Understanding microstructure evolution during additive manufacturing of metallic alloys using phase-field modeling

Yanzhou Ji[1], Lei Chen[2], Long-Qing Chen[1]
[1]Department of Materials Science and Engineering, The Pennsylvania State University
[2]Department of Mechanical Engineering, Mississippi State University

Additive manufacturing (AM), due to its flexibility and capability to deal with parts with complicated geometries, have attracted extensive research attention and is believed to be a promising candidate for accelerating the growth of advanced manufacturing industries. Benefiting from the recent development of manufacturing tools and integrated platforms, AM has been successfully extended to the manufacturing of metallic materials [1-13]. Many industries, such as biomedical and aerospace, are being poised to benefit from the metallic AM. Some examples include (1) on-site, rapid fabrication of metallic bone implants with patient and injury-specific designs, and (2) fabrication of replacement parts in remote locations (e.g. outer space). However, in a technical aspect, a more comprehensive understanding on the "processing-microstructure-properties" correlation is still lacking for the metallic AM. The metallic AM process involves non-uniform temperature distributions and rapid thermal cycles that result in microstructures featured with porosity and anisotropy, which differ drastically from their cast or wrought counterparts. Such different microstructure features critically affect the mechanical properties of the AM builds. Therefore, understanding the microstructure development and evolution during the AM process of metallic alloys is an important prerequisite for the optimization of the AM parameters to achieve desired mechanical properties of the AM builds.

1. Microstructures in additively manufactured metallic alloys

Microstructures are compositional and/or structural inhomogeneities developed during the processing of materials [14]. Microstructure evolution is a kinetic process to reduce the total free energies towards the thermodynamically equilibrium states in a material system under applied external fields [14]. Specifically, for AM of metallic alloys, the complex microstructure evolution characteristics arise from many aspects. On one hand, the multi-component, multi-phase nature of most of the commercial alloys enables the diverse microstructure patterns. The multiple alloying elements added to the alloy system, primarily for the purpose of improving the comprehensive mechanical properties or performances in practical applications, can cause substantial compositional inhomogeneities. Meanwhile, the multiple alloying elements and their inhomogeneous distributions within the system enable the formation of various thermodynamically stable and/or metastable phases. The various possible phase transformation kinetic pathways during the fabrication, treatment and processing of the alloy largely complicate the possible microstructure patterns and evolution paths.

On the other hand, the AM processing conditions further add to the complexity of the microstructures. To densify the initial metallic powders or wires into usable metallic parts, melting or partial melting (e.g., sintering) is required. AM techniques realize this process by a layer-by-layer processing fashion involving multiple rapid cycles. The marked temperature gradient subsequently induces the anisotropic growth behavior of certain microstructure features, resulting in the anisotropic mechanical properties of the builds. Meanwhile, the non-uniform temperature distribution and dynamic temperature

variation significantly complicate the phase transformation mechanisms and sequences in terms of both thermodynamics and kinetics principles. Microstructure features rarely reported in conventional manufacturing processes can be observed in the metallic AM. In addition, defects such as voids or pores may develop during the AM process, so as to augment the complexity of the microstructures.

1.1 Experimental observations

Tremendous efforts have been made in experimental characterization of the microstructure features in additively manufactured metallic alloys. Typically, optical microscope (OM), scanning electron microscope (SEM) and transmission electron microscope (TEM) are used for microstructure morphology characterization and electron backscatter diffraction (EBSD) is used for texture measurements. These experimental observations enable to outline the overall microstructure evolution processes during the metallic AM. The underlying mechanisms of microstructure evolution and their effects on the resulting mechanical properties of the AM builds can be hypothesized and verified as well. Especially, the AM of Ti-6Al-4V (in wt.%) has attracted extensive research interests [11,13,15-31], due to its excellent comprehensive mechanical properties, corrosion resistance and biomedical compatibility. Here ~~we~~ use an additively manufactured Ti-6Al-4V alloy as an example for the illustration purpose.

The AM of Ti-6Al-4V alloys has been accomplished by different AM techniques, and the resulting microstructures are prominently affected by the specific AM technique used. In terms of powder/wire supplies, powder-bed-fusion (PBF) AM techniques[15,17,18,25,30,32-34] and direct-energy-deposition (DED) techniques[13,16,20,29,31] are both reported for Ti-6Al-4V; in terms of power source, laser beam[11,15,20-22,24,29-35], electron beam[17-19,25,28,32,36] and arc[26,27] can be applied; in terms of processing methods, melting and sintering (followed by hot isostatic pressing (HIP)) are both reported. Nevertheless, since all of these different AM techniques and their combinations require melting or partial melting to densify the material, the thermal effect plays a critical role in the microstructure development during the AM building process. For illustration purpose, ~~we~~ describe the microstructure evolution during selective electron beam melting (SEBM) PBF of Ti-6Al-4V alloys, and briefly discuss the effect of AM techniques on microstructure evolution in Ti-6Al-4V.

For the building process of each layer in SEBM, micro-scale metal powders of Ti-6Al-4V are first uniformly spread. The electron beam heat source then follows the pre-set scanning path to melt the powders, forming a dynamic melting pool near the scanning probe. The shape of the melting pool depends on not only the specific materials system, but also AM parameters such as the heat source power, the shape of the scanning probe and the scanning speed.

