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A modified cellular automaton method for polydimensional modelling of dendritic growth and microsegregation in multicomponent alloys

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Abstract. Numerous numerical models for simulating solidification of metals on a microscopic scale have been proposed in the past, among them are most importantly the phase-field method and models based on cellular automata. Especially the models based on cellular automata (adopting the virtual front tracking (VFT) concept) published so far are often only suitable for the consideration of one alloying element. Since industrial alloys are usually constituted of multicomponent alloys, the possibility of applying cellular automata is rather limited. With the aim of enhancing this modelling technique, a new, modified VFT model, which allows for the treatment of several alloying elements, in the low Péclet number regime is presented. The model uses the physical fundamentals of solute and heat diffusion in two dimensions as a basis for determining the solidification progress. By a new and effective approach, based on a functional extrapolation of the concentration gradient, dendritic growth in multicomponent Fe-C-Si-Mn-P-S alloys could be studied. The model shows the typical behaviour of dendritic solidification, such as parabolic tip and secondary dendrite arm formation as well as selection of preferably aligned columnar dendrites. A validation of the model is performed by the evaluation of morphological parameters and comparing them to experimentally determined values. The results for free and constrained dendritic growth effectively demonstrate the capabilities of this new model. The model is especially attractive for bridging the gap between one-dimensional microsegregation models and multidimensional morphology models with regard to modelling the complex interrelations between segregation on a multidimensional level and morphology formation.

1. Introduction

Generally speaking, the formation of microstructure during solidification is partly the basis for the properties of a metallic material. Regarding solidification-related influences on the properties, the following two factors are essential: On the one hand the topology of the microstructure itself and on the other hand the physical and chemical implications of the microstructure (in alloys most explicitly seen by solutal segregation, \textit{i.e.}, microsegregation); both items are inevitably connected to each other. Specific and very thorough research on either phenomenon has been carried out in the past and is published in the relevant textbooks \cite{1, 2}.

In the last years special attention has been paid to providing appropriate modelling techniques especially for deepening the understanding of microstructure formation. In the literature three
different approaches to microstructure modelling can be identified: Firstly, front tracking models [3–5] are used to precisely reproduce the interface between solid and liquid by a dynamically refined meshing in the region. Secondly, Phase-Field (PF) models deal with the solid-liquid interface by introducing a continuous transitional layer of finite thickness using an additional PF quantity, thus eliminating the problem of explicitly tracking the interface. Several reviews on the methodology and capabilities of the PF models are available [6–10]. Thirdly, cellular automata (CA) were either employed for mesostructure simulations during solidification (when coupled with finite element modelling, this technique is referred to as CA-FE modelling [11–13]) as well as for modelling microstructure. Early attempts were published in [14] and in [15], however having the drawback of lacking an arbitrary dendrite growth direction due to the mesh anisotropy. This drawback was resolved in [16, 17] where the authors developed a virtual front-tracking (VFT) method of the solid-liquid interface.

In any case, most of the above models were developed for simulating only binary systems (mostly Al-Cu, Al-Si or Fe-C). Despite the remarkable reproduction of dendrite morphology, columnar as well as equiaxed solidification by VFT models, the application of such models to industrial materials is limited.

Next to the question of microstructure modelling, the effect of microsegregation is of key importance on material properties. Kraft and Chang [18] give a review on more than 60 different microsegregation models that were published in the last century, a number which emphasises the importance of the topic. Virtually all models mentioned in this publication feature the study of microsegregation either by an analytical or a numerical one-dimensional approach. The models mostly differ in the topology of the calculation domain, in the number of alloying elements and phase boundaries and in their assumptions regarding the solute diffusivity in liquid and solid. However, most models have the shortcoming of neglecting physical effects (such as interface curvature); moreover, some models even do not account for thermal and solutal diffusivity. Furthermore, the interfacial growth velocity is often derived from empirical relations. Although one-dimensional models are an excellent basis for a locally focused consideration of solidification, physically correct results are questionable when the problem domain exceeds the very focused local interest of solidification.

