A coupled diffusion-deformational phase field formulation for precipitate growth during laser-aided additive manufacturing process

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Abstract. A coupled stress-diffusion model for diffusion-controlled growth of precipitates in the powder-based laser additive manufacturing (AM) of metallic parts is presented using phase field approximation. The model is developed in a thermodynamically consistent way, with free-energy expressions associated with microstructural processes, the postulation of energy and mass balances and demonstration that the classical entropy inequality (Clausius-Duhem inequality) is satisfied. The governing equations that follow from the balance laws involve the phase variable, the atomic displacement field, the plastic hardening variables, and the solute concentration field, with significant couplings between all equations. The model includes concentration dependent eigen strain, strain due to phase transformation, strain dependency on precipitate growth and plastic hardening effects. To describe the effects of temperature and fluid velocity distributions and thermal history on the precipitate growth mechanisms a linking of microscale model with the macroscopic AM processing conditions is discussed.

1. Introduction
Laser metal deposition (LMD) with powder injection is one of the promising laser additive manufacturing (AM) technology based on adding powder materials layer by layer for making 3D parts directly from computer-aided design data. The applications of LMD include coatings, rapid prototyping and tooling, and part refurbishment. The mechanical properties of build parts essentially depend on the solid-state phase transformation behavior as well as the microstructure of the build material. To control the microstructure evolution during AM and achieve the desired properties of the final products, a fundamental knowledge about the microstructure formation and growth mechanism is required.

Mathematical modeling can enhance our capability to quantify the influence of process parameters and material properties on the quality of AM builds. Current modeling tools used to simulate materials processing are actively being extended to model LMD. Multi-scale and multi-physics models are needed that will incorporate knowledge of the morphological and microstructure development in materials within the macro-scale continuum models. The microstructural features in the LMD components can include secondary phases (precipitates), dendrites, oriented columnar grains, flat defects and periodic
band structures. The microstructure that results from the phase transformation process plays a critical in determining the physical properties and mechanical behavior of the build materials.

Therefore, understanding the microstructure formation and growth mechanisms during the LMD is an important prerequisite for the process optimization. However, existing LMD models mainly have focused on analyzing macroscopic temperature, solute concentration and fluid velocity fields [1,2]. Modeling of microstructure formation processes in solidification and solid-solid phase transformation (mesoscale processes) during the LMD has received a little attention.

Due to the layer-by-layer build-up of AM parts, the deposited layers experiences cyclic re-heating in the form of “temperature spikes” [3]. This heat treatment is in some cases strong enough to induce solid-state phase transformations, in particular formation of precipitates from supersaturated solid solutions. Solute atom diffusion and precipitate formation processes are also accompanied by composition and phase deformations that significantly complicate the growth kinetics. Controlling heat and mass transfer and crystallization with providing optimized processing parameter set is, therefore, essential to achieve the required residual stress field of the AM parts. The measurement of the stresses and their effects on the growth kinetics of secondary particles are, however, very difficult to be achieved experimentally, as the phase transformation occurs within very short time period. The phase field modelling of processes governing solid-solid phase transformations is one of the most powerful methods for such purposes.

During the past three decades, the phase-field approach has emerged for computational modeling many types of microstructures in various materials processes including dendritic solidification, solid-state structural phase transformations, thermally or diffusional controlled grain growth, coarsening, nucleation, solute trapping, pattern formation on surfaces, etc. [4-7]. The phase-field model (PFM) describes the microstructure development of a material by using a set of field variables (conserved and non-conserved) that are continuous (differentiable) across the interfacial regions. The key advantage of this modeling is that, it considers diffuse interface description; there is no need to track the interfaces during microstructure evolution.