1.1.1 Grain structures and textures

Fig. 1 illustrates the grain structure and texture development during AM of Ti-6Al-4V. β grains with body-centered-cubic (bcc) crystal structure develop near the trailing edge of the melting pool, as the melting pool moves, as shown in Fig. 1(a). The β grain growth direction is largely affected by the temperature gradient direction inside the melt pool [9]. Manufactured by specific AM parameters, the β grains may form different dendrite structures, in which columnar grains following the maximum temperature gradient directions are frequently seen (Fig. 1(c)); the specific morphology of the β grains depends on the interplay between the temperature gradient G and the interface velocity

(or cooling rate) R, as shown in Fig. 1(b). Solute segregation is also observed at the columnar grain boundaries, where the segregation of minor alloying elements may cause the formation of secondary particles. For the melting of the subsequent layer, the pre-existing layers may be partially re-melted; the development of grain microstructures may change if the scanning path changes.

The dynamic non-uniform temperature distribution in the build is one of the primary causes for the development of grain textures in different intersections of the build sample. For example, if the longitudinal section is longer than the wall section, and the scanning probe follows the same zig-zag path on each layer, then the grains in the longitudinal section are mostly columnar along the building direction [37]; whereas in the wall section, smaller grains appear near both walls while larger grains form and develop along the maximum thermal gradient inside the section [17]. These grain textures, as a result of the temperature gradient and cooling rates along the building direction, will cause anisotropies in mechanical properties of the build [6,11,23,36,37].

1.1.2 Solid state phase transformation

In addition to the grain morphology and textures, the temperature gradient and thermal history during AM will also affect the microstructure evolution inside β grains. When the temperature decreases below the β transus (about 1000°C for Ti-6Al-4V), the β→α allotropic phase transformation may take place, forming α products with hexagonal-close-packed (hcp) crystal structure. The specific β→α transformation modes may differ under different thermal conditions, which do not only contain the competition between diffusional and diffusionless transformation modes, but also, under a fix transformation mode, involve the interplay between the nucleation and growth of α products, leading to distinct microstructure features. For example, based on experimental observations during continuously cooling of Ti-6Al-4V, the β→α transformations may take place in different modes under different cooling rates [38-40]:

(1) under small cooling rates (~0.1K/s), the α phase primarily forms near β grain boundaries due to the higher undercooling and heterogeneous nucleation sites, and then develop coarse α colonies consisting of laths of the same α structural variants;

(2) under intermediate cooling rates (<20K/s), with increasing cooling rates, there is a decrease in the α-colony size and an increase in the intragranular α nucleation sites, resulting in basket-weave-type microstructures;

(3) under large cooling rates, partitionless β→α transformation, such as massive and martensitic transformation may take place, resulting in acicular α products inside prior β grains.

During AM processes, the situation becomes more intricate since multiple heating-cooling cycles are present, and cooling/heating rates at a selected region are generally not constant. As a result, different α products may sequentially develop and dissolve during thermal cycles, resulting in complicated (α+β) two phase mixtures [13]. The size, morphology and textures of α laths and colonies can significantly influence the mechanical properties of the sample[40]. Besides, due to the compositional inhomogeneities originated from solute segregations at either grain boundaries or α/β interface boundaries, minor precipitates or inclusions may also be present inside prior β grains (such as Ti_3Al) or at grain boundaries, which may have different effects on the mechanical properties of the build. Fig. 1(d) shows the typical (α+β) basket-weave microstructures in the AM build of Ti-6Al-4V.

1.1.3 Effect of different AM techniques

As mentioned above, the microstructure morphology may be affected by the different AM techniques. For example, as reviewed in [11], the as-fabricated Ti-6Al-4V by PBF laser melting is generally finer than that by DED laser melting techniques. In as-fabricated Ti-6Al-4V by PBF laser melting, the grain size is generally smaller and the acicular α' martensites are frequently observed, in contrast to the fully laminar α plates in DED-fabricated samples. The primary cause for the microstructure difference is identified to be the different laser spot size for the two techniques. The smaller laser spot size in PBF AM technique leads to smaller melt pools and larger temperature gradient, which result in finer microstructures.

The type of heat source can also affect the microstructure morphology. For example, Ti-6Al-4V builds fabricated by SEBM contain fewer α' martensites (only at top surfaces), in contrast to that fabricated by SLM[2,32]. The microstructure difference can be attributed to the faster moving velocity and the capability for *in situ* heat treatment of SEBM techniques[2].

In addition, the densification methods also influence the final microstructures. For example, comparing SLM with selective laser sintering (SLS), the SLS process generally has lower laser energy input and/or faster scanning speed since the metal powders are only partially melted. The melt pool is generally smaller in SLS and the densification of metal powders is mainly driven by surface tension rather than melting. Since thermal history is also present in SLS, the anisotropic microstructure features, such as columnar grains, are also observed in SLS-fabricated Ti-6Al-4V[41].

Based on these understandings, the microstructure evolution mechanisms can be hypothesized, and the AM parameters that lead to the change of microstructures can be identified. To further understand the processing parameter-microstructure-property relationship, systematic high-throughput experiments should be conducted in the entire AM processing parameter space, which is not only financially expensive but also time-consuming. Meanwhile, due to the difficulties in *in situ* observation of the microstructure evolution process during AM, the direct evidences and details of the microstructure evolution kinetics are usually lost.

