On the phase field model for microstructure evolution during selective laser melting of powder bed including diffusion-induced stresses

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Abstract. In this paper, we develop a phase field model for microstructure evolution in laser powder-bed additive manufacturing processes, which allows for the effect of elastic stresses due to diffusion of atomic defects (vacancy and interstitial atom) generated at the moving liquid-solid interface. The governing equations of the model including a time-dependent Ginsburg-Landau equation for the phase field with additional stress term, a diffusion-drift Cahn–Hilliard like equation describing atomic defect dynamics, an energy balance equation for the temperature change, and finally the mechanical equilibrium equation for the elastic displacement fields are derived within a thermodynamic framework based on entropy generation guaranteeing thermodynamic consistency. The most simple 1D and isothermal equilibrium situation is considered to study phenomenon of diffusion-flexural instability with the atomic defect clustering and periodic distributions of defects. The effect of this instability on the exfoliation of deposited thin layers on a substrate is also discussed.

1. Introduction

Selective laser melting (SLM) is a popular additive manufacturing (AM) process in which a high energy laser beam scans the surface of a powder bed and the melted powder solidifies to form the bulk part. Nowadays, SLM technology is widely used in aerospace, biomedical, energy, and automotive industries [1-3]. This process has many parameters (power, beam size, scan velocity, layer thickness, etc.), that in combination with material properties influence quality and mechanical properties of SLM parts [3, 4].

An important role in the AM process is played mechanical stresses and formation of various defects. Manufacturing defects and their effects on microstructural and physical properties of AM parts are the most difficult problems. It is important to understand the types of defects, the mechanisms of their formation, as well as detection methodologies for assessing mechanical properties and quality control. Currently, the task of reducing them is not solved. As a rule, these effects are associated with both the process of melting the powder material (gas porosity) and the solidification of the melt. In the first case, the significant effects affecting the formation of defects (pores) are beam power density, scanning speed, surface tension, Marangoni convection and recoil pressure [5-7]. In the second case, the formation of micropores is due to the generation of atomic point defects (PD) (vacancies and interstitials) formed by fluctuations at the solid site of the liquid-solid (l-s) interface and their clustering during diffusion.

It is experimentally sufficiently difficult to determine the parameters and their combinations that have the greatest influence on the mechanical and physical properties of the product. SLM is an
inherently multi scale, multi physics process and calls for an approach using multiple coupled models. Existing modeling tools used to study laser processing of materials are currently being actively extended to AM modeling. There are many different approaches to modeling thermal history and the resulting profiles of parts manufactured using AM processes. Most of the existing studies use numerical methods because of the complexity of the phenomena that appear [6–10]. However, multiscale models of solidification are needed, which will include morphological evolution and growth of various microstructures (grain growth, dendrites, precipitates, etc.). In Refs [11, 12], computational thermo-fluid dynamics analysis is used to predict macroscopic characteristics (melt pool geometry, thermal history and temperature distributions) and phase field modeling based on Kobayashi model [13] is employed to simulate microstructure development during SLM process. In these studies influence of processing parameters on different features of SLM microstructure such as segregation of secondary elements, dendrite sizes, dendritic orientation and morphology are also investigated and validated through comparison with experimental results.

One of the important tasks is to take into account the elastic stress effects in the multiscale models of the microstructure evolution during SLM. This is of general interest for applications in which stress fields act as a stable driving force for phase transformations (PT) or as a basic field for sequential modeling processes such as generation and motion of non-equilibrium PDs, plasticity and others that accompany rapid solidification of the melt. PDs lead to local changes in interatomic distances and, as a result, to lattice distortions. During the cooling process, PDs can migrate either due to a chemical potential gradient or elastic interaction, or to be trapped by other defects or so-called absorbers, such as dislocations, grain boundaries. The density and stability of the absorber will determine the accumulation of mobile PDs created at the (l-s) interface, which affects the stability of the phase and / or structure, as well as the kinetics of microstructure development and material properties. Residual stresses and distortions that are caused by non-uniform thermal phenomena (heating and cooling) and PD generation, which occur in AM, degrade the mechanical properties and dimensional accuracy of the parts [14].

In the present contribution a physically thermodynamically consistent phase field model (PFM) proposed by Wang et al. [15] is extended to describe the microstructure development during SLM taking into account the coupled processes of heat transfer, stress effects and PD generation on the (l-s) interface. Constitutive relations are derived on the basis of a single entropy functional and the principle of entropy production positiveness. The application of this principle allows obtaining consistent evolutionary equations for the phase field coupled with the fields of temperature, PD concentration and atomic elastic displacements for non-isothermal conditions of crystallization. Some applications of the developed coupled model of defect formation during solidification are considered. Diffusion-flexural instability with the formation of PD clusters (micropores and dislocation loops) and periodic distributions of PDs and their effect on the exfoliation of deposited thin layers on a substrate are also discussed.

2. Formulation of the problem
Microstructure evolution during SLM 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, scan speed, beam diameter, et al. With these assumptions, the microstructure evolution during SLM 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 SLM; (ii) the PFM for microstructure evolution during solidification at the SLM process.

We assume that the second phase can have a different lattice parameter from the mother phase, and therefore elastic stress is generated. It is also assumed that the generation of PDs on the (l-s) interface occurs. Within the phase field conception, the state of considered system at time $t$ is defined by the five field variables: the temperature field $T(r,t)$, the atomic displacement field $(u(r,t))$ representing the mechanical motion, the phase field $\phi(r,t)$ for microstructure evolution, and the concentration of PDs
\( n_v(n, T) \) \((d = v \text{ for vacancies and } d = i \text{ for interstitials})\). The order parameter \( (\phi) \) is defined as 0 in liquid and 1 in solid, and smoothly varies over an interface. We denote by \( \mu(r, t), \eta(r, t) \) and \( e(r, t) \) the diffusion potential of PDs, the entropy and internal energy densities (per unit volume), respectively. We use a superposed dot to denote time differentiation.