Nowadays the methodology of a thermodynamically consistent derivation of PFM is well established and is employed in many papers (e.g. [6-8]). The governing equations can be derived from an entropy functional following the formalism of irreversible thermodynamics. Wang-Sekerka et al [6] derived phase field equation coupled with thermal and solute concentration fields. Gong et al [9] coupled the thermal conduction modeling with the PFM to study the dendrite morphology evolution during electron beam AM of Ti-6Al-4V alloys. In our recent study [10] we have generalized the model [6] by including the elastic stress fields to describe non-isothermal crystallization of a double-component melt during LMD with powder injection. It has been shown that strain due to phase transformation, transformation plasticity and thermal expansion are the dominant factors that need to be included in the study of the microstructure evolution during LMD. In Ref. [11] a thermodynamically consistent coupled temperature-displacement phase field theory for grain growth during laser-aided metal deposition has been presented. The influence of the temperature dependent material properties on microstructure formation at the phase interface during the melt crystallization has been discussed in this study.

In the present paper, a coupled diffusion-stress formulation for diffusion-controlled growth of precipitates in the powder-based laser additive manufacturing (AM) of metallic parts is presented using phase field approximation. The model is developed in a thermodynamically consistent way, with free-energy expressions associated with microstructural processes, the postulation of energy and mass balances and demonstration that the classical entropy inequality (Clausus-Duhem inequality) is satisfied. The governing equations that follow from the balance laws involve the phase variable, the atomic displacement field, a set of hardening variables related to plasticity, and the solute concentration field, with significant couplings between all equations. The model includes concentration dependent eigen strain, transformation dilatation, strain dependency on precipitate growth and plastic hardening effects. To describe the effects of temperature and fluid velocity distributions and thermal history on the precipitate growth mechanisms a linking of microscale model with the macroscopic AM processing conditions is discussed.
2. The continuum model

Microstructure evolution process during AM has negligible effect on heat transfer and temperature distribution in the build, while the heat transfer is mainly affected by the heat conductivity of the materials, as well as the AM processing parameters such as laser power, laser scan speed, beam diameter, powder feed rate et al. With these assumptions, the precipitate growth process during AM can be decoupled into two sub models on different length scales: (i) the macroscopic thermo-hydrodynamic model to obtain the temperature and fluid velocity distributions and thermal history in the build during the whole AM; (ii) the phase-field model for precipitate growth kinetics to study the precipitate morphologies and their concentrations.

In this section, a coupled diffusion-deformational phase field model is presented for describing the growth of the precipitates occurring during the solid-solid phase transformation of binary alloys. We assume that the second phase can have a different lattice parameter from the mother phase, and therefore elastic stress is generated. The state of considered system at time $t$ is defined by the three field variables: the concentration of solute atoms ($c(\vec{r},t)$), the atomic displacement field ($u(\vec{r},t)$) representing the mechanical motion, and the phase field ($\phi(\vec{r},t)$) representing the precipitate evolution. $\phi = 0$ in the matrix and $\phi = 1$ in the precipitate, and $\phi \in (0,1)$ in the interface between matrix and precipitate. We denote by $\mu(\vec{r},t)$ and $e(\vec{r},t)$ the diffusion potential of the solute and the internal energy density (per unit volume), respectively.

The balances of total energy and mass can be expressed as

$$
\dot{\varepsilon} = \sigma : \dot{\varepsilon},
$$

$$
\dot{\varepsilon} + \nabla \cdot \dot{j}^{(c)} = 0,
$$

where $\dot{j}^{(c)}$ is the diffusion flux, $\sigma$ the Cauchy stress tensor, $\varepsilon$ the total stain tensor consisting of elastic strains ($\varepsilon^{(el)}$), transformation strains ($\varepsilon^{(tr)}$), composition strains ($\varepsilon^{(c)}$) and plastic strains ($\varepsilon^{(p)}$):

$$
\varepsilon = (\nabla \cdot \vec{u}) = \varepsilon^{(el)} + \varepsilon^{(tr)} + \varepsilon^{(c)} + \varepsilon^{(p)},
$$