1.2 Computational Simulations

Recently, with the development of computation tools and numerical methods, the computational simulation has become a promising alternative to the experimental investigations of AM, in a cost-effective manner. However, AM is a complicated process involving the interactions among different applied external fields, resulting in complicated microstructure features. To accurately predict and reconstruct the microstructure evolution process during AM, the realistic AM processing parameters, materials parameters and geometries should be input into the computation models. Moreover, careful validations through quantitative comparison with experimental results should be performed to ensure the robustness of the model. There have been a series of existing attempts on computational simulations of the microstructure evolution during AM.

Since the prominent feature of metal AM is the complicated thermal effects, the initial computational efforts in AM of alloys largely ignored the microstructure aspects. Instead, macroscale prediction of temperature distribution and history as a function of AM parameters such as power, spot size, scanning speed and scanning directions of the

heat source by solving the heat equations [28,42-48] were the main computational focus. These simulations can further couple with finite-element-based mechanical models to predict the macroscopic mechanical properties of the AM build. Notably, Michaleris et al.[28,44,45,48] developed a finite-element-based software for thermo-mechanical modeling of AM which can provide accurate solutions with lower mesh density and higher computation efficiency. Although not directly predict the microstructure evolution, these efforts can provide indications for the geometries of the melt pool, the temperature distributions and thermal history in the build, which lay the foundation for further microstructure predictions.

By coupling with thermal calculations, there have been a few existing efforts to develop computational models for predicting the grain morphology during AM of alloys. For example, Nie et al.[49] used a microscopic stochastic analysis including temperature-dependent nucleation rate, solute diffusion and growth anisotropy, to simulate the dendrite morphology evolution as a function of cooling rate and temperature gradient in IN718. Similar ideas have been applied by Zhou et al.[50] for AM of a stainless steel using Cellular Automata. By assuming that solidification direction is parallel to the local maximum heat flow direction, and the columnar to equiaxed transition occurs beyond a critical G/R (temperature gradient/cooling rate) ratio, Wei et al.[51] applied a simple two-dimensional (2D) grain growth model to predict and validate the grain orientation development during AM of an Al-alloy.

Moreover, the phase-field approach[14,52-61], which uses the diffuse-interface description to avoid the explicit tracking of interfaces, shows the great potential to simulate microstructure evolution during AM of metallic alloys. There have been some initial attempts to employ the phase-field method for modeling microstructure evolution during AM. For example, Gong et al.[32,62] coupled the thermal process modeling with a phase-field model to simulate the dendrite morphology during AM of Ti-6Al-4V, while considering undercooling effect. Lim et al.[63] proposed a rather preliminary modeling framework using both the phase-field approach and a crystal plasticity finite element (CP-FE) method which could be applicable to microstructure evolution modeling in AM.

However, to date, due to the complexity of the AM process, microstructure evolution modeling of alloys during AM is still at its early stages. Existing investigations mainly focus on the modeling of certain microstructural aspects. The model validations and the construction of an integrated model containing major microstructure features of AM are still lacking. In addition, the temperature distribution and thermal history during the building process may affect the microstructure features at different length scales, ranging from the grain structures developed during solidification with a typical length scale of several microns, to the intra-granular microstructures such as precipitations, micro-segregations and defects at nanoscale. Therefore, it is currently numerically challenging and computationally expensive to simulate the cross-length scale microstructure features during AM.

2. Multi-scale phase-field model for AM of alloys

In this work, ~~we~~ present a multi-scale computational framework based on the phase-field approach to simulate the microstructure evolution during AM of alloys. In particular, ~~we~~ consider a SLM or SEBM process. To highlight the major governing factors of the

microstructure evolution and simplify the numerical model, ~~we~~ make the following overall assumptions:

(1) The microstructure evolution processes have negligible effect on heat transfer and temperature distribution in the build, while the heat transfer is mainly affected by the heat conductivity and capacity of the material, as well as the AM parameters such as power, scanning probe size and scanning speed of the heat source;

(2) The solidification and the development of grain structures take place in the high temperature regime followed by possible solute segregation and inclusions occur near grain boundaries, while the phase transformation and microstructure evolution inside grains take place in the low temperature regime with negligible grain structure change.

With the assumptions above, the entire microstructure evolution model can be decoupled into three different sub-models on different length scales, as illustrated in Fig. 2: (i) the macroscopic thermal model to obtain the temperature distribution and thermal history in the build sample during the whole AM process;

(ii) the grain-scale solidification or grain growth phase-field model to study the grain morphology and texture development and/or solute distribution and segregation;

(iii) the sub-grain-scale phase-field model to simulate the intra-granular phase transformations which may include diffusional transformations such as precipitation and diffusionless structural transformations, depending on the specific materials system.

