

# On a modified Becker-Döring model for two-phase materials

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## Abstract

This work reconsiders the Becker-Döring model for nucleation under isothermal conditions in the presence of phase transitions. Based on thermodynamic principles a modified model is derived where the condensation and evaporation rates may depend on the phase parameter. The existence and uniqueness of weak solutions to the proposed model are shown and the corresponding equilibrium states are characterized in terms of response functions and constitutive variables.

**Keywords:** Becker-Döring equations. Nucleation. Phase transitions. Reaction diffusion equations.

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## 1 Introduction

Nucleation phenomena occur in various applications of great technological importance. Here we only mention the formation of liquid droplets in semiconductors as GaAs, [10, 11], nucleation as final step of recrystallisation, [25], and the importance of nucleation in steels, e.g. due to cold rolling, accompanied by investigations of local stresses or the chemical composition of the material. Nucleation and growth phenomena play a key role on the morphology and macroscale properties of a variety of metallic structures, which include bulky materials, thin films and nanoparticles (refer, e.g., to [13] and references therein). The thermomechanical formation process of metallic polycrystals is strongly affected by the orientation of the crystal lattice in the individual grains, lattice curvature, dislocations, and the migration of grain boundaries [1]. The

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mathematical modeling of such phenomena needs to account for the formation, nucleation and growth of voids within the material through suitable constitutive parameters [28].

Becker and Döring introduced their model in 1935 to predict the formation of liquid droplets or bubbles in vapour in a stationary setup, [3]. At the heart of this model, droplets may grow or shrink solely by one mechanism, the attachment or detachment of one single molecule (monomer) or atom. The problem was reformulated by Frenkel [12]. Later, Burton [8] was the first to study the dynamical aspects of the system. Recent numerical studies on this subject can be found in [16] and references therein.

The metastability of the equations was studied in [2], [22]. The transition of the Becker-Döring model to the Lifshitz, Slyozov, and Wagner (LSW) model is studied in [19, 20], [23]. In [2], a Lyapunov functional for nucleation is introduced which is not in accordance to the second law of thermodynamics. This led to a corrected version of the standard model proposed by Dreyer and Duderstadt in [11]. In [14], the existence and uniqueness of this non-standard model were studied, where many results of [2] could be reused. A thorough survey on nucleation from the physical point of view is [17], where a discussion of the driving forces, heterogeneous nucleation models and an analysis of equilibrium states can be found, among others. The results of this article also have significant implications on the theory and the understanding of dynamic recrystallization (DRX). Typically, during DRX, new nuclei essentially free of dislocations are formed within a highly damaged material. Evidently, the lattice orders of nuclei and surrounding substrate are very different. DRX is of great technological importance, especially during the hot and cold rolling of industrial steels, and is still the subject of intense research. Here we only mention the recent articles [27, 29, 7, 26] and references therein.

In this article we investigate the implications of the second law of thermodynamics on the reaction scheme imposed by the Becker-Döring system. In order for the equations to fulfill the second law of thermodynamics, the reaction rates representing the condensation (attachment) and evaporation (detachment) of monomers have to depend on the order parameter  $\chi$ . The formalism goes back to [4], where the oxidation of a solid precipitate is formally modelled by chemical reactions.

The outline of this article is as follows. In Section 2 we introduce the new model and show the validity of the second law of thermodynamics. In particular, this allows us to study the dependence of the reaction rates on the order parameter in the generalized Becker-Döring scheme. In Section 3 we discuss special cases of the new model and compare with the non-standard Becker-Döring model introduced by Dreyer and Duderstadt. Section 4 is dedicated to the existence and uniqueness of weak solutions to the new model. Section 5 contains a short characterization of the equilibrium states. We end with some concluding remarks and an outlook.

## 2 Derivation of the model

We want to study nucleation in a two-phase material that is contained in  $\Omega \subset \mathbb{R}^d$ . The main application we have in mind is a solid such as a single crystal or a polycrystal with nucleating droplets. Throughout the text we shall assume that all nuclei have a perfect spherical shape.

For a given stop time  $0 < T < \infty$ , let  $\Omega_T := \Omega \times (0, T)$ . The shape of the two phases is determined by an order parameter  $\chi : \Omega_T \rightarrow [0, 1]$  which is an indicator function of one selected phase. This ansatz gives rise to a diffuse interface model with mushy regions. To simplify the thermodynamic reasoning, we assume that the temperature  $\theta$  is kept constant in  $\Omega_T$ .

