A scale adaptive dendritic envelope model of solidification at mesoscopic scales

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Abstract. We present the Scale Adaptive Dendritic Envelope (SADE) model of solidification at mesoscopic scales. The new approach is based on the rescaling of the microscopic laws on the desired resolution scale of simulation. The diffusivity, the Gibbs Thomson coefficient, and the fraction which solidifies with the solid front are modified to create large fictitious dendrites (envelope) whose tips growth at the same speed as the microscopic tips. The model is inspired from the methodology of the turbulence models that filter the scales that are smaller than the simulation grid size, such as the Large Eddy Simulation (LES) approach. Here, the solidified structure scales which are smaller than the grid size, such as the tip radius and the smallest arms, are modelled with sub-grid scale models. The envelope growth is coupled with a sub-grid model to account for the contribution of the unresolved secondary arms to the phase transformation. The model is applied to constitutionally undercooled domains with different grid sizes that are larger than the initial secondary arm spacing and much larger than the microscopic tip radius. A similar primary arm spacing is predicted regardless of the grid resolution. However the result obtained with smaller meshes resolve more dendrites branches than with coarser meshes.

1. Introduction
Dendrites are common solidification morphology in both pure materials and alloys. Complex interplay between macroscopic heat and solute transport and microscopic interfacial phenomena results in various dendritic solidification structures. The interaction between dendritic branches and tips are responsible for the features of the grain inner structure. These features are responsible for the macroscopic properties, such as the mush permeability, as well as for the development of freckles and channel segregations. In term of modelling a huge scale gap exists between microscopic phase-field simulations of the dendrites shape, and the volume-averaged simulations dedicated to the prediction of the macrosegregation and freckles. The phase-field method necessitates extremely small grid size to correctly catch the diffusion of solute through the liquid/solid interface. Typically the grid size is several orders of magnitude smaller than the tip radius. Great progress in numerical implementation were recently achieved by using adaptive meshing techniques, multigrid methods, and GPU parallelization. However, even with those improvements the simulations of dendritic arrays still requires considerable computing resources. To fill the gap between the phase-field scale and the full volume-averaged simulation, Steinbach and Beckerman [1-2] have introduced a model where the complex dendrite morphology is approximated with its envelope. The envelope growth model is deduced from the velocity of dendrite tips which are calculated with an LGK model. The melt on the envelope surface as well as inside is assumed to be well mixed and in equilibrium with the solid field. The actual structure of the dendritic solid is not resolved, but a volume-averaged solid fraction is used. The model was successfully applied to equiaxed dendritic interactions, as well as for columnar growth
(Delaleau et al.[3]). More recently, Tourret et al. [4] presented the multiscale dendritic needle network approach to model well ramified dendritic microstructures. The primary, secondary and higher order branches are represented by a network of needles that interact through the solutal field. The range of stable primary arm spacing for an Al-7% Si alloy was calculated.

We present here an alternative approach based on the rescaling of the microscopic growing laws to desired scale of simulation. The aim is to be able to simulate the generation and the growth of primary arms for meshes where the grid size is much larger than the tip radius but still smaller than the expected primary arm spacing, \( \lambda_1 \). A previous work was presented in 2012 [5], where this approach was applied to the macroscale solidification of an NH\(_4\)Cl-H\(_2\)O alloy. In order to catch the smallest solute plumes scale the solidification model simulated directly the envelope of the columnar dendrites. The mushy interior of the dendrites made of secondary and tertiary arms is modelled with a volume-averaged method. The predicted solidification front and the primary arms spacing fitted with the observed ones. The envelope front started from all the walls with a planar front. The growth morphology changed quickly into cellular, and finally bifurcated into a columnar dendritic type [5]. The model was able to predict the occurrence of several flow regimes resulting from the interplay between thermal and solutal natural convections. Flow regimes such as turbulent, stratified and meandering flows were predicted [6-8]. In the present analysis we further develop the theoretical foundation of the model. It will be shown that it is possible to build a dendrite envelope where the tip radius is larger than the real dendrite tip by a specific factor. The factor is directly related to the solid fraction being solidified with the growth of the envelope. The model self-adapts to resolve only the scales that are larger than the chosen mesh size, smaller dendrite arms spacing are modelled with a sub-grid model.