$$
\varepsilon^{(c)} = \Omega (c - c_{ref}) + e^{(c)}_{ref}.
$$

Here $\Omega = \partial\varepsilon / \partial c$ is the dilatation parameter, representing the volume change associated with the substitution of species in lattice sites (for the particular case of isotropy $\Omega = -\alpha I$, $\alpha = \Delta V (3V_p)^{-1}$, $\Delta V$ is the relaxed lattice volume after one mole of solute atoms is removed from the lattice, and $V_p$ the volume occupied by a mole of atoms; $\varepsilon^{(c)}_{ref}(T)$ is the eigen strain tensor corresponding to the reference solute concentration ($c_{ref}$) at temperature $T$; $\varepsilon^{(p)}$ is the misfit strain due to phase transformation. For the isotropic case $\varepsilon^{(p)} = \chi I$, $\chi$ is equal to $(a_p - a_m)a_m^{-1}$, where $a_p$ and $a_m$ are the lattice parameters of the precipitate and mother phase, respectively.

The dissipation inequality is given by

$$
h_{\phi} \dot{\phi} + \sigma : \dot{\varepsilon} - \psi + \mu \dot{c} - \dot{j}^{(c)} \cdot \nabla \mu \geq 0,
$$

where $\psi$ is the Helmholtz free energy density per unit volume at constant temperature, $h_{\phi} = (\partial h / \partial \phi) + a_\phi \nabla^2 \phi$ ($a_\phi$, the gradient coefficient).

The free energy density ($\psi$), the stress ($\sigma$), and the chemical potential ($\mu$) can be expressed as a function of the following internal state variables characterizing the system behavior: the phase field ($\phi$), the elastic strain ($\varepsilon^{(el)}$), the solute concentration ($c$) and a set of variables ($q$) related to plasticity, such that:

$$
\psi = \dot{\psi}(\dot{A}), \quad \sigma = \dot{\sigma}(\dot{A}), \quad \mu = \dot{\mu}(\dot{A}),
$$

(5)
where \( \tilde{A} \) denotes the list \( \tilde{A}=(\phi,c,e^{(p)},q) \). Substituting these constitutive equations into the free-energy imbalance (4), we find
\[
\left(h_{\psi} T - \frac{\partial \psi}{\partial \phi}\right) \phi + \sigma : \dot{e}^{(p)} + \frac{\partial \psi}{\partial q} \dot{q} - \tilde{j}^{(e)} \cdot \nabla \mu \geq 0,
\]
where we have used
\[
\sigma = \left(\frac{\partial \psi}{\partial q}\right)_{q_0},
\]
\[
\mu = \left(\frac{\partial \psi}{\partial c}\right)_{q_0} - \alpha \text{tr}(\sigma).
\]
From the inequality (6) it is possible to identify four dissipative processes. The first term represents the phase field dissipation, which is related to the rearrangement of atoms during the evolution of the phase fields. The second and third terms represent the dissipation caused by plastic deformation. The last term is the mass transport caused by solute atom diffusion. The positivity of entropy production can be locally assured having chosen the following relationships for the dissipation potential
\[
\mathcal{F}(\sigma,\varepsilon^{p}, \phi, \zeta) \quad \text{is the dissipation potential due to the plastic strains,} \quad \gamma \geq 0 \quad \text{is the plastic multiplier,} \\
\zeta = \partial \psi / \partial q. \quad \text{Eq. (9) represent the solute atom flux. Eq. (10) is the kinetic equation for phase field} \quad \phi. \quad \text{Eq. (11) describes the evolution of the internal variables} \quad (q) \quad \text{in the plasticity model associated with phase field} \quad \phi \quad \text{and the evolution of the plastic strains in phases, respectively.}
\]

3. Free energy functional

The total free energy density of the system is written as function of the phase field \( \phi(r,t) \), the solute concentration \( c(r,t) \), the atomic displacement field \( \tilde{u}(r,t) \) and the set of internal variables \( (q) \) associated to material hardening
\[
\psi(\phi,u,c) = \omega g(\phi) + \psi^{(e)}(\phi,c) + \psi^{(p)}(\phi,u,c) + \psi^{(s)}(\phi,c,q),
\]
where \( g(\phi) = \phi^{2} (1-\phi)^{2} \) is the double-well potential energy and \( \omega > 0 \) is the barrier for the phase transformation. \( \psi^{(e)} \), \( \psi^{(p)} \) and \( \psi^{(s)} \) are, respectively, the elastic, chemical and plastic free energy densities. The chemical free energy density \( \psi^{(c)} \) is interpolated from the chemical energies of the two coexisting phases using the interpolation function \( p(\phi) \) and is given by
\[
\psi^{(c)} = p(\phi) \psi^{(c)}_{m} + (1-p(\phi)) \psi^{(c)}_{p}.
\]

