Investigation of the microstructure adjustment by velocity variations during the directional solidification of Al-Ag-Cu with the phase-field method

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Abstract. Directional solidification is a favored process to manufacture homogeneous microstructures which lead to macroscopically unique properties for a material. The dependence of the spacing and type of the arising microstructure from the solidification velocity for constant velocities is extensively investigated. However the effect of changes in the solidification velocity on the resulting microstructure adjustment processes is still unclear. Therefore large-scale (3D+t) simulations of the ternary eutectic system Ag-Al-Cu with changing solidification velocities are conducted with a phase-field model based on the grand potential approach. To study the spatially complex rearrangement process during velocity changes in statistical representative volume elements, simulations with different velocity profiles are calculated in large-scale domains. The results show, that the evolving microstructure continuously rearranges by splitting and merging of the rods despite constant growth conditions. By increasing the velocity, the microstructure refines by splitting of the Al\textsubscript{2}Cu phase. Whereas by decreasing the velocity, the microstructure coarsens by overgrowing events of both intermetallic phases.

1. Introduction

The process of directional solidification allows to produce different tailored materials with long ranges of homogeneous microstructures and consistent properties. The microstructure characteristics can be controlled by the applied process conditions. Especially the spacings and phase arrangements in the microstructure can be influenced by the externally applied solidification velocity. Despite the well understood correlation between the spacings of the evolving microstructure and the solidification velocity [1], the adjustment processes in the microstructure during a variation of the velocity is in focus of current research. An increase of the solidification velocity leads to a refinement whereas a decrease leads to a coarsening of the microstructure [2, 3, 4, 5]. In ternary eutectic systems a velocity change during directional solidification leads to split, merge, overgrowth and nucleation events as well as to a possible change of the overall evolving microstructural arrangement of the rods and lamellae. To investigate these complex three-dimensional adjustment processes, 3D samples of the microstructure are needed. Experimentally, a high technical effort like synchrotron
tomography [6, 3] is required, to observe these rearrangement processes. Beside experiments and theory, simulations have become a powerful method for the scientific investigation of physical phenomena. For the three-dimensional investigation of the diffusion driven phase transformations in the microstructure during solidification processes, the phase-field method has been established [7].

To resolve representative volume elements with complex spatial microstructure arrangements comparable to experimental micrographs [8, 9, 10], large simulation domains are needed. Further, the calculation on current high performance computers is crucial to solve the compute-intensive models in the necessary large-scale domains in reasonable time. Therefore massively-parallel and highly optimized solvers, like PACE3D [11, 12] and WALBERLA [13, 14], are required. By using optimized and scalable solvers on high performance systems, the directional solidification of binary [8, 5] and ternary eutectic reactions [15, 2, 10, 16, 17, 18, 19] is investigated in large-scale phase-field simulations. A detailed description of the used model can be found in [20, 21, 10]. The implementation of the model is presented in [10, 14, 22, 12].

Two- and three-dimensional simulations of the microstructure adjustment processes due to velocity variations during directional solidification for the two-phase eutectic system NiAl-34Cr are performed in [4] and [5], respectively. In these works, separate acceleration and deceleration steps of the temperature gradient velocity are used to realize variations in the solidification velocity. A comparison between three-dimensional phase-field simulations and two experiments of the ternary system Ag-Al-Cu with velocity variations is given in [2]. In the experiments a single velocity increase and decrease is executed, respectively, whereas the conducted simulations include multiple subsequent velocity variations. A pure experimental observation via synchrotron tomography of a decelerated solidification of Ag-Al-Cu is published in [3]. For this study the same analysis methods using a graph based approach are used as for the investigation of the three-dimensional phase-field simulations in [2] and in this work.

Due to the complexity of the microstructure adjustments in simulations with multiple velocity variations, large-scale 3D simulations of the system Ag-Al-Cu with single controlled acceleration and deceleration steps are conducted in this work to identify the underlying mechanism controlling the rearrangement processes in ternary eutectic systems.