2. The Model

2.1. Rescaling of the microscopic dendritic growth

Let us consider a microscopic dendrite with tip velocity, \( V_t \), radius, and its secondary arms (Figure 1a). The secondary arms that are generated at the dendrite trunk grow and compete, resulting in progressive increase of the secondary arm spacing with the distance from the trunk. Let us consider further a fictitious dendrite that has the same tip velocity but much larger tip radius. The microscopic tips are located inside the volume of the fictitious dendrite renamed as “dendrite envelope”. Similarly to the microscopic dendrite, the dendrite envelope must develop new secondary and tertiary arms. These envelope arms are assumed to surround tightly the major microscopic arms surviving the competition between smaller arms. The envelope contains an undefined volume of liquid, and a volume of solid equal to the solid contained within the real dendrite.

The aforementioned quality of the dendrite envelope can be achieved by assuming that the dendrite envelope solidifies a fraction of solid smaller than 1. We will show later the solid fraction must be equal to the ratio between the envelope tip radius, \( R_E \), and the microscopic real tip radius, \( R_t \) (Figure 1b).

Let us first define the ratio \( f^0 = R_t / R_E \). If the diffusivity is scaled with this factor, \( D_E = D_t / f^0 \), the tip envelope Péclet number becomes strictly equal to the microscopic tip Péclet number:

\[
\text{Pe}_E = \frac{V_t R_E}{2D_E} = \text{Pe} = \frac{V_t R_t}{2D_t}.
\]

This Péclet number is directly related to the melt super-saturation, \( \Omega \), at a distance, \( \delta \), by the exact steady state 3D solution derived by Cantor-Vogel [9]:

\[
\Omega = \text{Pe} \exp(\text{Pe})(E_1(\text{Pe}) - E_1(\text{Pe}(1 + \frac{\alpha}{\text{Pe}})))
\]

Where \( E_1 \) is the exponential integral function. Although the simulations are performed in 2D Hele-Shaw cell, the microscopic tip is small enough so that it develops a 3D paraboloid of revolution
between two plates. The parameter $\alpha$ is the ratio between the distance $\delta$ and solute diffusion layer $\delta_{lV} = \delta/(D_l/V_t)$, in the present study this parameter is chosen to be in order of 1 ($\alpha \sim 1$).

**Figure 1.** Schematic illustration of the envelope model used in the SADE approach. (a) A tight (white) envelope and a loose (red) envelope of an ammonium-bromide crystal in a supersaturated aqueous solution [10]. The SADE model targets to predict a tight envelopes with meshes of sufficiently small resolution scale. If larger grid size are used looser envelopes are predicted. (b) Concept of envelope used in the SADE model. The envelope tip has a much larger tip than the real microscopic tip, but shares the same tip velocity. With the interface advection the real dendrite solidifies totally the volume passed, the envelope solidifies only a fraction of solid equal to ratio between the real to the envelope tip radius.

With the choice of scaling, the original microscopic Ivantsov equation is still valid for the envelope. The tip radius and the solute diffusivity are both modified to conserve the magnitude of the Péclet number. To complete the determination of the tip radius and velocity, the tip selection criterion must also be rescaled. It can be achieved by again rescaling the diffusivity and the Gibbs-Thomson coefficient $\beta$ with the factor $0/sf$ we can build an object which develops a tip radius $R_E$ larger than the real tip radius $R_t$, and growth at the same speed as the microscopic tip, $V_t$. In other words, the Stefan condition at the envelope surface must fulfil

$$V_n (C_S^* - C_l^*) = D_E \frac{\partial C}{\partial n},$$

where $V_n$ is the normal velocity of the envelope-liquid interface, and $\partial C/\partial n$ denotes the macroscopic normal solute gradient. Since the interface curvature and the Gibbs-Thomson coefficient are both rescaled, the curvature undercooling of the envelope is equal to microscopic one. Therefore, the equilibrium concentration at the envelope interface $C_l^*$ is not modified. By simply transferring $f_s^0$ to the left hand side of Eq. (4), we can re-interpret the scaling factor $f_s^0$ as a solid fraction:

