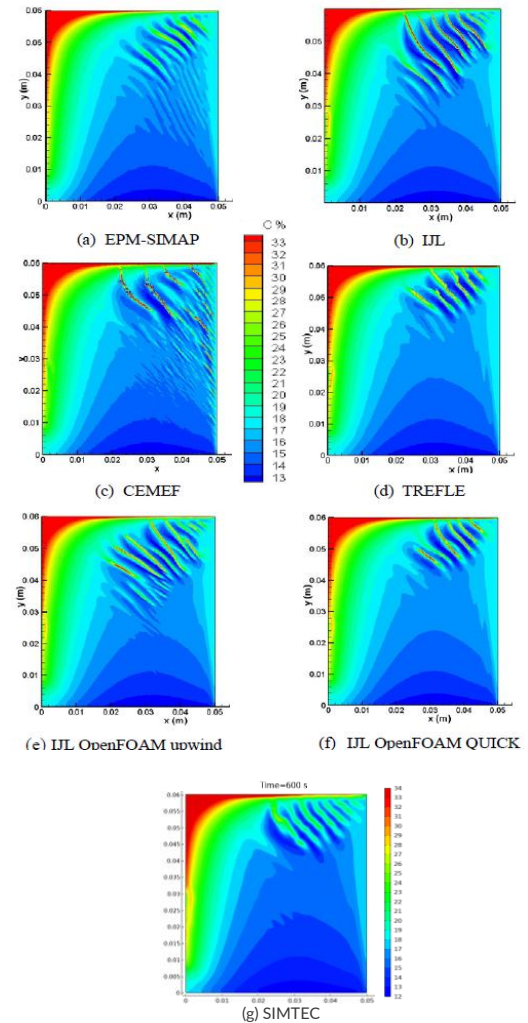


Figure 6. Case of Pb-18% Sn. Averaged solute mass fraction, $\langle C \rangle$ in %, at the end of solidification ($t = 600$ s) computed by different research groups [3, 6].

The volume fraction of the liquid phase is plotted as a function of x at mid-height in Figure 7 and demonstrates also quantitatively similar trends.

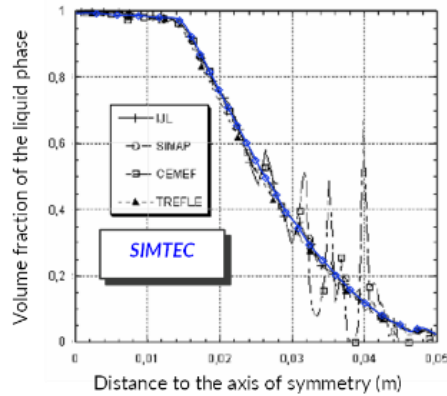


Figure 7. Case of Pb-18% Sn. Volume fraction of the liquid phase at $t = 120$ s as a function of x at mid-height computed by different research groups [6].

In **case 2**, Sn is the solvent and Pb is the solute rejected during solidification. Since Pb is denser than Sn, the solute-enriched liquid near the solid/liquid interface becomes locally denser than the bulk liquid. This solutal buoyancy force acts in the same direction as the thermal buoyancy force (the cooled liquid near the wall is also denser and sinks). The two driving forces therefore cooperate, reinforcing the convective flow and strongly destabilizing the mushy zone. This is why case 2 produces pronounced, well-developed channel segregates inclined at about 45° [2].

The comparison of the computational results by different research groups is presented in Figure 8. As in case 1, the final spatial distributions of the averaged solute mass fraction $\langle C \rangle - C_0$ are qualitatively similar. As before, the differences mainly appear in the fine structure of the segregated channels. These differences are attributed to variations in solution behavior at high liquid fractions, both in the central region, which solidifies last, and in the channels, which also form under high-liquid-fraction conditions.

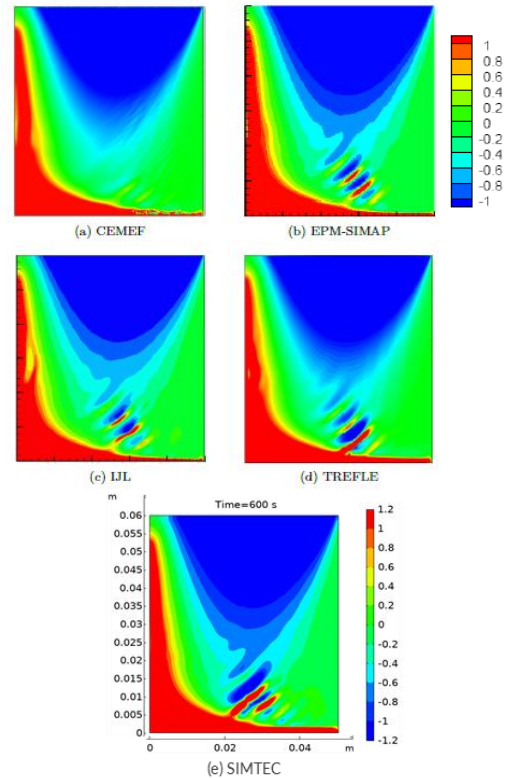


Figure 8. Case of Sn-10% Pb. Averaged solute mass fraction, $\langle C \rangle - C_0$ in %, at the end of solidification ($t = 600$ s) computed by different research groups [2].