In order to provide a possible improvement to the above mentioned shortcomings of microstructure models on the one hand and microsegregation models on the other hand a modified CA approach with the VFT scheme has been developed which considers multicomponent alloys in several dimensions. The basics of this model have been published in [19]; the present contribution will shortly summarise the model assumptions and then show some of the results to illustrate the model capabilities.

2. Short Model Outline
The developed model features a two-dimensional consideration of the microstructure formation and the microsegregation by the technique of cellular automata. In the model each cell is characterised by a number of basic quantities (where $i$, $j$ and $t$ are spatial and time indices):

- centre coordinates $x(i, j)$
- temperature $\vartheta(i, j, t)$
- concentration $C^\tau_v(i, j, t)$ of the alloying element $v$ in phase $\tau$ (here $\tau = L$ when liquid or $\tau = S$ when solid)
- fraction of solid $f_S(i, j, t)$
- interfacial curvature $\kappa(i, j, t)$
- growth direction $\varphi(i, j)$

According to the CA technique, the quantities in each cell at a time step $t$ will results from the neighbouring cells’ values in the current time step $t$ and the preceding time step $t-1$. Therefore,
the model procedure can be summarised as follows: Starting from a fully balanced liquid system, the chemical equilibrium is disturbed by artificially placing nuclei in the whole domain. Thus, certain cells’ state is set to solid and the rejected solute is distributed to its neighbours. Due to the governing laws of solute diffusion, the concentration of neighbouring cells will then again be influenced. Additional cooling of the system will also cause thermal convection in the system. Both influences will cause the solid phase to grow (or dissolve), rejecting further solute and latent heat of fusion. Owing to the growth of the solid phase, the curvature of the nuclei will steadily change, also affecting the interfacial conditions.

The rules for determining the quantitative values of each cell are governing physical laws which are iteratively applied. These rules are already described in a published work [19] and will therefore not be repeated here. Nonetheless, it is important to mention that when modelling microstructure formation multicomponent alloys (i.e., more than one alloying element in the matrix) the combination of solute diffusion and interfacial growth due to chemical (solutal) undercooling leads to an ill-posed problem due to the direct dependence of either phenomenon on each other. In the present model, the ill-posed problem is overcome by a functional extrapolation of the concentration gradient towards the solid-liquid interface (shown in Fig. 1) which yields the unknown interfacial concentration \( C^{\nu,*} \). Details to this approach are derived in [19].

![Figure 1. Illustration of the equilibrium concentration extrapolation at the interface.](image)

2.1. Model Simplifications and Assumptions

The model uses the following simplifications and assumptions:

(i) The solid-liquid interface is not treated explicitly in the thermal field, in contrast to the treatment in the solutal field. As no heat balance is made over the interface, dendritic growth driven by thermal diffusion rather than by chemical undercooling cannot be reproduced.

(ii) Kinetic undercooling is not considered in the model which limits its application when treating rapid solidification conditions.

(iii) Cross diffusion between the alloying elements and a dependence of the partition coefficient on more elements than the superimposed binary systems is not considered, also partially lacking appropriate material data. Nonetheless, it is believed that there is a considerable influence of a multicomponent functionality of the partition coefficient, which should therefore be resolved in near future.

(iv) Fluid flow is not considered.
3. Model Results
In the following, some exemplary results of simulations with the model shall illustrate the potential of this novel approach. The material parameters used for the present study are the same as in [19]. In advance, it should be stated that fundamental comparisons of the model results with well-established analytic models—like the LGK-model [20] and the microsegregation model by Ohnaka [21]—have been carried out in advance to this study and are published in [19].

3.1. Equiaxed Solidification of a Single Nucleus
At first, an equiaxed dendrite in the system Fe-0.6%C-0.5%Si-0.5%Mn-0.005%P-0.005%S in a domain size of $100 \times 100 \, \mu m^2$ is considered. The initial conditions of the simulation are uniform liquidus temperature and a single, one-celled seed in the domain centre. The domain is globally cooled at all sides with a heat transfer coefficient of $500 \, \text{W m}^{-2}\text{K}^{-1}$. Figure 2 shows the concentration fields for all alloying elements after a solidification time of 0.02 s.