The balance laws for the energy and PD concentration can be written as

\[
\dot{\varepsilon} = -\nabla \cdot \mathbf{j}^{(e)} - \nabla \cdot \left( \mu \mathbf{j}^{(v)} \right) - \nabla \cdot \left( \mu \mathbf{j}^{(i)} \right) + \sigma \dot{\varepsilon},
\]

(1)

\[
\dot{n}_v = -\nabla \cdot \mathbf{j}^{(v)} + Q_v(T, \phi) - R_v(n_v, n_i, T),
\]

(2)

\[
\dot{n}_i = -\nabla \cdot \mathbf{j}^{(i)} + Q_i(T, \phi) - R_i(n_i, n_v, T),
\]

(3)

where \( \mathbf{j}^{(v)} \) is the energy flux, \( \mathbf{j}^{(v)} \) and \( \mathbf{j}^{(i)} \) the fluxes of \( v \) and \( i \)-defects, respectively, \( \mu_v \) and \( \mu_i \) the diffusion potentials of PDs, \( \sigma \) the stress tensor, \( \varepsilon \) the total strain tensor; \( Q_{v,i}(T, \phi) \) the PD generation rates on \((l-s)\) interface, \( R_{v,i}(n, T) \) characterize the losses of PD concentration associated with their recombination. The second and third terms on the right side of the Eq. (1) represent the flux of energy carried by the PD fluxes [16] and the last term - changes due to elastic strain.

We restrict attention to small strains, thus the total strain tensor is given by the sum of the elastic, thermal, concentration and phase strains, \( \varepsilon^{(d)}, \varepsilon^{(a)}, \varepsilon^{(d)} \) and \( \varepsilon^{(a)}, \) respectively

\[
\varepsilon = \varepsilon^{(d)} + \varepsilon^{(a)} + \varepsilon^{(d)} + \varepsilon^{(a)},
\]

(4)

\[
\varepsilon^{(a)} = \mathbf{B} \left( T - T_{ref} \right), \quad \varepsilon^{(d)} = \sum_{d=e,s} \Omega_d \left( n_d - n_{d0} \right),
\]

where the tensor \( \mathbf{B} = \partial \varepsilon / \partial T \) takes into account the strains due to thermal expansion; \( \varepsilon^{(d)} \) is the PD induced eigen-strain, the tensor \( \Omega_d = \partial \varepsilon / \partial n_d \) scales the defect concentration changes, \( n_{d0} \) and \( n_{d0} \) are the equilibrium PD concentrations. For the particular case of isotropy: \( \Omega = -\alpha \mathbf{I}, \quad \alpha = (1/a) da/dn, \) \( a \) is the lattice parameter and \( \mathbf{I} \) is the second order identity tensor; \( \varepsilon^{(a)} \) is the strain generated from the difference in the volumes of phases \( l \) and \( s \). For the isotropic case \( \varepsilon^{(a)} = \chi_0 \mathbf{I}. \) The transformation expansion coefficient \( \chi_0 \) is expressed by the formula obtained by converting the rate of volumetric change into a linear expansion coefficient \( \chi_0 \approx (v_s - v_l) / 3v_l. \) The total strain tensor is given by \( \varepsilon_{tot} = 1/2 \left( \nabla_v u_l + \nabla_l u_v \right). \)

The Clausius-DuGue inequality is given by

\[
\eta \geq -\nabla \cdot \left( T^{-1} \mathbf{j}^{(v)} \right) - \alpha^2_{\phi} \dot{\phi} \nabla^2 \phi
\]

(5)

\( \alpha^2_{\phi} \) is a gradient-energy coefficient). With the help of Eqs. (1)-(3), the inequality (5) becomes

\[
\alpha^2_{\phi} \dot{\phi} \nabla^2 \phi + \sigma \cdot \dot{\varepsilon} - \eta T + \mu_v \dot{n}_v + \mu_i \dot{n}_i - \mathbf{j}^{(v)} \cdot \nabla \mu_v - \mathbf{j}^{(i)} \cdot \nabla \mu_i + T \mathbf{j}^{(e)} \cdot \nabla T^{-1} \geq 0,
\]

(6)
where we introduced Helmholtz free energy function $\psi$, defined by $\psi = e - T \eta$. The function $\psi$ (and all other functions under consideration (the stress ($\sigma$), the entropy ($\eta$))) can be expressed in terms of the following internal state variables characterizing the system behavior: the phase field ($\phi$), the temperature field ($T$), the elastic strain ($\varepsilon_{ik}^e$) and PD concentrations ($n_v$ and $n_i$), such that

$$\psi = \hat{\psi}(A), \quad \sigma = \hat{\sigma}(A), \quad \eta = \hat{\eta}(A), \quad (7)$$

where $A$ denotes the list $A = (\phi, T, \varepsilon_{ik}^e, n_v, n_i)$.

Substituting Eq. (7) into Eq. (6), we get

$$
\begin{align*}
\left(\alpha_\phi \nabla^2 \phi - \frac{\partial \hat{\psi}}{\partial \phi}\right) \hat{\phi} &- \left(\eta + \frac{\partial \hat{\psi}}{\partial T} - \beta \text{tr}(\sigma)\right) \hat{T} - \left(\mu_i + \alpha_i \text{tr}(\sigma) - \frac{\partial \hat{\psi}}{\partial n_v}\right) \hat{n}_v \\
- \left(\mu_i + \alpha_i \text{tr}(\sigma) - \frac{\partial \hat{\psi}}{\partial n_i}\right) \hat{n}_i &+ \left(\sigma - \frac{\partial \hat{\psi}}{\partial \varepsilon_{ik}}\right) : \dot{\varepsilon} - j^{(e)} \cdot \nabla T - j^{(i)} \cdot \nabla n_i - j^{(e)} \cdot \nabla n_v \geq 0.
\end{align*}
\quad (8)
$$

