Numerical study on the effect of convection   
on grain growth structure

Abstract

The effect of convection within the droplet during the levitation experiment on the grain structure was investigated using phase-field simulation considering convection. This study aims to distinguish the effects of convection and heterogeneous nucleation on the grain structure. In binary alloy systems, convection generates a strong solute gradient near the solid-liquid interface, leading to faster solid growth. The effect of flow velocity on the degree of grain deformation was investigated, and it was found that the shape of grown grains tends to deform with an increase in convection velocity. Small vortices are generated for the further large velocity, creating the local concentration gradient, thus resulting in the short-wavelength grain interface. In the solidification of pure metals based on heat transport, the effect of convection on grain shape was relatively small compared to the case of binary alloys because of the large thermal diffusion.

*Keywords: Additive manufacturing, Phase-field method, Solidification, Levitation methods, Microgravity;*

1. Background and purpose

In metal additive manufacturing (AM) techniques, adding heterogeneous nucleation site particles improves grain structures, resulting in the strength of manufactured objects. As shown in Fig. 1, adding heterogeneous nuclei refines and equiaxed the microstructure, improving anisotropy. Previous research has revealed that adding 0.2 vol% of heterogeneous nuclei TiC to the base material Ti-6Al-4V results in approximately double the Vickers hardness1).

Levitation experiments using the electrostatic levitation furnace (ELF) on the International Space Station (ISS) effectively investigate the refinement effects of adding heterogeneous nucleation sites during the melting and solidifying of metal samples under microgravity. Experiments using the ELF can suppress convection that influences grain structures, allowing a focused analysis of the effects of heterogeneous nucleation. However, the high cost of ISS missions limits the data obtained. As alternatives, ground-based levitation methods, such as electrostatic levitation (ESL), aerodynamic levitation (ADL), and electromagnetic levitation (EML), are available. This study is part of the Hetero-3D project, which aims to elucidate the mechanisms of microstructural refinement through these levitation experiments.

This study aims to understand how convection affects crystal grain structures and to determine the role of adding heterogeneous nucleation sites in grain refinement.

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| **Fig.1.** The diagram of the microstructure of a product manufactured by metal AM.  (a) no additive, and (b) additive heterogeneous nucleus. |

**Add substantial literature on the subject matter.**

1. Methods
   1. Multi-phase-field model

In this study, a multi-phase-field (MPF) model2) was used to simulate the evolution of microstructures in multiphase and polycrystalline materials. The phase field is assumed to satisfy the following equation in the MPF model,

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where denotes the grain numbers locally present. The free energy functional describing the movement of the interface and undercooling using phase field is constructed as follows:

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Here the double well potential , the gradient energy density can be described with the phase field

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Using the total free energy , the time evolution equation is formulated as follows:

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where is phase field mobility. Substituting Eq. (3) and (4) into Eq. (5), we obtain:

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whereand denote the energy barrier, and gradient coefficient, respectively. Usually, subscripts satisfy , here These parameters are determined using the physical properties of the materials. The last term in Eq. (5) represents the chemical driving force, which is defined as follows:

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In pure metal, the chemical driving force during the solidification process is defined by the temperature difference between the melting point and the undercooled liquid phase:

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where are free energy in solid and liquid phases, and latent heat, respectively. The free energy difference in binary alloys is calculated from a linearized phase diagram.

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where describes the entropy of fusion, and the gradient of phase lines, and the concentration, and the reference concentration, the reference temperature.

* 1. Implementation and calculation parameters

The Academic version of the OpenPhase, which includes a standard implementation of the MPF model discussed in Section 2.1, was used for simulations. These simulations focused on microstructure evolution driven by nucleation and growth processes based on Gibbs free energy. OpenPhase combines the MPF model with orientation and concentration fields and the convection model using the Lattice Boltzmann method. This allows detailed analysis of the interaction between melt flow and solidification in levitated droplets. The simulations considered pure titanium and the Ti-Al titanium alloy, both widely used in metal additive manufacturing.