It can be seen that typical dendritic features with branching and development of secondary dendrite arms develop, whereby the generation of secondary arms is only a product of interfacial curvature. It should however be highlighted that the secondary dendrite arm spacing which can be observed in the present example is not comparable to the final secondary dendrite arm spacing as the mechanisms of selection and coarsening are not visible at this solidification time (the model however features these mechanisms as shown later). Moreover, due to the differences in partition coefficients and diffusivity, considerable differences in the distribution of solute are obvious.

![Figure 2](image_url)

**Figure 2.** Concentration Fields for Free Dendritic Growth in a Multicomponent Fe-Si-Mn-P-S-alloy after a Solidification Time of 0.02 s.

3.2. Columnar Solidification
As solidification usually occurs in columnar form, a rectangular model of $300 \times 100 \, \mu m^2$ with an initially solid first row of boundary cells at one of the longer sides was taken as a basis
to investigate the model’s behaviour in the quaternary system Fe-0.6%-0.5%Si-0.5%Mn. The domain was cooled with a heat transfer coefficient of 500 W m$^{-2}$K$^{-1}$ at this particular face; adiabatic isolation was prescribed at the remaining faces.

Thus, not only the behaviour of the model regarding the formation of irregularities (finally resulting in dendrites) but also the resulting interdendritic concentration profiles can be evaluated. Figure 3 shows the C-distribution after a solidification time of 0.3 s in the total computational domain—the solidification direction is downwards. It can clearly be seen that out of the initially planar front, primary dendrites developed; the selection of dendrite arms oriented more ideally (almost opposite the heat extraction direction) is evident. The results in the lower part of the domain must be regarded carefully, since the dendrites could not grow further (no mass flow) and thus are starting growth normal to the interface. Next to the formation of primary dendrites, the development of secondary arms is also well visible. In accordance with many practical observations, this formation is more or less pronounced, depending on the available diffusion length (i.e., distance to the next primary arm).

![Figure 3. Simulated C-concentration field after 0.3 s.](image)

### 3.2.1. Differences in Interdendritic Enrichment

Figure 4a shows a highlighted selection of Fig. 3. An interdendritic point marked P1 in this figure, lying well between the tip and the trunk of the two secondary arms, is used to demonstrate the differences in interdendritic segregation depending on the distance from the dendrite trunk. It is compared to position P2 which represents the enrichment at the triple point (meeting of two secondary dendrite tips and a different primary dendrite arm).

The results in Fig. 4b show the significant changes regarding the maximum value of segregation for the two cases: Silicon, showing the strongest segregation behaviour, enriches from a starting value of 0.5 % to 3.8 % between the dendrites—at the triple point this value rises to 7.4 %. In terms of the segregation ratio $C_L/C_0$ this corresponds to a factor of 7.6 interdendritically and 15 at the triple point. Pertaining to the segregation behaviour of Manganese, again a considerable increase in the segregation ratio is detected: here, a factor of 2.4 versus an increase by 3.6$C_0$ at the triple point. Regarding the profile of Carbon, an equalisation of the composition occurs at the triple point, leading to approximately the same segregation ratio as observed interdendritically ($C_L/C_0 = 3.3$).