If in inequality (8) we take into account that, and $\hat{T}$, $\hat{n}_a$, $\hat{n}_i$ and $\dot{\varepsilon}$ can change in an independent way, then to satisfy it at any values of $\hat{T}$, $\hat{n}_a$, $\hat{n}_i$ and $\dot{\varepsilon}$, we need to put

$$\sigma = (\partial \hat{\psi} / \partial \varepsilon)|_{\hat{\phi}, \hat{T}},
\eta = -(\partial \hat{\psi} / \partial T)|_{\hat{\phi}, \hat{T}} + \beta \text{tr}(\sigma), \quad \mu_i = -\alpha_i \text{tr}(\sigma) + (\partial \hat{\psi} / \partial n_i)|_{\hat{\phi}, \hat{T}},
\quad (9, 10)$$

at

$$
\begin{align*}
\left(\alpha_\phi \nabla^2 \phi - \frac{\partial \hat{\psi}}{\partial \phi}\right) \hat{\phi} - T^{-1} j^{(e)} \cdot \nabla T - j^{(i)} \cdot \nabla n_i - j^{(e)} \cdot \nabla n_v \geq 0.
\end{align*}
\quad (11)
$$

From the inequality (11) it is possible to identify three 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 term is the heat transport caused by thermal conductivity. The last two terms represent the dissipation caused by PD diffusion processes. The positivity of entropy production can be locally assured having chosen the following relationships for the diffusional flows and the time derivatives of variables

$$j^{(e)} \cdot \nabla T \leq 0, \quad j^{(e)}(\phi, T) = m_\phi(\phi, T) \nabla T^{-1}, \quad (12)$$

$$j^{(i)}(\phi, T, n_i) \nabla n_i, \quad j^{(e)}(\phi, T, n_i) = m_i(\phi, T, n_i) \nabla n_i, \quad (13)$$

where $m_\phi$ is the phase field mobility, $m_i > 0$, $m_e > 0$ and $m_e > 0$ characterize PD diffusion and thermal conductivity, respectively.
3. Derivation of the governing equations

We assume that the total free energy density is given by

$$\psi = \psi^{(dw)} + \psi^{(th)} + \psi^{(el)} + \psi^{(ch)}$$  \hspace{1cm} (15)

with

$$\psi^{(dw)}(\phi) = w_c g(\phi) = w_c \phi^2 (1 - \phi)^2,$$

$$\psi^{(th)}(T) = \Phi_i^{(th)}(T) + \Delta \Phi p(\phi),$$

$$\psi^{(ch)}(T, n_i, n_s) = \frac{N_a}{v_a} \left[ E_i' n_i + E'_s n_s + k_n T \left[ n_i \ln(n_i) + n_s \ln(n_s) - (1 - n_i - n_s) \ln(1 - n_i - n_s) \right] \right].$$

In the above equations $\psi^{(dw)}(\phi, T)$ denotes the double–well potential, guaranteeing that the free energy function has two local minima at $\phi = 0$ and $\phi = 1$ corresponding to the l and s phases of the system, $w_c$ is the height of the energy barrier between phases; $\Phi_i^{(th)}(T)$ is the free-energy density of the liquid ($\Phi_i^{(th)}(T) = c(T - T_m) - cT \ln(T/T_m)$, $c$, the heat capacity, $T_m$, the melting temperature); $\Delta \Phi$ is the difference between the energies of the l and s phases, $\Delta \Phi = L(T - T_m)/T_m$ ($L$ is the latent heat of fusion); $p(\phi) = \phi^3 (10 - 15\phi + 6\phi^2)$ is the interpolation function with $p(0) = 0$ and $p(1) = 1$ for variation of many material property between phases. $E_i'$ the formation energies of PDs, $N_a$ the Avogadro number; $v_a$ the molar volume.

The elastic energy density is interpolated between the two phases using a smooth function $p(\phi)$:

$$\psi^{(el)}(\mathbf{u}, T, n_i, n_s, \phi) = p(\phi)\psi^{(el)}(\mathbf{u}, T, n_i, n_s, \phi) + [1 - p(\phi)]\psi^{(el)}(\mathbf{u}, T, \phi).$$ \hspace{1cm} (16)

The elastic energies of the s and l phases are given by

$$\psi^{(el)}(\mathbf{u}, T, \phi, n_i, n_s) = 0.5 \left( \mathbf{e} - \mathbf{e}^{(s)} - \mathbf{e}^{(l)} \right) : \Lambda_i : \left( \mathbf{e} - \mathbf{e}^{(s)} - \mathbf{e}^{(l)} \right),$$ \hspace{1cm} (17)

$$\psi^{(el)}(\mathbf{u}, T, \phi) = 0.5 \left( \mathbf{e} - \mathbf{e}^{(l)} \right) : \Lambda_i : \left( \mathbf{e} - \mathbf{e}^{(l)} \right).$$ \hspace{1cm} (18)

($\Lambda_i$ and $\Lambda_i$ are constants). In Eq. (18) it is assumed that the elastic strain is equal in all the phases assumed to be present as a mixture at every point in the diffuse interface region corresponding to Voigt/Taylor homogenization model. It should be noted that the contribution to the elastic energy due to PDs ($\mathbf{e}^{(l)}$) is nonzero only for the solid phase. Combining Eqs. (4) and (16)-(18) we have

$$\psi^{(el)}(\mathbf{u}, T, \phi, n_i, n_s) = 0.5 \mathbf{e}^{(el)} : \Lambda(\phi) : \mathbf{e}^{(el)}.$$ \hspace{1cm} (19)

where $\Lambda(\phi) = \Lambda_i + p(\phi) \Delta \Lambda$, $\Delta \Lambda = \Lambda_i - \Lambda_i$. In the case of an isotropic medium $\Lambda_{ijkl} = \lambda \delta_{ij} \delta_{kl} + G (\delta_{ik} \delta_{jl} + \delta_{jk} \delta_{il})$ and the elastic moduli ($\Lambda$) depend only on the Lame constant ($\lambda$) and the shear elastic modulus ($G$), so that
$$\psi^{(el)}(u, T, \phi) = \frac{1}{2} k(\phi)(\nabla \cdot u)^2 + G(\phi)(\epsilon_{ik} - \frac{1}{3} \delta_{ik}(\nabla \cdot u))^2 - 3k(\phi)\left[\beta(\phi)(T - T_0) + \alpha_1(\phi)(n_i - n_{i0}) + \alpha_2(\phi)(n_v - n_{v0}) + \chi(\phi)\right](\nabla \cdot u).$$  (20)