Tables 1 and 2 show the calculation conditions for pure titanium and Ti-30at%Al, respectively. Fig. 2 shows the phase diagram of Ti-30at%Al used as the thermodynamic data. The phase diagram data was obtained from the materials database MatNavi CPDDB from NIMS and calculated using PyCALPHAD. The values of physical properties are cited from the literature4-8). As a comparative analysis between experimental samples and simulation results is planned for future works, the value of the interface anisotropy has been empirically determined.

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|  |  | **Table 3**. The phase diagram parameter of Ti-Al alloy. | | |
|  | Parameters | Value | Unit |
|  | Intersection concentration |  |  |
|  | Intersection temperature |  |  |
|  | Initial liquid concentration  of titanium (mole fraction) |  |  |
|  | Initial solid concentration  of titanium (mole fraction) |  |  |
|  | Initial liquid concentration  of aluminium (mole fraction) |  |  |
|  | Initial solid concentration  of aluminium (mole fraction) |  |  |
|  | Liquidus slope |  |  |
| **Fig. 2**. Phase diagram of Ti-Al alloy |  | Solidus slope |  |  |

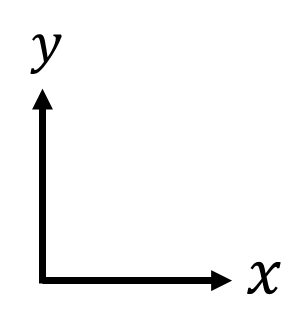
1. Results

Please, make reference to Tables 2 & 3. Also note that Tables 1, 2, & 3 are calculation parameters obtained from a source and do not constitute RESULT of your study. These Tables could better placed in Section 2.2.

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| **Table 1.** The material and calculation parameters of pure titanium. | | |  | **Table 2.** The material and calculation parameters of Ti-30at%Al. | | |
| Parameters | Value | unit |  | Parameters | Value | Unit |
| Mesh number |  |  |  | Mesh number |  |  |
| Grid spacing |  |  |  | Grid spacing |  |  |
| Interface width |  |  |  | Interface width |  |  |
| Equilibrium temperature |  |  |  | Initial temperature |  |  |
| Initial temperature |  |  |  | Cooling rate |  |  |
| Interface energy |  |  |  | Interface energy |  |  |
| Interface mobility |  |  |  | Interface mobility |  |  |
| Interface anisotropy |  |  |  | Interface anisotropy |  |  |
| Latent heat |  |  |  | Entropy of fusion |  |  |
| Thermal conductivity |  |  |  | Diffusion coefficient |  |  |
| Heat capacity |  |  |  | Diffusion coefficient |  |  |
| Kinetic viscosity |  |  |  | Kinetic viscosity |  |  |
| Prandtl number |  |  |  | Prandtl number |  |  |

* 1. Ti-Al alloy solidification simulation

A 2-dimensional domain representing part of a levitated droplet's cross-section was initialized with forced convection based on the findings of Usui et al3). as shown in Table 1.As shownin Fig. 3,all boundary conditions of the mm square 2D computational domain are periodic, and the flow under the conditions listed in Table 4 is set as the initial condition. Fig. 4 shows the result of the solidification simulation of Ti-30at%Al alloy. At the grain boundary positions perpendicular to the flow, the concentration gradient of enriched aluminum became steep, leading to faster grain growth toward the left side of the computational domain in Fig. 5(b). Fig. 5 shows the growth of multiple grains of Ti-30at%Al dendrites simulated in 2D. Based on previous research results, the number of nuclei is set to five and adjusted to the size of the current computational domain. Elliptical fitting was applied to the grains, and their shapes were evaluated by calculating the aspect ratio, defined as the ratio of the minor to the major axes. Fig.7 shows the aspect ratios of the five grains in the computational domain and their average for each levitation method condition. Fig. 7 shows that when convection at the magnitude of ESL or ADL is applied, the aspect ratio decreases, forming grains with anisotropy. The tendency of the aspect ratio can be explained by the gradient of the concentration field due to the mass transport by the convection. However, the aspect ratio increased again under a strong convection case corresponding to the EML. To discuss the grain morphology in detail, the distance from the center of the fitted ellipse to the interface is plotted in Fig. 8. The figure shows the morphology of the same grain ID, which was generated at the same location and has the same orientation under each convection condition. Although the long-wavelength curves look similar for all cases, the relatively short-wavelength components can be found in the case of strong convection.