The chemical energy densities \( \psi^{(c)}_{j}(c,T) \) of matrix \( (j=m) \) and precipitate \( (j=p) \) phases are designated as
\[
\psi^{(c)}_{j}(c,T) = \left(\kappa_{j}/2\right)(c-c_{j}(T))^2,
\]
where the coefficient \( \kappa_{j} \) is second derivative of the chemical free energy at equilibrium concentration at the temperature \( T \) in phase \( j \). \( c_{j}(T) \) is the equilibrium concentration of solute atoms for phase \( j \);
\[ p(\phi) = \phi^3 (10 - 15\phi + 6\phi^2) \] is a shape function that is equal to 0 and 1 in the matrix and precipitate, and smoothly changes from 0 to 1 across the interface.

The elastic energy density in (12) is interpolated between the \( p \) and \( m \) phases:

\[ \psi^{(el)} = p(\phi)\psi_p^{(el)} + [1 - p(\phi)]\psi_m^{(el)}. \] (13)

The elastic energy densities \( \psi_j^{(el)} \) given by

\[ \psi_j^{(el)} = \frac{1}{2} \mathbf{e}_j - \mathbf{e}_j^{(p)} - \mathbf{e}_j^{(c)} \] (14)

(\( A_j \) is the stiffness tensors of the phase \( j \)). Using the Voigt-Taylor model, is assumed that the total strain is the same in all coexisting phases (\( \epsilon_m = \epsilon_p = \epsilon \)), but each phase can have different transformation strains, composition strains and plastic strains. Then, combining (13) and (14), we have

\[ \psi^{(el)} = \frac{1}{2} \mathbf{e}^{(el)} : A_{eff}(\phi) : \mathbf{e}^{(el)}, \] (15)

\( A_{eff}(\phi) = A_p (\phi) + (1 - p(\phi)) A_m = A_j + \Delta A p(\phi) \) (\( \Delta A \) are the difference between \( A_j \) and \( A_m \)). For the isotropic case, the stiffness tensor components

\[ A_{ijkl} = \lambda \delta_{ij} \delta_{kl} + G (\delta_{ik} \delta_{pj} + \delta_{ij} \delta_{pk}) \] with Lame elastic constants \( \lambda \) and \( G \). Then \( \psi^{(el)} \) takes the form

\[ \psi^{(el)} = \frac{1}{2} k(\phi)(\nabla \cdot \mathbf{u})^2 + G(\phi)(\epsilon_{ik} - \frac{1}{3} \delta_{ik} (\nabla \cdot \mathbf{u}))^2 - 3k(\phi) (\alpha(\phi)(c - c_\alpha) + \chi(\phi))(\nabla \cdot \mathbf{u}). \] (16)

In (16) \( k(\phi) = k_m + p(\phi) k + \Delta G/2 \) and \( G(\phi) = G_m + p(\phi) \Delta G \) are the bulk and shear moduli of the mixture, \( \Delta k = k_p - k_m, \Delta G = G_p - G_m; \lambda = k - 2G/3 \) (\( k_j \) is the bulk modulus of the phase \( j \), \( G_j \) is the shear modulus of the phase \( j \)); \( \alpha(\phi) = \alpha_m + p(\phi) \Delta \alpha \) is the dilatation volume of the solute atoms (\( \Delta \alpha = \alpha_p - \alpha_m \)). The last term including \( \chi = \chi_m + p(\phi) \Delta \chi \) allows for the stresses generated because of the difference in the volumes of \( p \) and \( m \) phases (\( \Delta \chi = \chi_p - \chi_m \)). For small strains, the total strain is given by \( \epsilon_{ik} = 1/2(\nabla \mathbf{u} + \nabla \mathbf{u} ^T) \).