In Eq. (20) \(k(\phi) = k_i + p(\phi)\Delta k\) and \(G(\phi) = G_s p(\phi)\) are the bulk and shear moduli of the mixture, \(\Delta k = k_s - k_i,\ \lambda = -2G/3\) \(k_{s,i}\) are the bulk moduli of the \(s\) and \(l\) phases, respectively, \(G_s\) the shear elastic modulus of the \(s\) phase; \(\beta(\phi) = \beta_i + p(\phi)\Delta \beta\) the linear thermal expansion coefficient of the mixture; \(\beta_{s,i}\), the thermal expansion coefficients of the \(s\) and \(l\) phases; \(\Delta \beta = \beta_s - \beta_l;\ \alpha(\phi) = \alpha_0 p(\phi),\) the dilatation volume of the PDs. The last term including \(\chi = \chi_0(T)p(\phi)\) allows for the stresses generated because of the difference in the volumes of \(l\) and \(s\) phases.

Then, the expression for the elastic stress, following from Eq. (9) and Eq. (20) has the form

$$\sigma_{ik} = \lambda(\phi)\delta_{ik}\nabla \cdot u + 2G(\phi)\epsilon_{ik} - 3k(\phi)\left[\beta(\phi)(T - T_0) + \alpha_1(\phi)(n_i - n_{i0}) + \alpha_2(\phi)(n_v - n_{v0}) + \chi(\phi)\right].$$  (21)

In view of Eq. (15), the constitutive Eqs. (10) take the form

$$\eta = n_0 + c \ln(T/T_0) - \nu_{\phi} g(\phi) + Lp(\phi) + \beta \text{tr} (\sigma) \tag{22}$$

$$-Rn^{-1}_p\left(n_i \ln(n_i) + n_v \ln(n_v) - (1 - n_i - n_v)\ln(1 - n_i - n_v)\right)$$

$$\mu_i = \hat{\psi}^{(el)} \hat{\nabla}_{n_i} - \sigma^{(el)} : \mathbf{\Omega}^{(el)} p(\phi),$$  (23)

$$\mu_i = \hat{\psi}^{(el)} \hat{\nabla}_{n_i} - \sigma^{(el)} : \mathbf{\Omega}^{(el)} p(\phi).$$  (24)

To obtain the phase field kinetic equation, we substitute Eq. (15) into Eq. (14), we get

$$m_{\psi} \phi = \alpha_0^2 T \nabla \cdot (\hat{\nabla} \phi) + \omega_\phi \frac{d\phi}{d\phi} + \left[\Delta \Phi(T) + 3k(\phi)(T - T_0) + \Delta \alpha_1 (n_i - n_{i0}) + \Delta \alpha_2 (n_v - n_{v0}) + \chi_0 \right](\nabla \cdot u) \frac{dp}{d\phi} \tag{25}$$

$$+ \left[0.5 \Delta k(\nabla \cdot u)^2 + G_s (\epsilon_{ik} - \frac{1}{3} \delta_{ik}(\nabla \cdot u))^2\right] \frac{dp}{d\phi}.$$

Using the energy balance Eq. (1), as well as the constitutive relation (7), we have the following energy equation

$$\dot{T}\eta + \frac{\partial \psi}{\partial \phi} \dot{\phi} + \frac{\partial \psi}{\partial n_i} \dot{n}_i + \frac{\partial \psi}{\partial n_v} \dot{n}_v = -\nabla \cdot \mathbf{j}^{(el)} + \nabla \mu_i \cdot m_n \nabla \mu_i + \nabla \mu_v \cdot m_n \nabla \mu_v. \tag{26}$$

If in Eq. (26) to substitute the expression for \(\eta\)

$$\eta = \left(\frac{\partial \eta}{\partial T}\right) T + \left(\frac{\partial \eta}{\partial \epsilon^{(el)}}\right) \mathbf{j}^{(el)} + \left(\frac{\partial \eta}{\partial \phi}\right) \dot{\phi} + \left(\frac{\partial \eta}{\partial n_i}\right) \dot{n}_i + \left(\frac{\partial \eta}{\partial n_v}\right) \dot{n}_v,$$
and take the designation for heat capacity \( c = -T \frac{\partial^2 \psi}{\partial T^2} \), then the energy conservation law takes the form of the heat equation, which takes into account the relationship between temperature, PD concentration, stress and phase field:

\[
c \dot{T} = \nabla (k \nabla T) - 3T \beta \phi \nabla(\phi) + \nabla \mu_s \cdot m_s \nabla \mu_s + \nabla \mu_v \cdot m_v \nabla \mu_v +
\]

\[
T \left( \frac{\partial \mu_s}{\partial T} \hat{n}_s + \frac{\partial \mu_v}{\partial T} \hat{n}_v \right) - \left( \frac{dp}{d\phi} L + Tw \frac{dg}{d\phi} \right) \phi.
\]

(27)

Here we used the total heat capacity \( c = c_s + (c_v - c_s) \lambda(\phi) \) (\( c_s \) and \( c_v \) are the heat capacity for the melt and solid, respectively) and the thermal conductivity \( k = m_s T \alpha_s = \kappa_s + (\kappa_v - \kappa_s) \lambda(\phi) \) (\( \kappa_s \) and \( \kappa_v \) are thermal conductivity for the melt and solid, which are measured experimentally). In (27) thermo-mechanical coupling term \( 3T \beta \phi \nabla(\phi) \) in the right-hand side is responsible for the Gough-Joule effects, i.e. when strain lead to structural heating and the coupling terms from third to sixth arise due to dissipative effects caused by the PD diffusion.