2.1 Linkage between the three sub-models

Rather than performing the three sets of simulations independently, the linkage among the sub-models is considered to reflect the AM processing conditions. The results of the larger-scale simulations will be used as the input parameters or initial microstructures for the smaller-scale simulations. Specifically, the thermal model focuses on the effect of various AM parameters, such as the power, shape and scanning speed of the heat source, the layer thickness and the scanning paths on the temperature distribution and thermal history of the build. The calculated temperature distribution and thermal history will be input into the grain growth or solidification phase-field model. For simulating the intra-granular microstructure evolution, several representative regions will be selected from the simulated grain structures; the grain structures and solute compositions of the selected region will be enlarged through interpolation methods to obtain better simulation resolutions for the sub-grain microstructures. Since the length scale of the selected region is comparable with the resolution of the thermal model, within the selected region, the temperature distribution can be assumed uniform while change with time during the AM process. Therefore, thermal history for the selected region will be input into the sub-grain-scale phase-field model to simulate the microstructure evolution. With this multi-scale model, the temperature effect during the AM process on the microstructure evolution can be thoroughly studied. With proper model validation, the effect of the AM parameters on the microstructure evolution of the additively manufactured alloys can also be understood.

2.2 Finite-element thermal model

Next, ~~we~~ present the details of the multi-scale microstructure model for AM of metallic alloys, using Ti-6Al-4V as an example. ~~We~~ start from the finite-element-based thermal calculations, using a Life-death Element Technique. Both substrate and printing component are fully discretized into finite elements, but the elements of printing component are inactive before printing. At the beginning of each simulation time step,

which corresponds to a real time period during AM, a new group of finite elements is activated to mimic the movement of the heat source, and those active elements that constitute a complete domain are calculated for thermal analysis.

Based on the ABAQUS software platform[64], the temperature field of both the printing component and the substrate can be calculated by solving the 3D heat conduction equation

$$\frac{\partial}{\partial x}\left(k_x\frac{\partial T}{\partial x}\right)+\frac{\partial}{\partial y}\left(k_y\frac{\partial T}{\partial y}\right)+\frac{\partial}{\partial z}\left(k_z\frac{\partial T}{\partial z}\right)+\rho Q=\rho c_p\frac{\partial T}{\partial t} \tag{1}$$

where k_x, k_y and k_z are thermal conductivities along the three coordinate axes, respectively. ρ (kg/m^3) denotes the density, c_p (J/(kg·K)) denotes specific heat capacity, Q (W/kg) denotes heat source density.

At each simulation step, the convection and radiation boundary conditions are updated and applied to outer surfaces of the active elements

$$k_x\frac{\partial T}{\partial x}n_x+k_y\frac{\partial T}{\partial y}n_y+k_z\frac{\partial T}{\partial z}n_z=h\left(T_a-T\right)+\varepsilon_R\sigma_R\left(T_a^4-T^4\right) \tag{2}$$

where h (W/m^2·K) is heat convection coefficient, T_a is ambient temperature, ε_R, σ_R are emissivity and Stefan-Boltzmann constant (5.67×10^{-8} W/m^2·K^4), respectively.

The electron beam heat source is regarded as a body heat flux, which is modeled by Goldak double-ellipsoid model[28]

$$Q=\frac{6\sqrt{3}UI\eta f_s}{a_1b_1c_1\pi\sqrt{\pi}}e^{-\left[\frac{3x^2}{a_1^2}+\frac{3\left(y+v_Qt\right)^2}{b_1^2}+\frac{3z^2}{c_1^2}\right]} \tag{3}$$

where U represents the electron beam acceleration voltage, I is the electron beam current, η represents the absorption efficiency, f_s represents the process scaling factor, x, y and z are the local coordinates of the heat source, a_1, b_1 and c_1 are the transverse, melt pool depth, and longitudinal dimensions of the ellipsoid, respectively, v_Q represents scanning speed of heat source, and t is the scanning time. The resulting temperature distribution within the build during the AM process and the thermal history in a selected representative volume element (RVE) are shown in Fig. 3.

2.3 Grain-scale phase-field model: grain growth & solidification

2.3.1 Model description

With the temperature distribution and history at hand, ~~we~~ then study the β grain growth behavior during AM using a grain-scale phase-field model. In this model, the texture (crystallographic orientation of each grain) in a simulation cell is specified by a set of continuous order parameters. The total free energy of a polycrystalline microstructure system can be described as follows[65,66]

$$F=\int\left[f_0\left(\phi_1,\phi_2,...,\phi_Q\right)+\sum_{q=1}^{Q}\frac{\kappa_q}{2}\left(\nabla\phi_q\right)^2\right]d\mathbf{r} \tag{4}$$

where $\{\kappa_q\}$ are positive gradient energy coefficients, and $f_0(\{\varphi_q\})$ is the local free energy density, which is defined as

$$f_0\left(\{\phi_q\}\right)=-\frac{\alpha}{2}\sum_{q=1}^{Q}\phi_q^2+\frac{\beta}{4}\left(\sum_{q=1}^{Q}\phi_q^2\right)^2+\left(\gamma-\frac{\beta}{2}\right)\sum_{q=1}^{Q}\sum_{s>q}^{Q}\phi_q^2\phi_s^2 \tag{5}$$

in which, α, β and γ are constants, for $\alpha=\beta>0$ and $\gamma>\beta/2$, f_0 possesses $2Q$ degenerate minima. Those minima are located at $(\varphi_1, \varphi_2, \ldots, \varphi_Q)= (\pm1, 0, \ldots, 0), (0, \pm1, \ldots, 0), \ldots, (0, 0, \ldots, \pm1)$, representing the finite number of possible of grain orientations in a polycrystal.