The plastic free energy \( \psi^{(pl)} \) is taken as

\[ \psi^{(pl)} = p(\phi)\psi_p^{(pl)} + [1 - p(\phi)]\psi_m^{(pl)}, \quad \psi_j^{(pl)} = (1/2) h_j q_j^2, \]

where \( \psi_j^{(pl)} \) is the plastic free energy of phase \( j \); \( h_j(T) \) is the plastic hardening modulus of phase \( j \); the internal hardening variable \( q_j \) is a function of the dislocation density \( (\rho_j^{(div)}) : q_j = (\rho_j^{(div)})^{1/2} - 1. \)

4. Phase field equation coupled to diffusion and stress fields

The local effective stress can be expressed in terms of the local stresses with respect to \( p \) and \( m \) phases. Substituting (14) into (7), for the stress of heterogeneous materials yields

\[ \sigma = A_{eff}(\phi) \left[ \epsilon - \epsilon^{(p)} - \epsilon^{(c)} \right] \] (17)

where the local eigen strains (\( \epsilon^{(c)} \) and \( \epsilon^{(p)} \)) and plastic strain \( (\epsilon^{(p)}) \) vary continuously between their respective values in each phase

\[ \epsilon^{(c)} = A_{eff}^{-1} \left[ p(\phi) A_p : \epsilon^{(c)}_p + (1 - p(\phi)) A_m : \epsilon^{(c)}_m \right], \]

\[ \epsilon^{(p)} = A_{eff}^{-1} \left[ p(\phi) A_p : \epsilon^{(p)}_p + (1 - p(\phi)) A_m : \epsilon^{(p)}_m \right], \]

\[ \epsilon^{(c)} = A_{eff}^{-1} \left[ p(\phi) A_p : \epsilon^{(c)}_p + (1 - p(\phi)) A_m : \epsilon^{(c)}_m \right], \]

\[ \epsilon^{(p)} = A_{eff}^{-1} \left[ p(\phi) A_p : \epsilon^{(p)}_p + (1 - p(\phi)) A_m : \epsilon^{(p)}_m \right]. \]
Substituting (12) into (10), we obtain the following kinetic equation of the phase field

\[ m_p^{-1} T \phi = \alpha_p^2 T \nabla^2 \phi - \omega \frac{d\phi}{d\phi} \left( \psi^{(c)} - \psi^{(c)} \right) \frac{dp}{d\phi} \]

\[ + 3 \left[ k(\Delta \alpha(c-c_0) + \Delta \chi) + \Delta k(\alpha(\phi)(c-c_0) + \chi(\phi)) \right] (\nabla \cdot \vec{u}) \frac{dp}{d\phi} \]

\[ + \left( 0.5 \Delta k(\nabla \cdot \vec{u})^2 + \Delta G(\varepsilon_\alpha - \frac{1}{2} \delta_\alpha (\nabla \cdot \vec{u}))^2 \right) \frac{dp}{d\phi} - \left( \psi^{(p)} - \psi^{(p)} \right) \frac{dp}{d\phi}. \]

(18)

Using the Equation (8), the diffusion flux becomes

\[ \vec{j}^{(c)} = -m_c(\phi, T) \nabla \left( \frac{\partial \psi^{(c)}}{\partial \phi c} - \alpha \text{tr}(\sigma) + \frac{\partial \psi^{(p)}}{\partial c} \right). \]

(19)

Equation (19) clearly reveals the two main driving forces controlling the solute atom flux: the concentration gradient term arising from inhomogeneities in the composition, and the mechanical contributions via the elastic stress field gradient and the plasticity effect. Then the diffusion-drive equation for evolution of solute atom concentration is obtained as

\[ \dot{c} + \nabla \cdot \left[ -m_c(\phi, T) \nabla \left( \frac{\partial \psi^{(c)}}{\partial \phi c} - \alpha \text{tr}(\sigma) + \frac{\partial \psi^{(p)}}{\partial c} \right) \right] = 0, \]