With the use of Eq. (23) and Eq. (24) the PD fluxes are expressed as

\[
j^{(k)} = -D_{jk} \nabla n_k + \frac{1}{3} \frac{D_k}{k_B T} n_k (1 - n_k) \nabla \left[ \phi \left( \frac{1}{\phi} \nabla : \Omega^{(s)} \right) \right]
\]

(28)

(\( D_k = k_B T m_n(T) / n_k (1 - n_k) \) the diffusivity of PDs, \( k = \{ i, v \} \)). Combining this expression with Eq. (2) and Eq. (3), we obtain the equations of the kinetics of the PDs subsystem in the form

\[
\frac{d}{d\tau} n_v = \nabla \left( D_v \nabla n_v \right) + \frac{1}{3} \frac{D_v}{k_B T} \nabla \left( n_v (1 - n_v) \nabla \left( \lambda(\phi) \phi \left( \frac{1}{\phi} \nabla : \Omega^{(s)} \right) \right) \right)
\]

\[
- n_v(T) 6 \lambda(\phi) \phi - \tau_1 n_v - R_{v,v}
\]

(29)

\[
\frac{d}{d\tau} n_i = \nabla \left( D_i \nabla n_i \right) + \frac{1}{3} \frac{D_i}{k_B T} \nabla \left( n_i (1 - n_i) \nabla \left( \lambda(\phi) \phi \left( \frac{1}{\phi} \nabla : \Omega^{(s)} \right) \right) \right)
\]

\[
- n_i(T) 6 \lambda(\phi) \phi - \tau_1 n_i - R_{i,i}, \quad (\phi \neq 0)
\]

(30)

with \( R_{v,v} = 4\pi (D_v + D_i) \lambda \exp \left( -E_{\psi} / T \right) \left( n_v n_i - n_v^{\text{eq}} n_i^{\text{eq}} \right) \) and \( n_v^{\text{eq}} = 0, \phi = 0 \). \( \tau_1 \) and \( \tau_1^{-1} \) denote the recombination rates of PDs on neutral sinks. The last term on the right-hand side (29) and (30) takes into account the effects of mutual recombination of PDs.

The elastic field can be obtained in terms of the phase field using the local equilibrium condition. Ignoring inertia and body forces, conservation of linear momentum yields: \( \nabla \cdot \sigma = 0 \) or

\[
\nabla \left[ k(\phi) (\nabla \cdot \mathbf{u}) - 3k(\phi) n_i + \alpha(\phi) n_v + \beta(\phi) T + \chi(\phi) p(\phi) \right] + 2 \nabla \left[ G(\phi) (\phi - \frac{1}{3} \delta_{\phi} \nabla \cdot \mathbf{u}) \right] = 0.
\]

(31)

Eqs. (25), (27) and (29)-(31) constitute a closed system of PFM equations for the microstructure development during SLM. If PD generation and stress effects are absent on the (i-s) interface the proposed model reduces to Wang model [15] for solidification kinetics. Some applications of the developed coupled model of PD formation during solidification process are considered in section 5.
4. Coupling between the microscopic and macroscopic models
The microstructure evolution 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 microstructure growth kinetics the developed theory should added to the macroscale model [17] of dynamic processes accompanied SLM. The model of the processes at the macroscopic level considered by [17] takes into account the interaction of laser radiation with the powder particles, 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 with allowance for the action of capillary and thermo-capillary forces on the free surface. For tracking the free surface of the melt, a volume of fluid method can be 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 SLM.

The temperature distributions found from the microscopic model can be used as boundary and initial conditions for solving the problem of the evolution of the microstructure. It allows investigating the influence on the microstructure formation of both the SLM parameters and the parameters of microprocesses such as diffusion of heat and defects, diffusion-induced stresss, strain-induced drift of PDs. In addition, based on it, instability phenomena accompanying the melt solidification process can be studied. In particular, at high PD concentration, due to the defect-strain positive feedback, both periodic distributions of defects and their localized states - stable PDs clusters can occur.

5. Diffusion-elastic instability in solids and elastic layers
One of the most important manifestations of the cooperative defect-strain interaction during microstructure evolution is the phenomenon of clusterization of similar atomic PDs, with the formation of micro- and nanometer-clusters. The clustering process occurs due to instability of the homogeneous distribution of carriers interacting through the elastic fields created by them (the diffusion-elastic instability (DEI)) [14]. DEI occurs when the average carrier concentration exceeds a certain critical value. With DEI, fluctuations in the concentration of carriers, being elastic inclusions, create a strain of the elastic continuum, which in turn leads to the appearance of strain-induced drift of the PDs. Due to the redistribution of carriers, forces appear that are proportional to the gradient of their concentration (\( \nabla n \)) and directed in the compression region (for \( v \)-defects with dilatation volume \( \alpha_v < 0 \)) and in the stretching region (for \( i \)-defects with \( \alpha_i > 0 \)). When a certain critical concentration \( n_{cr} \) is exceeded, these forces increase small fluctuations of PD concentrations, and an instability of the concentration field (as well as the field of elastic deformation of the medium) arises.