Grain growth is described by the temporal and spatial evolution of the order parameters, which yield the time-dependent Ginzburg-Landau equations

$$\frac{\partial \phi_q(\mathbf{r},t)}{\partial t} = -L_q \frac{\delta F}{\delta \phi_q(\mathbf{r},t)} \quad (q=1,2,...,Q) \tag{6}$$

where $\{L_q\}$ are kinetic rate coefficients influenced by temperature gradient, which can be calculated through the modified Arrhenius type equation such as

$$L_q = A \cdot \left(\frac{T}{T_0}\right)^m \cdot \exp\left(-\frac{E_a}{RT}\right) \tag{7}$$

where R, molar gas constant, is 8.314J/(mol·K) and m is a constant lying in the range -1<m<1, T_0 is the ambient temperature (293.15 K), A is a temperature-independent constant, E_a is the activation energy for interface movement. The equation is solved numerically using finite difference method.

To link the grain-growth phase-field model with the FEM temperature calculations and simulate the grain growth behavior during the layer-by-layer AM process, the following assumptions are made:

1) When the temperature is above the liquidus temperature (1660℃), it is set as liquidus temperature;
2) When the temperature falls below the β transus temperature ($\beta \rightarrow \alpha$, 1000 ℃), the kinetic rate coefficients $\{L_q\}$ are set to be zero.
3) It is assumed the temperature in the newly added layer is uniform, and the process of applying the uniform temperature is instantaneous;
4) ~~We~~ assume that the temperature gradient is fixed after the heat source left, and the direction of maximum heat flux always keeps vertical upward;

With these assumptions, ~~we~~ investigate the grain growth behavior during AM in the longitudinal and wall sections of the build, due to their distinct temperature distributions. ~~We~~ select one representative region in each section for the grain growth simulations, as shown in Fig. 4. To obtain simulated grain morphology with acceptable resolution, the required mesh size for the phase-field simulations (~μm) should be much less than that of the thermal calculations (~10μm). Therefore, interpolation method is used to fit the temperature distribution data from the thermal calculations, as shown in Fig. 4, for both longitudinal and wall sections.

2.3.2 Simulation results

With the temperature distribution and history available from the thermal calculations, 2D/3D phase-field grain growth simulations are performed. With randomly distributed grains as the initial state, new layers with adjustable thickness are introduced into the system at adjustable time intervals to mimic the AM process. The simulated grain microstructures can well reproduce the experimentally observed ones in both the longitudinal section[37] and the wall section[17], as shown in Fig. 5, which confirms the effect of temperature distribution during AM on the grain texture development. In particular, in the longitudinal section, since the temperature gradient is along the building direction and perpendicular to the layers, columnar grains parallel to the building direction are observed. In the wall section, since the temperature gradient is not uniform

as shown in Fig. 4, grains grow along the local temperature gradient direction to form the columnar grains in the middle and small grains near the edges of the section.

The development of columnar grains during AM may lead to the anisotropies in mechanical behaviors of the build, which is detrimental in the practical applications. Therefore, ~~we~~ further investigate the effect of the AM processing parameters, especially the scanning speed and the layer thickness, on the grain morphology evolution. Based on ~~our~~ simulations, as briefly illustrated in Fig. 6, coarse columnar grains develop as the layer thickness and/or the scanning speed decrease, since the larger grains below the newly added layers have enough time to swallow the smaller grains or select the preferred smaller grains inside the newly added layers, which should be avoided during real AM process.

2.3.3 Future directions

The current phase-field model only focuses on the grain morphology development and ignores the solute distribution during the solidification process. Moreover, even in the longitudinal section, the temperature distribution is not uniform, especially in the melt pool where the solidification and grain growth begins. To more accurately simulate the microstructure evolution in the melt pool and during the movement of the melt pool, the phase-field solidification model by Gong et al.[32,62] should be extended to polycrystal systems and consider the realistic geometry of the melt pool, as illustrated in the preliminary simulation results in Fig. 7. In addition, as has been experimentally reported, the change of scanning directions of the heat source during AM will also change the grain texture in the longitudinal section; the growth direction of the grains will not exactly follow the maximum temperature gradient direction[67]. These phenomena will be considered in ~~our~~ future phase-field grain growth/solidification models for AM processes.

2.4 Sub-grain-scale phase-field model: solid-state phase transformations

2.4.1 Model description

With the simulated β grain structures, ~~we~~ further simulate the sub-grain-scale microstructure evolutions in additively manufactured Ti-6Al-4V alloys. Ignoring the minor inclusions or defects, the β→α transformation is the major factor for the development of the intra-granular microstructure features. The β→α transformation happens when the temperature of a local region in the sample is below the β transus temperature (~1000°C) during AM, or during the follow-up thermo-mechanical processing of the as-build sample. The microstructure features during β→α transformation is not only affected by the temperature distributions and history, but also influenced by the solute distribution and grain structures. Therefore, ~~we~~ select different RVEs in the thermal calculation to obtain the local temperature history. The temperature distribution within each RVE is assumed uniform. ~~We~~ further select the grain structures of the corresponding RVE in the grain-scale phase-field simulations as the initial grain structure. Interpolation method is used to convert the grain structures in the grain growth simulations with larger grid size (~μm) into the sub-grain phase-field simulations with smaller grid size (~10nm). Since the β→α transformation may contain both compositional and structural changes, both solute compositions (X_{Al} and X_V) and structure order parameters $\{\eta_{p,g}\}$ are considered to describe the microstructures. Specifically, the Burgers orientation relation for the bcc→hcp transformation is considered, resulting in 12 symmetry-allowed α variants in each β grain[68]. The free energy functional of the system is[69-73]

$$F = \int_V (f_{bulk}\left(X_{Al}, X_V, \{\eta_{p,g}\}, T\right) + f_{grad}\left(\{\nabla \eta_{p,g}\}\right) + e_{el})dV \tag{8}$$

where f_{bulk} is the temperature-dependent bulk free energy density, f_{grad} is the energy contribution due to the inhomogeneity of order parameters, and e_{el} is the elastic strain energy density.