$$f_s^0 V_n (C_S^* - C_l^*) = D_l \frac{\partial C}{\partial n}.$$

The desired dendrite envelope can be built by using the normal solute diffusivity $D_l$, a rescaled Gibbs-Thomson coefficient, and by solidifying the fraction $f_s^0$ as the envelope passes.
The applicability of the model will depend mainly on the chosen grid size. The model can be applied in two ways. The first way is to choose $f_s^0$, then the envelope velocity is computed either with Eqs. (2) and (3) for highly curved regions, and with Eq. (5) when the envelope curvature is well resolved. In this case $f_s^0$ must be properly chosen so as not to build an envelope which is larger than the expected primary arm spacing. The second way is to shift the envelope with the microscopic tip velocity, Eqs. (2) and (5), and compute the solid fraction that would fulfill Eq. (3). After some simple algebra, Eq. (3) can be rewritten in the following form:

$$f_s^0 = \frac{\Gamma}{2|m_L|\left(C_s^* - C_i^*\right)\sigma R_e Pe}.$$  

Since the Péclet number is fixed, the required solid fraction to be solidified with the envelope is proportional to the tip curvature. This fraction is calculated every time step at the tips, and kept constant for further lateral growth where Eq. (5) is applied.

While the main tip envelope growth considers only one real tip is considered, the lateral growth represents the growth of numerous unresolved secondary arm tips. This situation is more difficult because the competition between secondary arm tips cannot be directly resolved. In order to keep the model simple, we consider the number of arms fixed. The grid size is assumed to be sufficiently large so that only one tip survives when crossing the next volume elements. These secondary arms will then be considered as a new lateral “primary” tip. New branches (primary, secondary and tertiary) are emitted by the inherently unstable growth of the dendrite envelope surface.

### 2.2. Volume averaged equations and sub-grid model for the unresolved secondary arms

The 2D isothermal equations governing conservation of species and phase fraction are written as follows

$$\frac{\partial (\rho f_s C_i)}{\partial t} = \nabla \cdot (f_s \rho D_i \nabla C_i) - \rho C_s \frac{df_s}{dt}$$

$$\rho \frac{df_s}{dt} = f_s^0 \int V dl + M_{la}$$

where $f_s$, $f_s^0$, $C_i$, $D_i$, and $\rho$ are the liquid and solid fractions, the concentration of the solute (NH4Cl), the solute diffusivity and the density. The terms on the right hand side of Eq. (8) stand for the mass transfer through an element of the envelope surface $dl$, and $M_{la}$ the sub-grid mass transfer due to the growth of unresolved secondary arms (see Eq. (10)). Depending on whether the envelope represents a tip or a lateral growth, the front velocity $V$ represents $V_t$ or/and $V_a$. The mush topology can be represented as parallel or crossed cylindrical sticks. We use here the multiphase volume-averaged approaches developed by Wu et al. [11]. This model was developed for periodic alignment of primary arm columnar trunks. Two arms are considered as unresolved if the distance that separates them is smaller than one grid size. The resolved arms are generated by instabilities of the dendrite envelope surface. Thus, the model does not expressively separate primary and secondary arms in the classical meaning; the number of resolved arms depends on the choice of mesh resolution. By “resolved” we mean that the presence of arms are reflected in the variation in volume fraction. The model of Wu et al. [11] is used in the present analysis with an average secondary arms spacing, $\lambda_a$. The number of secondary arms that germinates along the trunk can be estimated from the observation that the ratio $\lambda_s^0/R_e \approx 5$ [12] is approximatively constant, $\lambda_a^0$ represents the initial spacing. The initial number of sub-grid arms that grow in lateral directions is $\Delta x / \lambda_a^0$. This number is assumed constant within the volume element where they are generated. For the volume elements that has been passed by the envelop front, a diffusion-controlled growth model around cylindrical trunks is assumed. The growth velocity in the radial direction of a cylindrical trunk is assumed very slow, so that a quasi-steady growth can be used.
\[ V_a = \frac{D_a}{R_a} \Omega \ln\left(\frac{R_a}{R_a}\right) \]  
(9)

where \( R_a \) is the average radius of the unresolved arm trunks, \( R_a = \ell / 2 \) is the maximum packing radius of a secondary dendrite arm trunk. For our 2D dendrites, the trunks are assumed to be parallel and lying on the same plane, so that the surface area of the secondary arm trunks per volume is \( S_a = 2\pi R_a \left(\frac{\ell}{2}\right)^2 \). The net volume-averaged mass-transfer rate for elements within the inner dendrite envelope mushy region is:

\[ M_{la} = V_a \left(\frac{2\pi R_a}{(\ell^2)}\right) \rho f_j \]  
(10)