With the use of Eq. (28), the expression for the total flux of defects becomes

\[
\mathbf{j}^{(s)} = j_D + j_x = -D_v \left( 1 - nk\alpha^2 / k_B T \right) \nabla n = D_{eff} (n) \nabla n.
\] (32)

Inclusion of the elastic deformation effects gives rise to an additional term in the above expression (\( j_x \)) and it represents the flux opposite to the usual diffusion (\( j_D \)) in a system with defects of the same type (\( \alpha^2 > 0 \) for \( \alpha > 0 \) and \( \alpha < 0 \)). In the case of PDs of different types (\( \alpha, \alpha', < 0 \)) the additional flux has the same direction as the diffusion flux. The PDs with \( \alpha < 0 \), for example, vacancies, compress
the lattice (reduce the volume) and the compressed regions attract PDs with $\alpha_j < 0$ and repel those with $\alpha_j > 0$.

The quantitative conditions for the appearance of clusters in a system of PDs of the same type can be found by substitution of Eq. (32) into continuity equation $\dot{n} + \nabla \cdot \mathbf{j}^{(i)} = Q_0 - \tau^{-1}n$ ($Q_0 = \text{const}$) and linearization of the latter in the vicinity of the homogeneous solution ($n^{(0)} = Q_0 \tau$). The DEI increment ($\Gamma$) is then

$$\Gamma(q) = -Dq^2 \left(1 - n^{(0)}k\alpha^2/k_nT\right) - \tau^{-1}$$

($q$ is the wave number). It therefore follows that the system is unstable if

$$Q_0 > Q_{cr} = k_nT \tau k^{-3} \alpha^{-2}, \quad q^2 > q_{cr}^2 = k_nT_0 \tau^{-1} \left(k\alpha^2n^{(0)} - k_nT_0\right)^{-1}.$$  \hspace{1cm} (33)

Physically, the fulfillment of conditions (33) is equivalent to a local change in the sign of the effective diffusivity of PDs: $D_{\text{eff}} = D \left(1 - \alpha^{-2}k_n (k_nT_0)^{-1}\right)$. This means that the homogeneous distribution of PDs, starting from a certain critical rate of their formation, which is determined by temperature, dilatation volume, elastic modulus, and density of recombination centers, becomes unstable. A strain-induced current arises, leading to an increase in the areas of compression (for $\nu$-defects) or tension (for $i$-defects) of the concentration of defects, to their satiation and the formation of the corresponding clusters. The second inequality in (33) imposes a restriction on the spatial scale of inhomogeneities. If we accept that for the $\nu$-defects $|q| \approx 10^{-27} \text{cm}^{-1}$, $k = 5.10^{-11} \text{erg cm}^{-3}$ then $Q_{cr} = 10^{-2} \text{s}^{-1}$.

Another application of the proposed model is related to the formation of periodic spatial distributions of PDs in the process of solidification of thin layers on the substrates. In this case, the role of the strain field is played by the bending coordinate $\xi(x, y)$, which characterizes the displacement of the points of the median plane of the film along $z$-axis (the $z$-axis is perpendicular to the layer plane, the plane $z = 0$ coincides with the midplane of the layer). The strains $\varepsilon_f = (\nabla \mathbf{u})_f$ of von Karman type are $\varepsilon_f = (\nabla \mathbf{u})_f = \nu \left(z - \frac{h}{2}\right) \Delta \xi$, where $h$ is the layer thickness; $\nu = (1 - 2\sigma_x)/(1 - \sigma_y)$. It is assumed that the PD concentration rapidly adjusts to the $z$ axis strain distribution.

The equations of motion for the thin solid layers with defect-induced body forces $f_i = \partial_t \nabla n$ are given by $\sigma_{ij}^{(i)} + \partial_t \nabla n = 0$ ($\partial_x = \kappa \alpha_s$ is the PD deformation potential), which can be expressed in the following form by multiplying them by $zdz$ and integrating with respect to $z$:

$$M_{\alpha\beta, \alpha\beta} + f_{||} + \int_{-h/2}^{h/2} f_{\alpha\beta}dz + \int_{-h/2}^{h/2} f_{\alpha\beta}dz = 0,$$

where $M_{\alpha\beta} = \int_{-h/2}^{h/2} \sigma_{ij}^{(i)} z dz$ is the stress bending moment and $f_{||} = \sigma_{ij} \rho^{-1} \Delta \xi = c_s^2 \Delta \xi$ ($\sigma_{ij} = \sigma_{xx}$ is an axially symmetric stress in the film directed along the film and arising due to spatially inhomogeneous axially symmetric heating). The stress-strain constitutive relation for the bulk of the layer can be written as $\sigma_{ii}^{(i)} = \frac{1}{2} \left(\varepsilon_{ii} + \frac{1}{2} \varepsilon_{kk} \delta_{ll}\right)$. Young’s modulus ($E$) and Poisson’s ratio ($\nu$) are related to elastic Lame constants $\lambda$ and $G$ via $G = E/2(1+\nu)$ and $\lambda = E\nu/(1+\nu)(1-2\nu)$.

Using this expression we can obtain the following governing equation for the thin layer
\[
\frac{h^2 c^2}{12} \Delta^2 \xi - c_i^2 \Delta \xi = -\frac{1}{\rho h} \sum_{j \neq i} \partial_j \left( \int_{-b/2}^{b/2} (\partial n_j / \partial z) \, dz \right) - \nu \int_{-b/2}^{b/2} z \Delta n_j \, dz ,
\]

(34)

where \( c^2 = E \rho^{-1} (1 - \sigma^2)^{-1} \) is the layer rigidity. The right side of (34) takes into account the normal and tangential defect-induced bending forces, respectively, exerted on the layer. If \( \partial_j = 0 \) and surface heat effects are not considered, Eq. (34) reduces to the classical plate equation.