(20)

where the mobility \( m_c(\phi, T) \) is defined with respect to diffusivities \( D_\rho \) and \( D_m \) in precipitate and mother phases by means of the interpolation function \( p(\phi) \) as

\[ m_c = p(\phi) D_\rho \kappa^{-1} + (1 - p(\phi)) D_m \kappa^{-1}. \]

The time scale for the relaxation of the stress field, as described by the atomic displacement field, is much shorter than that of the solute diffusion field. Therefore, displacement rapidly reaches equilibrium the time scale of solute diffusion, giving rise to mechanical equilibrium: \( \nabla \cdot \sigma = 0 \), which with the use of (17) becomes

\[ \nabla \cdot \left[ \Lambda(\phi) \left( \varepsilon - \varepsilon^{(c)}(\phi, T, c) - \varepsilon^{(p)}(\phi, T, c, q) \right) \right] = 0 \]

(21)

or in the case of isotropic materials

\[ \nabla \left[ k(\phi)(\nabla \cdot \vec{u}) - 3k(\alpha(\phi)c + \chi(\phi)) \right] + 2 \nabla \cdot \left[ G(\phi)(\varepsilon_\alpha - \frac{1}{2} \delta_\alpha \nabla \cdot \vec{u}) \right] = 0. \]

Equations (18), (20) and (21) are the complete set of governing equations for the precipitate growth kinetics during LMD. With proper initial and boundary conditions, the system will be well-posed.

5. Conclusions

In this work, a coupled diffusion-stress formulation for diffusion-controlled growth of precipitates during AM of binary alloys has been presented using phase field approximation. The model has been developed in a formal thermodynamic setting, with entropy functional instead of a Helmholtz free energy functional associated with microstructural non-isothermal processes, the postulation of energy balance and demonstration that the classical entropy inequality is satisfied. Various couplings terms introduce dependencies between different processes. The governing equations that follow from the fundamental balance laws involve the phase variable, the atomic displacement field, and the solute concentration field, with significant couplings between all equations. The model includes concentration-dependent eigenstrain, strain due to phase transformation, elastic-plastic deformation, strain dependency on phase transformation and local mechanical equilibrium conditions.

The precipitate growth during LMD is determined by the thermal history of the materials, which is result of laser energy absorption by the powder particles and substrate, heat and mass transfer within the build part and heat losses. To describe the effects of temperature and fluid velocity distributions and thermal history on the precipitate growth kinetics the developed theory should added to the macroscale

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model [10] of dynamic processes accompanied LMD. The model of the processes at the macroscopic level considered by [10] takes into account the interaction of laser radiation with the powder particles, the powder injection, the evolution of the free surface (the interface between the melt-gas phase), hydrodynamics and heat and mass transfer in the fusion zone. The energy equation of the multiphase mixture takes into account convective and conductive flows, as well as the latent heat release accompanying the phase transition. The dynamics of convective currents is modeled with the use of a complete system of equations for the continuity of a multiphase mixture and the motion of a multiphase mixture (of the Navier-Stokes type) with allowance for the action of capillary and thermo-capillary forces on the free surface of the melt. For tracking the free surface of the melt, a combined volume of fluid and level-set method is used. At this structural level the processes in a scale of the condensed system in the whole are considered and processes in two-phase region, where the formation of the microstructure takes place are not included. Thus, macromodel predicts the temperature field coupled with the convection flow velocity, and changes in the form of the free surface in the process of LMD.

The resultant inhomogeneous temperature distribution and thermal history from the macroscopic thermo-hydrodynamic model can be used as the initial and boundary conditions for solute atom diffusion, which in turn controls the growth of precipitates in LMD. Linking of microstructure evolution by considering the relationship of micro- and macroparameters, including laser and powder parameters such as laser power, scanning probe size and scanning speed, powder feed rate will lead to a better understanding of different aspects of the precipitate growth and morphological development during the LMD.

We emphasize that the purpose of this paper was only to report on the formulation of the theory. We leave a report concerning its numerical implementation and use for analysis of strain effects on the precipitate growth, and the more difficult problem of stress generation in phase transformations including morphology instabilities, to future work.

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