Let  $Z_\alpha(x, t) \geq 0$  denote the number of nuclei of size  $\alpha$ ,  $\alpha \in \{1, 2, \dots, \nu\}$  at  $x \in \Omega$  and time  $0 \leq t \leq T$ , where  $\nu \in \mathbb{N} \cup \{+\infty\}$  specifies the largest occurring nucleus. We use the notations

$$Z \equiv Z(x, t) = (Z_\alpha(x, t))_{1 \leq \alpha \leq \nu} \equiv (Z_\alpha)_{1 \leq \alpha \leq \nu}$$

and drop the argument  $(x, t)$  when this is clear from the context.

In contrast to the original Becker-Döring model,  $Z$  may depend on the spatial position  $x$  which means that the function

$$\rho(Z(x, t)) := \sum_{\alpha=1}^{\nu} \alpha Z_\alpha(x, t)$$

may vary over  $\Omega$ . In the above context,  $Z_\alpha$  and  $\rho(Z)$  define the number density and the mass density of nucleating particles, respectively, with regard to a characteristic volume. The intuitive physical picture is that nucleation starts on a small microscopic length scale, whereas the phase profile determined by  $\chi$  is a macroscopic quantity.

The total number of nuclei is given by

$$N(Z(x, t)) = \sum_{\alpha=1}^{\nu} Z_\alpha(x, t), \quad (x, t) \in \Omega_T. \quad (2.1)$$

The time evolution of  $Z$  in  $\Omega_T$  is determined by the system of ordinary differential equations

$$\frac{d}{dt} Z_\alpha(x, t) = J_{\alpha-1}(Z(x, t), \chi(x, t)) - J_\alpha(Z(x, t), \chi(x, t)), \quad 1 \leq \alpha \leq \nu \quad (2.2)$$

with the initial condition

$$Z(\cdot, 0) = \tilde{Z} \quad \text{in } \Omega$$

and the fluxes

$$\begin{aligned} J_0(Z(x, t), \chi(x, t)) &= - \sum_{\alpha=1}^{\nu} J_\alpha(Z(x, t), \chi(x, t)), & (2.3) \\ J_\alpha(Z(x, t), \chi(x, t)) &:= \Gamma_\alpha^C(\chi(x, t)) Z_\alpha(x, t) - \Gamma_{\alpha+1}^E(\chi(x, t)) Z_{\alpha+1}(x, t) \\ &:= K(x) R_\alpha(\chi(x, t))^{1/b_\chi} Z_\alpha(x, t) \\ &\quad - R_{\alpha+1}(\chi(x, t))^{1/b_\chi} Z_{\alpha+1}(x, t), \quad 1 \leq \alpha \leq \nu. \end{aligned} \quad (2.4)$$

If  $v$  is finite, a further closedness condition is required. Here we only consider

$$\Gamma_v^C = \Gamma_{v+1}^E \equiv 0. \quad (2.5)$$

Other choices are discussed in [8], [22].

The equations (2.2)-(2.5) (completed with a governing equation for  $\chi$  below) are related to the classical Becker-Döring system, [3], but with rates that may additionally depend on  $x$  and on the phase parameter  $\chi$ . In the definition of  $J_\alpha$ , the functions  $\Gamma_\alpha^C(\chi) > 0, \Gamma_\alpha^E(\chi) > 0$  denote the *condensation* and the *evaporation* rates of a nucleus of size  $\alpha$ . In (2.4),  $K \in L^\infty(\Omega)$  is a given positive function, and

$$b_\chi := \chi b_1 + (1 - \chi)b_2 \quad (2.6)$$

for two positive constants  $b_l, l = 1, 2$  that appear in the definition (2.10) of the free energy of phase  $l$ . The particular form of  $\Gamma_\alpha^C, \Gamma_\alpha^E$  in (2.4) will be worked out later and can be justified a posteriori by thermodynamic considerations. Eqn. (2.3) ensures that  $\rho(Z)$  is conserved in  $\Omega_T$ .