For the strain-induced flux of PDs on the layer surface we have expression

\[
\mathbf{j}_d^{(i)} = -n_d^{(0)} D_d \left( \nu \partial_d h / 2k_b T \right) \nabla (\Delta \xi).
\]

Then PD dynamic linearized equation can be written as

\[
\dot{n}_d = D_d \Delta n_d + D_d \frac{\partial_d v_{n_d}^{(0)}}{2k_b T} \Delta^2 \xi - \tau_d^{(i)} n_d - \gamma_d n_i.
\]

Taking into account the above assumptions, as well as neglecting the tangential forces in the layer the following dispersion equation of DEI can be obtained (for the 1D case)

\[
2 \Gamma_{12} \left( q^2 \right) = -\alpha_4 \pm \left( \alpha_4^2 - 4 \beta_4 \right)^{1/2},
\]

(35)

\[
\alpha_4 = \left( n_v^{(0)} + n_i^{(0)} \right) v_T + \frac{1}{\tau_i} + D_v q^2 \left( 1 - \frac{m_v n_v^{(0)}}{l_i^2 q^2 + 1} \right) + D_i q^2 \left( 1 - \frac{m_i n_i^{(0)}}{l_i^2 q^2 + 1} \right),
\]

\[
b_4 = \left[ D_v q^2 \left( 1 - \frac{m_v n_v^{(0)}}{l_i^2 q^2 + 1} \right) + \frac{1}{\tau_i} + \gamma n_i^{(0)} \right] \left[ D_i q^2 \left( 1 - \frac{m_i n_i^{(0)}}{l_i^2 q^2 + 1} \right) + \frac{1}{\tau_i} + \gamma n_i^{(0)} \right] - \\
\left( \gamma n_v^{(0)} + D_v q^2 \frac{m_v n_v^{(0)}}{l_i^2 q^2 + 1} \right) \left( \gamma n_i^{(0)} + D_i q^2 \frac{m_i n_i^{(0)}}{l_i^2 q^2 + 1} \right),
\]

\[
m_v = \nu \frac{\partial_n v_T}{2k_b T \rho c_v^2}, \quad m_i = \nu \left( \partial_n \right)^2, \quad l_i^2 = \frac{(hc)^2}{12 c_v^2},
\]

where \( n_d^{(0)} \) satisfy the equations \( Q_v = \gamma n_v^{(0)} n_i^{(0)} + \tau_i^{-1} n_i^{(0)} \), \( Q_v = \gamma n_v^{(0)} n_i^{(0)} + \tau_i^{-1} n_i^{(0)} \).

Consider the case when the parameters of both components of the defect subsystem coincide \( D_v = D_i = D, \quad \tau_i = \tau_v = \tau \quad n_v^{(0)} = n_i^{(0)} = n_0 \). Then we have from Eq. (35)

\[
\Gamma_{12} \left( q^2 \right) = \begin{cases} 
-D q^2 \left( 1 - \frac{2m n_0}{l_i^2 q^2 + 1} \right) - \tau^{-1} & , \\
-D q^2 - \tau^{-1} - 2\gamma n_0 & .
\end{cases}
\]

(36)

It follows from Eq. (36) that when the condition \( 2m n_0 > 1 \) is met, that is \( n_0 > n_c = k_b T \rho c_v^2 / \nu \partial_n \) \( 0.5n_{cr} \), there exists a range of wave-number values \( q_1^2 < q^2 < q_2^2 \) (values of...
critical fluctuations \((q_{1,2}^2)\) are determined from the condition \(\Gamma_i(q_{1,2}^2) = 0\) for which \(\Gamma_i > 0\), that is, the Fourier amplitudes of the perturbations increase indefinitely with time. As a result, instability arises in the PDs subsystem, the development of which leads to the formation of coupled periodic structures of PDs and bending deformation. The maximum value of the instability increment \(\Gamma_m\) is achieved when
\[
q^2 = q_m^2 = \left(\sqrt{m - 1}\right)l_0^2 \quad \text{or} \quad \Gamma_m = D \left(m + 1 - 2\sqrt{m}\right)l_0^2 - \tau^{-1},
\]
and the structure period is
\[
d_{lat} = 2\pi/q_m = 2\pi l_0 \left(\sqrt{m - 1}\right)^{1/2}.
\]

The reason for the occurrence of coupled concentration-bending deformation structures is the appearance of a strain-induced carrier drift due to interaction with the strain field \((m \neq 0)\) and the presence of a longitudinal stress (due to thermoelastic expansion) \(\sigma_{\parallel} \neq 0\) in the layer \((m \rightarrow 0)\), due to which the dependence \(\Gamma_i(q)\) has a maximum. In this case, against the background of homogeneous fluctuations \((q = 0)\), inhomogeneous fluctuations of the concentration of PDs with a wave-number \(q_m\) appear increasing with a rate \(\propto \Gamma_m\) several orders of magnitude higher than the rate of increase of homogeneous fluctuations in the subsystem of defects.

Note that the instability threshold is two times less compared with a system with one type of defects \((n_{cr})\) [14], which is due to the superposition of fluxes of \(i\) and \(\nu\)-defects. The absence in Eq. (36) of the mutual recombination coefficient \((\gamma_i)\) is due to the corresponding flux compensation.

In the general case, the instability region is determined by the conditions: with strong recombination \((4\gamma_iQ_{\tau_i} \tau_i \gg 1)\) and \(n_{cr}^{(0)}(m_n + m_i) > 2\) or \(n_{cr}^{(0)} > n_{cr}^{(0)} = 2k_B T \rho c_i^2/\nu_\parallel \left(\langle \theta_i \rangle^2 + |\langle \theta_i \rangle|\right)\), and with weak recombination \((4\gamma_iq_\tau_i \tau_i \ll 1)\): \(n_{cr}^{(0)} m_i > 1\).