Table 1: Boundary conditions and effective parameters used for simulations.

| Property/Symbol | Value | Property/Symbol | Value |
|-----------------|-------|-----------------|-------|
| \( C_0 \)       | 0.3   | \( C^*_s \)     | 1     |
| \( C_j^* \)     | 0.266 | \( \alpha \) (Eq 1). | 1.0   |
| Temperature     | 17 °C | \( \rho_l \)     | 1078 kg/m³ |
| \( \beta \) (Ref.[13]) | 12.6e-18 | \( m_L \)      | 4.8 K   |
| \( \Gamma_j \)  | 8.05e-5 K m | \( D_L \)   | 2.5e-9 m²/s |

3. Application of the model

The dendritic envelope model is applied to the solidification of a H₂O-30wt.%NH₄Cl alloy at 290 K (\( \Omega = 0.1 \)), with the thermophysical properties given in Table 1. To illustrate the capability of the model to self-adapt to different mesh sizes, the simulations are performed with the same grid, but with two different scales. The first scale is 20 mm x 200 mm with a grid size of 200 \( \mu m \) (Fig. 2) and the second 0.5 mm x 5 mm with a grid size of 50 \( \mu m \) (Fig. 3). In order to prevent the interface from remaining flat, a random factor is applied to the computed front velocity at a level of 1%. Initially one primary tip is introduced per volume element. Table 2 summarises some quantitative results. The initial interface (mush/liquid) is initially assumed flat on the right half of the bottom boundary. The second half on the left is perturbed with a sinusoidal. In Fig. 2b the interface develops kind of lamellae structures which very quickly transforms into columnar. The “secondary” arms are generated as soon as the distance between primary vertical arms is large enough (Fig. 2d). These arms represent the resolved part of the secondary horizontal arms. The unresolved arms are invisible, hidden in the solid fraction predicted in each volume element. The solid fraction reaches its maximum on the arm axis at about 7 %. It can be noticed that few volume elements are almost free of solid.

During the growth (Fig. 2b-f) the number of primary arms decreases by a mechanism of competition. In the very beginning \( \lambda_0 \sim 0.4-0.8 \) mm, in the end of the calculation \( \lambda_0 \) in average much larger 0.6-3 mm. As could be expected different spacing developed on the perturbed left side compared to the right side. It is interesting to notice that the “regularly” perturbed side develops a more irregular interface (Fig. 2b-d). This irregularity leads to a faster selection of the primary arm spacing compared to the right side. However even in the right hand side, \( \lambda_0 \) seems to tend towards \( \sim 3 \)mm. At the later stage (Fig. 2e-f), a relatively steady tip velocity (\( \sim 450 \) \( \mu m/s \)) is reached which correspond to an effective tip super-saturation of 0.06 smaller by 40% from the global super-saturation (\( \Omega = 0.1 \)).
The simulation is now performed on a mesh which is 4 times finer. The initial state assumes a perfectly plane interface. The very first developments shows an irregular growth of two islands (Fig. 3a). Later columnar structures with side branches become visible. The dendrite envelope is now able to resolve tertiary arms (Fig. 3d-f). These tertiary arms transform in new primary arms when the conditions allow it (Fig. 3d-f). It must be emphasized that even in this higher resolution case many secondary arms are still unresolved. In each volume element located on an arm axis, 7 to 20 small unresolved arms are predicted according to the ratio $\lambda_0^2 / \lambda$. These arms are considered within the sub-grid model and are assumed to not develop more than one grid size in length. As in the previous case the competition between the numerous initial primary arms result in the progressive increase of $\lambda_0$, from 0.15-0.5 mm in Fig. 3a to 0.4-5 mm in Fig. 3f. Although not yet stable the primary spacing predicted with the finer mesh is very similar to the one predicted with the coarser mesh. Due to the smaller mesh size, the maximum solid fraction reached in the dendrite trunks is naturally higher 13% (compared to 7% previous calculation). Due to the higher resolution of the dendrite envelope surface, the volume elements which are almost free of solid (in blue) are more numerous than in the coarser case.