The bulk free energy density includes the molar Gibbs free energy of all the phases in the systems as well as energy variations due to structure change. Although many stable and metastable phases have been identified in Ti-6Al-4V, ~~we~~ primarily focus on the (α+β) two phase system. Therefore, the following bulk free energy density is used:

$$\begin{aligned} f_{bulk}(X_{Al}, X_V, \{\eta_p\}, T) &= V_m^{-1} \cdot \sum_p h(\eta_p) \cdot f^{\alpha}(X_{Al}^{\alpha}, X_V^{\alpha}, T) \\ &+ V_m^{-1} \cdot \left(1 - \sum_p h(\eta_p)\right) \cdot f^{\beta}(X_{Al}^{\beta}, X_V^{\beta}, T) + g\left(\{\eta_p\}\right) \end{aligned} \tag{9}$$

where f^{α} and f^{β} are molar Gibbs free energies of α and β phases (in J/mol), respectively, which are directly taken from the Ti-Al-V thermodynamic database[74]; V_m is the molar volume, $h(\eta) = 3\eta^2 - 2\eta^3$ is an interpolation function, and $g\left(\{\eta_p\}\right)$ is a Landau-type double-well potential describing the energy variation due to structure change:

$$g\left(\{\eta_p\}\right) = w \cdot \sum_p \eta_p^2 (1 - \eta_p^2) + w' \cdot \sum_{p \neq q} \eta_p^2 \eta_q^2 \tag{10}$$

w and *w'* are parameters characterize the barrier height.

The gradient energy term in Eq. (8) is the part of interfacial energy due to the inhomogeneous distribution of order parameters at interfaces. ~~We~~ assume the gradient energy coefficient is in the matrix form and express the gradient energy as:

$$f_{grad} = \frac{1}{2} \sum_{p=1}^{12} \kappa_{p,ij} \left(\nabla_i \eta_p\right)\left(\nabla_j \eta_p\right) \tag{11}$$

where $\kappa_{p.ij}$ is the anisotropic gradient energy coefficient tensor for variant *p*, which, together with the barrier height *w*, can be obtained from α/β interfacial energy[75] and interface thickness values through thin-interface analysis[76].

The elastic strain energy originates from the mismatch between the crystal structures of both phases. Based on the lattice constants and the lattice correspondence between the crystal structures of the two phases, the stress-free transformation strain (SFTS) can be derived in the local reference coordinates under finite-strain approximation:

$$\varepsilon_{local}^{00}=\begin{pmatrix}\frac{2a_\alpha^2}{3a_\beta^2}-\frac{1}{2} & \frac{a_\alpha^2}{6\sqrt{2}a_\beta^2} & 0\\ \frac{a_\alpha^2}{6\sqrt{2}a_\beta^2} & \frac{7a_\alpha^2}{12a_\beta^2}-\frac{1}{2} & 0\\ 0 & 0 & \frac{1}{4}\left(\frac{c_\alpha^2}{a_\beta^2}-2\right)\end{pmatrix} \tag{12}$$

where a_α, c_α and a_β are the lattice constants of α and β phases, respectively[77]. The SFTS of each α variant $\varepsilon_{global,ij}^{00}(p)$ can be calculated by axis transformation from the local reference frame to the global reference frame. The elastic strain energy can then be calculated as[78]:

$$e_{el}=\frac{1}{2}C_{ijkl}\cdot\left(\varepsilon_{ij}-\varepsilon_{ij}^0\right)\cdot\left(\varepsilon_{kl}-\varepsilon_{kl}^0\right) \tag{13}$$

where C_{ijkl} is the elastic stiffness tensor, ε_{ij} is the total strain solved from the stress equilibrium equation $\sigma_{ij,j}=0$ under certain mechanical boundary conditions, ε_{ij}^0 is the overall eigenstrain:

$$\varepsilon_{ij}^0=\sum_p h(\eta_p)\cdot\varepsilon_{global,ij}^{00}(p) \tag{14}$$

The microstructure evolution is governed by the numerical solution of both the Cahn-Hilliard equation (for solute compositions)

$$\frac{\partial X_{Al}}{\partial t}=\nabla\cdot\left(\sum_p h(\eta_p)\cdot\left(\tilde{D}_{AlAl}^{\alpha}\nabla X_{Al}^{\alpha}+\tilde{D}_{AlV}^{\alpha}\nabla X_V^{\alpha}\right)+\left(1-\sum_p h(\eta_p)\right)\cdot\left(\tilde{D}_{AlAl}^{\beta}\nabla X_{Al}^{\beta}+\tilde{D}_{AlV}^{\beta}\nabla X_V^{\beta}\right)\right) \tag{15a}$$