2. Results and discussion

To investigate the microstructure rearrangement mechanisms during a velocity change, the ternary eutectic system Ag-Al-Cu is used. The simulation setup is presented together with a summary of the material and process parameters in [10, 2]. During the solidification the matrix phase Al and the two intermetallic phases Ag2Al and Al2Cu evolve simultaneously. The velocity changes are realized by variations of the temperature gradient velocity $v_G$. The solidification velocity adjusts according to the changes in the analytic temperature field. The velocity variations are performed similar to [5] with deceleration and acceleration steps instead of a jump function like in [2]. Due to the unnaturally enlarged temperature gradient, which is used in the conducted system, to reduce the computational effort [2], the acceleration and deceleration step can be performed in less than 20 000 time steps. One simulation is decelerated from 0.325 µm/s to 0.25 µm/s, which corresponds to a velocity decrease of 30%. The other simulation is accelerated from 0.175 µm/s to the same end velocity of 0.25 µm/s, resulting in a velocity increase of 30%. It is assumed that the microstructure refines during an acceleration of the solidification by splitting and nucleation of the rods and coarsens during a deceleration by merging and overgrowing of rods. To investigate these rearrangement events, the graph-based analysis approach described in [3] is used. Despite rarely observed nucleation events found in [3] it is assumed that for low solidification velocities the nucleation barrier is to high [2, 23]. Hence no nucleation mechanism is considered in the model. The simulations with a computational domain size of 800 × 800 × 275 voxel cells are calculated on 11 264 cores for approximately 40 h on the Hazel Hen supercomputer.
Figure 1. Computed micrographs perpendicular to the solidification direction (left and right images) and selected rods (central images) showing the effect of velocity variations.

[24]. By applying a moving window approach, this results in a growth height of 14,672 voxel cells for the deceleration and 10,950 voxel cells for the acceleration simulation. The difference in the growth heights is caused by the different initial velocities.

An overview of the two simulation results is composed in fig. 1. The two intermetallic phases, $\text{Ag}_2\text{Al}$ and $\text{Al}_2\text{Cu}$ are shown in green and blue, respectively, and the matrix phase $\text{Al}$ is indicated in red. The rod diameters adjust following the velocity-undercooling-spacing relationship described by Jackson and Hunt [1]. For both simulations, computed micrographs parallel to the solidification front are depicted before and after the velocity change and at the end of the simulation. The overall evolving pattern remains the same for both simulations. Selected rods of the intermetallic phases over the growth height are highlighted in the center of fig. 1. For the deceleration simulation (left), an increase of the rod size is observed, whereas the rod size decreases for the acceleration (right). In both simulations the spacing in the microstructure adjusts by multiple split, merge and overgrowth events. Also multiple sequences of splitting and afterwards overgrowing of one splitted arm are observed in both intermetallic phases. For a better visualization of the microstructure adjustment events selected examples are magnified. The selected rods indicate that the adjustment processes are not only driven by the corresponding events for refinement and coarsening. Splitting events, leading to a refinement of the microstructure, are also multiple times observed in the deceleration simulation as well as overgrowing events, leading to a coarsening of the microstructure, in the acceleration simulation.

To analyze the microstructure events quantitatively, the area of the two intermetallic rods perpendicular to the solidification direction at the events is plotted over the growth height in figs. 2 and 4. The type of event is indicated by the symbols and the phases are marked by the colors. Due to the continuous structure of the $\text{Al}$ matrix, split, merge and overgrowing events lead not to separate parts of this phase and hence they are not detected. In fig. 2 more events can be observed before the velocity decrease. Despite the similar phase fractions of the intermetallic phases of $\sim 1/4$ and the similar number of rods, the rod area at which they split and merge differs. The area of the $\text{Al}_2\text{Cu}$ rods is approximately twice as large as of the $\text{Ag}_2\text{Al}$ rods. For a better comparison of the event occurrence rate, the average number of events per cell over the growth height is shown in fig. 3. Whereas in the beginning the microstructure adjustment is dominated by splitting and merging events, after the velocity decrease, mostly
splitting and overgrowing events occur. For a growth height less than 1 000 µm a higher number of events is observed due to the evolution of the artificially used Voronoi tessellation as initial condition to a stable chain-like pattern [25]. After self similar growth is reached for a constant velocity, the number of events is stable. After the velocity decrease, the number of overgrowing events increases, leading to a coarsening of the microstructure. Due to a rod area of zero for overgrowing events, the symbols in fig. 2 are strongly overlapping. Therefore the peak of the overgrowing events in fig. 3 is not visible identifiable in fig. 2. A similar behavior of the events as in fig. 3 is also observed experimentally in [3, 2].