![Figure 2. Solid fraction (0-0.1(red)) of columnar growth simulated with the dendrite envelope model on a low resolution mesh (domain width: 2 cm wide, grid size: 0.2 mm).](image)
The concentration field near the dendrite tips is shown in Fig. 4. In the inter-dendritic region the equilibrium concentration (~0.266) is reached at a distance of about 3 mm from the liquid/envelope surface, at larger distance solidification is thermodynamically stopped. The model predicts the same decreasing length for both high and low resolution simulations.

After an initial transient regime, the front made of envelope tips reaches a relatively stable velocity (~360 µm/s). Some strong fluctuations exist especially during the elimination of trunks due to competition between the tips. Around 20% difference exists in the predicted steady tip velocity between the coarser and finer mesh size simulations. The main reason lies in the fact that the domain used in the simulation with the finer mesh is probably not long enough to reach a final steady state. Another problem is the correct estimation of the concentration field ahead of the tip, which in the present model depends on the solidification rate within the first cell containing the envelope surface. The concentration of interest is at a distance $\delta = D_Y/V$ of the tip, which depending on the mesh accuracy is close but not strictly equal to the concentration given by the volume averaged equation (Eq. 7). The ratio of the envelope to the microscopic tip radius $R_E/R_T$ is typically in the range of 120-250 for the coarser mesh and in the range of 40-60 for the finer mesh.

![Figure 3](image)

**Figure 3.** Solid fraction (0-0.2 (red)) of columnar growth simulated with the dendrite envelope model on the higher resolution mesh (domain width: 5 mm cm wide, grid size: 0.05 mm).
Figure 4. Zoom on a typical concentration field near the tips of the dendrite envelopes predicted with high (left) and low resolution (right) simulations (0.266 (blue), 0.3 (red), domain height: 4 mm).

Table 2. Summary of some quantitative results.

|                     | Coarse mesh       | Fine mesh       |
|---------------------|-------------------|-----------------|
| Mesh size           | 0.2 mm            | 0.05 mm         |
| Calculation domain  | 20x100 mm²        | 5x20 mm²        |
| Dendrites trunk solid fraction | 7 %             | 13%             |
| Steady tip velocity | 460 µm/s          | 360 µm/s        |
| Final $\lambda_1$   | 0.6-3 mm          | 0.4-5 mm        |

4. Final remarks and conclusions

We presented a dendrite envelope approach based on the rescaling of the microscopic growing laws to desired scale of simulation. This is an alternative approach based on the rescaling of the microscopic growing laws to desired scale of simulation. The aim is to be able to simulate the generation and the growth of primary arms for meshes where the grid size is much larger than the tip radius but still smaller than the expected primary arm spacing, $\lambda_1$. This model is inspired from the methodology of the turbulence models that filter the scales that are smaller than the simulation grid size, such the Large Eddy Simulation (LES) model. The elimination of the small scales from the Navier–Stokes equations reduces the computational cost of the simulation with respect to the Direct Numerical Simulation (DNS). The latter resolves every scale of the solution, is prohibitively expensive for almost all geometry or flow configurations. In the LES model the equations are transformed by using a Sub-Grid Scale (SGS) model which implicitly accounts for the eddies smaller than the filter width (grid size). A large number of specific SGS grid-dependant models was built according to the turbulence theory and the available computational resources. Only the large scales of the flow field are resolved allowing better fidelity than alternative approaches based on the Reynolds-Averaged Navier-Stokes (RANS) methods. It is noteworthy that most of the results of LES are grid dependent, however, depending on the capability of the sub-grid scale model, statistical grid independence can be achieved.

If a parallel can be made between the field of dendritic solidification and turbulence, the contrast between the phase-field/sharp interface and the volume-averaged methods can be represented by the contrast between the DNS and RANS methods. Just as for the LES with turbulent eddies, the SADE approach models the smallest (and most expensive) scales of the solution, rather than resolving them as phase-field does. The simulated dendrites are grid-dependant. However if simulations on two different meshes are performed, only the mean and higher order statistics of results must be compared. In the present work we have focused on the largest scale of the variations of the solid fraction (i.e. $\lambda_1$) and the concentration field. If the mesh is sufficiently small, the ratio $f_{10}^0 = R_1 / R_g$ will tend towards 1, the SADE model transforms into a classical sharp-interface method for dendritic growth.
If we persist in the comparison between LES and the present model, a great effort should be put on the development of two sub-grid models, the first is for the growth and competition of the unresolved arms, and the second for an accurate estimation of the thermo-solutal conditions ahead of the dendrite tips.

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