$$\frac{\partial X_V}{\partial t}=\nabla\cdot\left(\sum_p h(\eta_p)\cdot\left(\tilde{D}_{VAl}^{\alpha}\nabla X_{Al}^{\alpha}+\tilde{D}_{VV}^{\alpha}\nabla X_V^{\alpha}\right)+\left(1-\sum_p h(\eta_p)\right)\cdot\left(\tilde{D}_{VAl}^{\beta}\nabla X_{Al}^{\beta}+\tilde{D}_{VV}^{\beta}\nabla X_V^{\beta}\right)\right) \tag{15b}$$

where $\tilde{D}_{ij}^{\phi}(i,j=Al,V;\phi=\alpha,\beta)$ are the inter-diffusivity coefficients available from literature[79,80]; and Allen-Cahn equation (for structure order parameters):

$$\frac{\partial\eta_p}{\partial t}=-L\left(\frac{\partial f_{bulk}}{\partial\eta_p}-\kappa_{p,ij}\nabla_i\nabla_j\eta_p+\frac{\partial e_{el}}{\partial\eta_p}\right),\ p=1,2,...,12 \tag{16}$$

where L is a kinetic coefficient related to interface mobility, which can be evaluated according to thin-interface analysis if the interface mobility values are available[76]. To improve the computation efficiency, the governing kinetic equations, as well as the stress equilibrium equation are solved using fast Fourier transform (FFT) algorithm. The stress equilibrium is assumed to be much faster than the microstructure evolution.

Due to the anisotropic interfacial energy and SFTS, the morphology of α products, both diffusional and diffusionless ones, are generally anisotropic with lath or acicular shape and habit planes[52,81-85]. With these anisotropies considered, ~~we~~ reconstruct the morphologies of α product, which quantitatively agrees with the experimentally reported habit planes[52,81-85], as shown in Fig. 8. With the energy anisotropies and the temperature-

dependent thermodynamic and kinetic coefficients, the non-isothermal growth behavior of α products can be accounted for. To further consider the temperature-dependent nucleation behavior, ~~we~~ apply the classical nucleation theory[59,86,87]:

$$j = ZN_0\beta^* \exp\left(-\frac{\Delta G^*}{RT}\right)\exp\left(\frac{t}{\tau}\right) \quad (17)$$

where j is the nucleation rate, Z is Zeldovich's factor, N_0 is the number of available nucleation sites in the corresponding system (here a simulation cell), β^* is atomic attachment rate, ΔG^* is nucleation barrier, t is elapsed time and τ is incubation time for nucleation. The detailed formulation of these quantities can be found in[88]. For simplicity, ~~we~~ assume the initial α nuclei is of spherical shape. The diffusionless β→α transformation is assumed to be much faster than the diffusional one. Therefore, the new α nuclei put into the system are assumed to only include order parameter change without composition change.

2.4.2 Simulation results

With the temperature-dependent description of both the nucleation and growth behavior, phase-field simulations are performed in β single crystals under different cooling rates (1K/s, 10K/s, 100K/s). The simulation results shown in Fig. 9 can capture the increased α number density and decreased α size with increasing cooling rates, which qualitatively agrees with the experimental observations[40]. Based on the phase-field simulations, the microstructure evolution during β→α transformations under different cooling rates is governed by the competition between nucleation and growth. Under high cooling rates (100K/s), there is an enhanced nucleation rate due to the high undercooling, which results in larger nucleation rate; on the other hand, the growth of the α nuclei is largely retarded, due to the decreased growth rates and increased competitions among different α nuclei (since the increased nucleation rate would lead to significant decrease in average spacing of α nuclei) at low temperatures. Under low cooling rates (1K/s), the system can stay at the high-temperature regime for a sufficiently long time, so that the α nuclei can grow larger, which, on the other hand, retards the further nucleation of α products.

For a specific RVE in the macroscopic thermal model, its thermal history during AM involves multiple cooling/heating cycles with non-constant cooling/heating rates. Based on the thermal history calculations, the cooling/heating rates increase significantly when the RVE is near the heat source, and the overall effect for the multiple thermal cycles is a cooling process, as shown in Fig. 3(b). Therefore, in a RVE of the thermal model, below the β transus, after each cooling/heating cycle, the α products will form and dissolve, while the overall effect is the formation of certain amount of α products without dissolution. The α products left over after a cooling/heating cycle are generally the ones formed at the earlier stages during the precious cooling process, which will also affect the formation of subsequent α products. Therefore, it is critical to understand the formation and growth sequence of α products during cooling, especially in polycrystals. As mentioned in[3,89], during early stages of cooling, the driving force for nucleating an α nucleus is limited, resulting in low nucleation rate in grain interiors. In this case, grain boundaries and/or pre-existing dislocations can facilitate the nucleation of α particles through heterogeneous nucleation, resulting in grain boundary α (GB-α) products as the initially observed α products, especially during low cooling rates. The GB-α products are

usually of plate shapes parallel to the GB and form protrusions which develop into α-colonies consisting of the same α variants parallel to each other. The formation of α-colonies is either due to the instabilities during the growth of GB-α or the elastic interactions between the α nuclei and pre-existing GB-α plate, as has been discussed in [90]. During the growth of α-colonies, new α nuclei can form inside β grains to form basket-weave-type microstructures. To illustrate this process, we simulate the microstructure evolution during slow cooling in a Ti-6Al-4V polycrystal, as shown in Fig. 10, which captures the sequential microstructure evolution features. Based on the current simulation results, further simulations will be performed in Ti-6Al-4V polycrystals by applying the predicted thermal history during AM from the macroscopic thermal model and the initial grain structure from the grain growth model. Furthermore, the effect of AM parameters on the microstructure development will be investigated.