Thus for the occurrence of DEI it is necessary that the PD concentration exceeds a critical value \(n_{cr}^{(0)} > n_{cr}^{(0)}\). The flexural strain, interacting with the PDs generated as the film grows, induces under certain conditions a PD drift directed counter to the ordinary diffusion. In this case the compression regions attract the \(\nu\)-defects, while the tension regions repel them. Becoming localized in the compression region, the \(\nu\)-defects themselves deform the film, enhancing thereby the initial strain fluctuations. The ensuing instability leads to formation, in the vacancy localization regions, of a high supersaturation sufficient for pore nuclei to appear and grow. In result, the adhesion of a layer to its substrate becomes weaker at the positions of such pores clusters and peeling off of the SLM layer can be observed.

For the characteristic values of the parameters \(|\alpha_n^{(\nu)}| = 10^{-23}\text{cm}^3\), \(k = 5.10^{11}\text{erg}\text{cm}^{-3}\), \(\beta = 10^{-5} \text{K}^{-1}\), \(T = 700 \text{K}\), may be estimated the critical concentration of defects: \(n_{cr}^{(\nu)} = 8.10^{18}\text{cm}^3\). Since, \(l_{\parallel} \approx \sqrt{10h} \approx 10^{-5}\text{cm} \quad (h = 5.10^{-6}\text{cm})\), \(E = 10^{11}\text{erg}\text{cm}^2\), \(\sigma_{\parallel} = 10^{6}\text{erg}\text{cm}^{-2}\), we obtain for \(m = 1 + \delta_\parallel\), where
\[
\delta_\parallel \approx 2q_m^2 l_0^2 = 8\pi^2 l_0^2 / d_{lat} \approx 10^{-2} \text{ at } d_{lat} = 3.10^{-3}\text{cm}.
\]
Then the maximum growth increment of the instability is:
\[
\Gamma_m \approx D\delta_\parallel^2 / 2l_0^2 \approx 0.5 \text{ s}^{-1} \text{ at } D = 10^{-6}\text{cm}^2\text{s}^{-1}.
\]
Consequently, the formation of a spatially periodic structure of \(\nu\)-defects with a scale \(d_{lat}\) occurs at times \(\tau_m \approx \Gamma_m^{-1} \approx 2\text{s}\).

6. Conclusions

In this article, the formulation of a conjugate temperature-diffusion-stress PFM has been presented for the microstructure evolution during SLM with the generation of PDs at the \((l-s)\) interface. The governing equations that follow from the fundamental balance laws involve the phase variable, the atomic displacement field, the defect concentration field and the temperature field with significant couplings.
between all equations. The model includes concentration-temperature-dependent eigen-strain, strain due to PT, strain dependency on PT and local mechanical equilibrium conditions.

The defect dynamics equations take into account the diffusion (ordinary and stress-induced drift) of PDs, their recombination (mutual, on neutral absorbers). The energy transfer equation includes contributions to the energy balance associated with dissipative effects generated by a change in the PD subsystem, as well as by the generation of latent heat due to phase changes. Some applications of the proposed coupled model of PD evolution have been discussed under the consideration on isothermal equilibrium situations. Phenomenon of diffusion-flexural instability with the PD clustering and the effect of this instability on the exfoliation of deposited thin layers on a substrate has been also studied.

The thermodynamically consistent approach used to derive control equations is characteristic not only for the solidification during SLM, but covers a wide class of problems (laser heating of solids, laser synthesis of thin films, etc.), in which thermal conductivity, diffusion of PDs, and elastic effects are related. In particularly, the proposed DEI model can be used to interpret the direct formation of micro- and nanostructures on the surface of a quartz optical glass by picosecond ultraviolet laser pulses [18].

The microstructure development during SLM is determined by the thermal history of parts, which is result of laser beam energy absorption by the powder particles and substrate, heat and mass transfer within the build part and heat losses. The resultant melt pool geometry and inhomogeneous temperature distribution and thermal history from the macroscopic thermo-hydrodynamic model can be used as the initial and boundary conditions for defect diffusion, which influences grain growth during SLM. Linking of microstructure evolution by considering the relationship of micro- and macroparameters, including laser and powder parameters such as laser power, scanning spot size and scan velocity will lead to a better understanding of different aspects of the microstructure evolution during the SLM.

7. References

[1] Mani M, Lane B M and Donmez M A 2016 Int. J. Prod. Res. 55(5) 1
[2] Huang Y, Leu M C, Mazumder J and Donmez A 2015 J. Manuf. Sci. Eng. 137 014001
[3] Yap C Y, Chua C K and Dong Z L 2015 Applied Physics Reviews 2 041101
[4] Cherry J A, Davies H M and Mehmoond S 2014 Int. J. Adv. Manuf. Technol. 76 869
[5] Khairallah S A, Anderson A T, Rubenchik A and King W E 2016 Acta Mater. 108 36
[6] Xia M, Gu D, Yu G, Dai D, Chen H and Shi Q 2016 Sci. Bull. 61(13)1013
[7] Martin A A, Calta N P, Khairallah S A 2019 Nat. Commun. 10 1987
[8] Li Y and Gu D 2014 Mater Des. 63 856
[9] Zang D Q, Cai Q Z 2010 Int. J. Adv. Manuf. Technol. 51(5-8) 649
[10] Panwisasawas C, Qiu Ch and Anderson M J 2017 Comput. Mater. Sci. 126 479
[11] Acharya R, Sharon J A and Staroselsky A 2017 Acta Mater. 124 360
[12] Mirzade F Kh and Dubrov A V Proc. SPIE 11057 110571Q
[13] Kobayashi R 1993 Physica D: Nonlinear Phenomena 63(3-4) 410
[14] Mirzade F Kh 1996 Phys. Usp. 39 1
[15] Wang S -L, Sekerka R F and Wheeler A A 1993 Physica D 69 189
[16] Gurtin M E 1996 Physica D 92 178
[17] Mirzade F Kh 2019 Proc. SPIE 11057 110571P
[18] Zavyalova M A 2016 Surface modification of silica glass by pulses of a picosecond laser Computer Optics 40(6) 863-870 DOI: 10.18287/2412-6179-2016-40-6-863-870

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