The dependence of the velocity magnitude on time is shown in Figure 9 at midpoint of mid-height. The first peak reflects the onset of thermosolutal convection at high liquid fraction, before permeability drops as the mushy zone thickens; the second peak results from flow reorganizing into the high-permeability segregated channels as they form. The present results demonstrate again a very good quantitative agreement between other software.

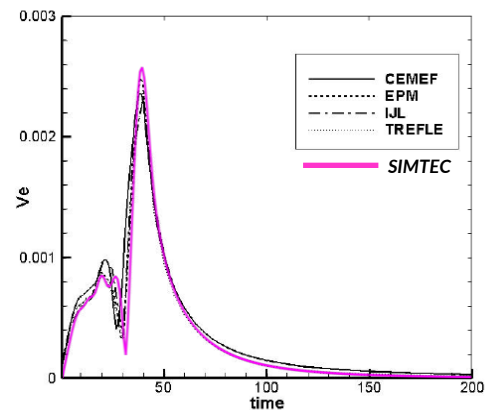


Figure 9. Case of Sn-10% Pb. Velocity magnitude as function of time, computed by different research groups [2].

Conclusions

This work has demonstrated the capability of COMSOL Multiphysics® to reproduce the coupled thermo-solutal convection and phase-change phenomena involved in the 2D columnar solidification benchmark problem, including the macrosegregation instability that manifests as oblique channels of alternating liquid fraction and solute concentration. The numerical implementation, based on an enthalpy formulation solved in a weak sense together with the Brinkman equations and a dual solute transport scheme, proved able to capture the essential physics of the problem with good stability and convergence behavior. The mesh and time-step sensitivity studies confirmed a trend already reported in the literature: while the local, fine-scale structure of the segregated channels is strongly dependent on the spatial and temporal discretization, the overall pattern and intensity of the segregation instability remain robust even on relatively coarse meshes and larger time steps. Additionally, varying the heat transfer coefficient showed that the extent of the segregated-channel region decreases as the cooling rate is reduced, with channels disappearing altogether below a threshold value.

Comparison with previously published results for both the Pb-18wt%Sn and Sn-10wt%Pb reference cases showed that the present model reproduces the final macrosegregation maps, the concentration and liquid-fraction profiles, and the time evolution of the velocity magnitude in good qualitative and quantitative agreement with the four independent codes considered in the benchmark. As observed by other contributors, the main discrepancies between solvers remain concentrated in the fine structure of the channels and in the high-liquid-fraction regions, which are the most sensitive to the numerical scheme employed.

Overall, these results confirm that COMSOL Multiphysics® provides a reliable first-order prediction tool for complex columnar solidification problems involving macrosegregation. This constitutes an encouraging basis for future work, which should focus on refining the solidification model — for instance by incorporating more realistic microsegregation and permeability laws — and on extending the validation against the experimental data available for this type of benchmark configuration.

References

- [1] M. Bellet, H. Combeau, Y. Fautrelle, D. Gobin, M. Rady, E. Arquis, O. Budenkova, B. Dussoubs, Y. Duterrail, A. Kumar, C. A. Gandin, B. Goyeau, S. Mosbah and M. Zaloznik, "Call for contributions to a numerical benchmark problem for 2D columnar solidification of binary alloys," *International Journal of Thermal Sciences*, vol. 48, no. 11, pp. 2013-2016, 2009.
- [2] H. Combeau, M. Bellet, Y. Fautrelle, D. Gobin, E. Arquis, O. Budenkova, B. Dussoubs, Y. Duterrail, A. Kumar, S. Mosbah, T. Quatravaux, M. Rady, C. A. Gandin, B. Goyeau and M. Zaloznik, "A numerical benchmark on the prediction of macrosegregation in binary alloys," *Proceedings of frontiers in solidification science. Warrendale, USA: TMS2011*, 2011.
- [3] H. Combeau, M. Bellet, Y. Fautrelle, D. Gobin, E. Arquis, O. Budenkova, B. Dussoubs, Y. Duterrail, A. Kumar, C. A. Gandin, B. Goyeau, S. Mosbah, T. Quatravaux, M. Rady and M. Zaloznik, "Analysis of a numerical benchmark for columnar solidification of binary alloys," *IOP Conference Series: Materials Science and Engineering*, vol. 33, no. 1, p. 012086, 2012.
- [4] D. J. Hebditch and J. D. Hunt, "Observations of ingot macrosegregation on model systems," *Metallurgical transactions*, vol. 5, no. 7, pp. 1557-1564, 1974.
- [5] E. Feulvarch and J.-M. Bergheau, "An implicit fixed-grid method for the finite-element analysis of heat transfer involving phase changes," *Numerical Heat Transfer, Part B: Fundamentals*, vol. 51, no. 6, pp. 585-610, 2007.