2.4.3 Future directions

The sub-grain microstructure evolution varies with the specific material systems. The current sub-grain phase-field model focuses on the β→α transformations, which is the major cause for microstructure evolution during AM of Ti-6Al-4V. For other materials systems, the phase transformations in sub-grain scale can become more complicated. For example, during AM of superalloy IN718, different types of precipitates can appear at GBs and/or inside grains, including the Laves phase, δ-Ni_3Nb, γ'-Ni_3Al and γ''-Ni_3Nb precipitates, which have different effects on the mechanical properties of the build. Specifically, the intra-granular γ' and γ'' precipitates, either sequentially precipitated or co-precipitated, can remain coherency with the γ-matrix and provide notable precipitate hardening[91]; the GB-δ can partially enhance the mechanical property of the build by impeding grain growth, while the intragranular δ, which has limited coherency with γ-matrix, is detrimental to the overall strength of the build[91]. To simulate the microstructure evolution of these precipitate phases, the sub-grain phase-field model should be further extended to multi-component, multi-phase systems coupled with corresponding temperature-dependent thermodynamic database and diffusion kinetic information.

3. Summary and outlook

In summary, the proposed multi-scale phase-field modeling framework is capable of capturing the major grain-scale and sub-grain scale microstructure features during the AM of Ti-6Al-4V. The effect of temperature distribution and thermal history on the microstructure evolution mechanisms can be investigated, which can be further correlated with the macroscopic AM processing parameters. As a preliminary framework, ~~we~~ also show the possibility and capability of constructing such an integrated computation framework for further understanding and predicting the microstructure evolution during the realistic AM process. The correlation between AM processing parameters and the resulting microstructures during the metallic AM can also be investigated. More accurate and reliable predictions of the microstructure evolution would rely on the improvement, extension and validation of the current model. Especially,

(1) The numerical accuracy, robustness and efficiency should be further improved. This does not only include the numerical improvement for the three sub-models individually: macroscopic thermal model, grain-scale grain growth/solidification phase-field model and sub-grain scale phase-field model for solid state phase transformation;

but also include the improvement for the connect-interface among the three models, so that the key information related to the AM process from the larger-scale model can be input into the smaller-scale model.

(2) The quantitative validation of the predicted microstructure morphology during AM should be performed with existing experimental results. This can be accomplished by performing parallel AM experiments with the same material and AM processing parameters as that in the phase-field model, and characterize the as-build sample using different techniques, such as EBSD for grain texture and variant distribution and SEM/TEM for microstructure morphology analysis. By comparing the simulation results and experimental observations, new insight can be obtained for better improving the simulation methods, numerical treatments and model parameters.

(3) The accuracy of phase-field simulations is also related to the accuracy of the input materials parameters, as well as the temperature-dependent thermodynamic and diffusion mobility databases of the material system. These largely rely on more accurate experimental calibrations. For the database development, more experiments on phase boundary identification and inter-diffusion measurement are desired. First-principles calculations on the relative phase stabilities, self-diffusivities and impurity diffusivities can also be an important contribution.

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Figure Captions

Fig. 1: Illustration of the effect of processing on texture and grains in AM, adapted from[9] (Credit: P. C. Collins et al.)

Fig. 2: Illustration of the multi-scale phase-field framework for AM of alloys: (a) finite-element-based thermal model; (b) grain growth phase-field model; (c) sub-grain-scale phase-field model for solid phase transformation.

Fig. 3: Calculation results of the thermal model: (a) temperature distribution (in °C) during AM; (b) temperature history of selected RVEs (nodes).

Fig. 4: Connection between the thermal model and the grain growth phase-field model: representative longitudinal section and wall section for grain growth simulation, as well as the interpolated temperature distribution data for phase-field simulations in the two sections.

Fig. 5: Comparison between the phase-field simulated grain morphology and experimental observations. (a1) Experimental results in the longitudinal section[37] (credit: A. A. Antonysamy); (a2) simulation results in the longitudinal section; (b1) experimental results in the wall section[17] (credit: A. A. Antonysamy et al.); (b2) simulation results in the wall section.

Fig. 6: Effect of AM parameters on grain morphology: (a) layer thickness; (b) scanning speed.

Fig. 7: Preliminary phase-field simulation results for dendrite morphology in a moving melt pool. (a) Initial grain configuration; (b) dendrite morphology; (c) solute distribution. The red rectangle represents the heat source and the arrow represents the scanning direction.

Fig. 8: Morphology of a single α variant in β matrix with the consideration of the anisotropic energy contributions.

Fig. 9: Phase-field simulation of β→α transformations under different cooling rates in β single crystal and the comparison with experimental observations: (a) phase-field simulations, system size 10μm×10μm×10μm; (b) experimental observations[40] (Credit: G. Lütjering).

Fig. 10: Sequential formation of α products during slow cooling of a polycrystal: (a) initial grain structure; (b) formation of GB-α; (c) development of α-colonies; (d) basket-weave+colony microstructure. System size: 10μm×10μm×10μm.