Figure 2. Area of the rods perpendicular to the solidification direction at which the microstructural adjustment events take place over the growth height for a growth velocity decrease.

Figure 3. Average number of the microstructural adjustment events per cell over the growth height using a moving average approach for a growth velocity decrease.

In fig. 4 the rod size at the occurring events over the growth height is plotted for the velocity acceleration. Before the velocity increase, the number of events is reduced in contrast to the higher velocity afterwards. Similar to the deceleration simulation, different rod areas at which the events occur can be observed depending on the phase. Also more events occur initially due to the Voronoi tessellation until stable growth is established. Despite the overall refinement of the microstructure, multiple overgrowing events can be observed after the velocity increase. This is also indicated in fig. 5 by the average number of events per cell over the growth height. To refine the microstructure, various rods split as shown by the peak directly after the velocity change. Thereby mostly the blue Al₂Cu rods split as displayed in fig. 4.

Figure 4. Area of the rods perpendicular to the solidification direction at which the microstructural adjustment events take place over the growth height for a growth velocity increase.

For both simulations, the events to adjust the spacing occur with a delay after the new solidification velocity has been established. This behavior is in accordance with the observations of [5]. The number of the microstructural events is found to be dependent on the growth rate.
and increases with higher solidification velocity. Despite constant growth velocities, an ongoing rearrangement process of the two intermetallic rods by splitting and merging events is observed. Thereby a split event of one rod often leads to a merge of rods of the other phase and vice versa. This is also reported in experimental data in [3] and in phase-field simulations [2]. Independent of the initial velocities, both simulations have a similar number of ongoing events for the same imprinted final velocity. For all investigated velocities the size of the rods at which the event occurs is larger for the $\text{Al}_2\text{Cu}$ rods than for the $\text{Ag}_2\text{Al}$ rods. Despite the ongoing rearrangement at constant velocities, the coarsening of the microstructure is dominated by overgrowing events and the refinement by splitting events. This is in accordance to phase-field simulations with velocities variations of the two-phase eutectic system $\text{NiAl-34Cr}$ as presented in [5] and of the ternary system $\text{Ag-Al-Cu}$ shown in [2]. Similar findings are found as well in experimental data of $\text{Ag-Al-Cu}$ containing a velocity decrease [3]. In contrast to the investigations of the pseudo-binary two-phase eutectic system $\text{NiAl-34Cr}$, the number of events is higher and the rearrangement process is more complex in ternary systems. In order to understand the basic mechanisms, more studies of defined and idealized setups are needed.

3. Summary and conclusions
In this work the influence of single independent velocity variations on the microstructure rearrangement process of the ternary eutectic system $\text{Ag-Al-Cu}$ during directional solidification is investigated. This investigation leads to the following points: (i) An ongoing rearrangement process of the two intermetallic rods by splitting and merging events is observed for constant growth velocities. (ii) Depending on the observed phase the same microstructure events take place at different rod sizes, despite similar phase fractions and number of rods. (iii) During a velocity decrease the coarsening of the microstructure is dominated by overgrowing events, whereas (iv) during a velocity increase the refinement is dominated by splitting events. (v) The events occur delayed to the velocity adjustment, caused by the spatial rearrangement of the material. (vi) However splitting events are also observed during the deceleration simulation and overgrowing events during the acceleration simulation. This indicates that the ongoing rearrangement processes are still continuing through the microstructure coarsening and refinement processes. (vii) For the same velocity at the end of the simulations, both microstructures show a similar behavior of the events. (viii) The results show that the used phase-field model is able to reproduce experimental phenomena of the microstructure rearrangement.

These investigations lead to a better understanding of the rearrangement and adjustment processes in the microstructures required for the development of tailored materials with locally defined microstructures.

Acknowledgments
We are grateful for the provided computational resources at the High Performance Computing Center Stuttgart and thank Martin Bauer from the Friedrich-Alexander-Universität Erlangen-Nürnberg for helping to implement the solver in the wALBERLA framework. The authors thank
Henrik Hierl, Christopher Seer and Marco Seiz for their support in the generation and analyses of the data. Further the authors thank for the financial support within the project SKAMPY (Ultra-scalable multiphysics simulations for solidification processes in metals) funded by BMBF and the cooperative graduate school Gefügeanalyse und Prozessbewertung by the ministry of Baden-Wuerttemberg.

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