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| **Table 4.** The magnitude of internal convective flow velocity in droplets during the levitation methods | | | | | |
|  | Levitation methods | ESL | ADL | EML |  |
| Velocity order [m/s] |  |  |  |

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| 1. No convection | 1. ESL condition | 1. ADL convection | 1. EML condition |
| **Fig. 5.** Multi-dendrite simulation of Ti-30at%Al with and without forced convection.  Each grain has a different grain ID and orientation field, distinguished by color. | | | |

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| **Fig. 3**. The computational domain. Forced convection in the x-axis direction is set as initial condition. | 1. No convection | 1. ESL condition |
| **Fig. 4.** Single dendrite simulation of Ti-30at%Al. | |



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| 1. ESL condition | 1. ADL condition | 1. EML condition |
| **Fig. 9**. Streamline around grains. Snapshots with similar grain sizes are compared. | | |

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| 1. No convection | 1. ESL condition |
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| 1. ADL condition | 1. EML condition | |
| **Fig 8**. Morphology of grain surface shape (.  The lines show the distance from the center of the fitted ellipse to the grain boundary interface. | | |

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| **Fig. 6.** The elliptical fitting.  This grain has in ADL condition. | **Fig. 7.** Aspect ratio of grains. The colors of the points correspond to the grain ID in Fig.6. The bars show their averages. |

Fig. 9 shows the streamlines around the grains in each convection condition. The generation of short-wavelength components at the interface is related to the vortices. As convection becomes stronger, the smaller vortices are generated, and their number also increases. The vortices play a role in enhancing the local concentration gradient near the interface. Once the velocity of the flow exceeds a certain threshold, stronger vortices create a shorter wavelength interface instead of reducing the aspect ratio.

* 1. Pure titanium solidification simulation

A 2D single dendrite simulation related to heat transfer was carried out considering pure titanium. The computational parameters for pure titanium are shown in Table 1. Fig. 10 shows the results of the solidification simulation for pure titanium under the four convection conditions listed in Table 4. Only the grains under EML conditions show a significantly different shape than those under other conditions. In Fig. 11, the case of EML shows the presence of twin vortices. The convection effect is smaller for heat-transfer-based solidification than in mass-transfer cases. The grain structures changed for the sufficiently strong flow, like the EML case.

Heat diffusion is faster than the convective heat transfer for the low-Prandtl number liquid. The effect of convection on the temperature gradient is smaller than in the case of concentration-based growth. In the pure substance cases, the relatively weak flow, up to the ADL case, does not affect the grain structures.

1. Conclusion

MPF simulations were conducted to investigate the effect of internal convection in levitation experiments on grain morphology. In the titanium binary alloy Ti-Al system, convection under ESL and ADL conditions reduced the aspect ratio of the grains. Large-scale convection in EML induces small vortices, altering local concentration gradients and forming grains with short wavelength interfaces. In the solidification of pure titanium based on thermal diffusion, the grain shape does not change very much except in the EML condition. The results of the present study showed that the droplet internal convection in the levitation methods may affect the final grain structures.

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| 1. No convection | 1. ESL condition | 1. ADL convection | 1. EML condition |
| Fig. 10. Single dendrite simulation of pure titanium. | | | |

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| ロゴ, 会社名  自動的に生成された説明 | | グラフ  低い精度で自動的に生成された説明 | | グラフ  低い精度で自動的に生成された説明 | | ロゴ が含まれている画像  自動的に生成された説明 |  |
| 1. No convection | 1. ESL condition | | 1. ADL condition | | 1. EML condition | |
| **Fig. 11.** Streamlines around grains. Snapshots with similar grain sizes are compared. | | | | | | |

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