Although it has to be pointed out that these absolute values are still rather qualitative (using non-constant diffusion parameters possibly changes the composition fields), it becomes clear that the present model is generally able to predict these variations of a larger scale than a conventional microsegregation model. Several solidification-related phenomena like for example macrosegregation or hot tearing involve a polydimensional view of the issue of microsegregation.
3.3. Influence of Cooling Rate

The effects of changing cooling rate on a solidification structure are well known. In a second example with the same preconditions as the previous (mesh size and alloy), the effect of cooling rate on the microstructure shall be compared to values observed in reality as well as to analytical derivations for multicomponent alloys from the literature. In [22] the authors deduce a relationship between the secondary dendrite arm spacing and the solidification time. Since this empiric relation is based on observations at room temperature (i.e., coarsening of dendrite arms has already taken place), evaluations of the present model are also conducted at a late stage of solidification. Furthermore, in [23] dendritic solidification in multicomponent alloys is treated from the analytical viewpoint. In this work, one finds a relationship for the secondary dendrite arm spacing as a function of local solidification time and enrichment in the liquid for multicomponent alloys. Both studies are taken as a basis for the following comparison.

Figure 5 shows the Si-distribution for a late solidification stage according to the previously mentioned conditions, however for an increased cooling with \( h = 1000 \text{ W m}^{-2}\text{K}^{-1} \); the formation of secondary arms is obvious. As mentioned before, not all secondary arms would actually survive the process of coarsening. Therefore, only those secondary arms which exhibit a clear tendency for growth and which will not suffer from a potential remelting or from overgrowth by a near neighbour were selected for distance measuring. In the present example, these dendrite trunks are marked with a short white dash in the figure.

The numerical results of the evaluation for different cooling rates are displayed in Fig. 6. The solidification time was determined from the simulation, i.e., it represents the time when the last gaps between the dendrites close in the system. Additionally, the figure shows the prediction of the model by [22]. It has to be stated that this model was actually derived for higher solidification times (at least several seconds). However, for reasons of computation time, such simulations cannot be performed with the present model in a reasonable period of time. The figure additionally shows the results of the model by [23]. The final composition of the liquid at the base of the dendrite —as a necessary input parameter for this model—was determined from the simulation. The consistency between the present model and the results derived from
experimental observations as well as those from an analytical approach to the topic is remarkable and underlines the correctness of the assumptions in the presented model.

**Figure 5.** Si-distribution after a solidification time of 0.25 s for $h = 1000$ W m$^{-2}$K$^{-1}$. The white dashes indicate the origins of the secondary arms that were considered for the $\lambda_2$-evaluation.

**Figure 6.** Comparison of $\lambda_2$-spacings calculated with the present model to those of [22] and [23]; heat transfer coefficients $h$ in W m$^{-2}$K$^{-1}$.

### 4. Summary and Conclusions

In the present paper a recently developed model for the simulation of microstructure formation and microsegregation for multicomponent alloys has been used to study several model cases and to evaluate the capabilities of this model.

From the perspective of microstructure it could be shown that the model features unconstrained equiaxed dendritic growth, the evolution of dendritic solidification structures
from a planar front and the development of secondary dendrite arms. Moreover, the influence of cooling rate on the secondary dendrite arm spacing was evaluated and compared to results from practical observations.

Next to the microstructural viewpoint, the phenomenon of microsegregation was investigated. It could be shown that by a multidimensional view on the problem of microsegregation, substantial differences regarding the microsegregation between two secondary dendrite arms and the enrichment of solute at a triple point are observed. This result becomes highly important when calculating local solidification temperatures as these differences cause a considerable decrease of the solidus temperature.

It is believed that the present model bridges the gap between phase-field models, which feature an exact resolution of dendritic features—however, at very high computational effort—and macrostructural solidification models. Thus, phenomena which are currently of high practical relevance in the field of casting of steel can in future be addressed by a quicker means than previously available. Examples of such questions are the columnar-to-equiaxed transition, the formation of hot tear segregations (which also involves the introduction of a mechanical or pseudo-mechanical model) or the study of the effects of rapid solidification conditions on the formation of segregation patterns.

In order to bring the model even closer to practise, several modification steps are planned in the future: Firstly, by obvious modification of the algorithm a second phase boundary shall be achieved. Moreover, temperature and concentration-dependent material properties shall be employed. Later, the upgrade of the model to a three-dimensional treatment of solidification should be persued by obvious modification of the presented equations. In order to further assess the quality of the model, comparisons to phase-field models are planned.

References