The free energy  $F$  of the system is

$$F = F(Z, \chi) = \int_{\Omega} f(Z, \chi, \nabla \chi) dx$$

with the free energy density  $f = f(Z, \chi, \nabla \chi)$ . For  $f$  we make the ansatz

$$f(Z, \chi, \nabla \chi) = \chi f_1(Z) + (1 - \chi)f_2(Z) + \theta \left( W(\chi) + \frac{\gamma}{2} |\nabla \chi|^2 \right). \quad (2.7)$$

Here,  $f_l$  is the free energy density of phase  $l, l = 1, 2$  and the last term is due to the entropy of mixing. The scalar  $\gamma > 0$  determines the square root of the thickness of the boundary layer between the two phases (assumed constant here), and

$$W(\chi) := \chi \ln(\chi) + (1 - \chi) \ln(1 - \chi)$$

is a double well potential.

In order to formulate the governing equation for  $\chi$ , we need to smoothen the spatial variation of  $x \mapsto Z(x, t)$  in  $\Omega$ . To this end, we fix  $\varepsilon > 0$  and choose a function  $\varphi \in C^\infty(\mathbb{R}^d)$  with  $\varphi \geq 0$  and  $\int_{\mathbb{R}^d} \varphi(y) dy = 1$ . We regularize  $Z$  by the convolution

$$Z_\varepsilon(x, t) := (Z(\cdot, t) * \varphi_\varepsilon)(x) = \int_{\mathbb{R}^d} \varphi_\varepsilon(x - y) Z(y, t) dy \quad (2.8)$$

with the kernel  $\varphi_\varepsilon(x) := \varepsilon^{-d} \varphi(x/\varepsilon)$ . For the validity of (2.8),  $Z(\cdot, t) \in L^1(\Omega)$  is required. So we postulate for the initial value of (2.2)

$$\begin{aligned} \tilde{Z} &\in L^1(\Omega), \quad \rho(\tilde{Z}) > 0 \text{ in } \Omega, \\ \rho(\tilde{Z}) &\text{ is bounded uniformly in } \Omega, \\ \tilde{Z}_\alpha &\geq 0 \text{ in } \Omega \text{ for } \alpha \geq 1, \quad \tilde{Z}_1 > 0 \text{ in } \Omega. \end{aligned} \quad (A1)$$

For the time evolution of  $\chi$ , a variety of different laws may be used, as long as they are compatible with thermodynamics. Here we choose the Allen-Cahn type formula

$$\tau \partial_t \chi = - \frac{\partial F}{\partial \chi}(Z_\varepsilon, \chi), \quad (2.9)$$

where  $\tau = \tau(\theta)$  is a positive constant that adjusts the time scale of the propagation in  $\chi$ . The presence of  $Z_\varepsilon$  in Eqn. (2.9) states that the number of nuclei has to be integrated (summed) over a small spatial region of size  $\varepsilon$ . Hence,  $\varepsilon > 0$  introduces a length scale into the model.

In the definition (2.7), the functions  $\{f_l\}_{l=1,2}$  are smooth and convex and represent the specific free energy density of phase  $l$ . A convenient choice for reactive systems is

$$f_l(Z) := k_B \theta \sum_{\alpha=1}^{\nu} Z_\alpha \left[ b_l \ln \left( \frac{Z_\alpha}{N(Z)} \right) + \frac{E_\alpha^l}{k_B \theta} \right] \quad \text{in } \Omega_T, \quad l = 1, 2, \quad (2.10)$$

where  $k_B$  is the Boltzmann constant,  $E_\alpha^l > 0$  are enthalpic energy terms, and  $0 < b_l \leq 1$  are constants representing the local lattice geometry of phase  $l$ ,  $l = 1, 2$ . In case of  $b_l < 1$ , certain lattice sites are locked, e.g. by impurities, geometrically necessary dislocations, or other immobile constituents, see Figure 1. In [9], entropic terms of the form (2.10) are derived from lattice models.

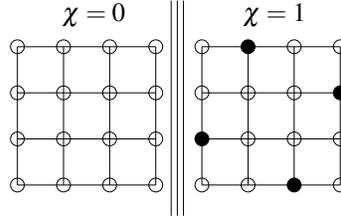


Figure 1: Schematic illustration of a phase transition with two different lattice geometries. Filled dots represent locked lattice sites. Left a rectangular lattice with  $b_1 = 1$ , right a rectangular lattice with  $b_2 = 0.75$ . The lines in the center represent the mushy region.

In the Becker-Döring model, the nucleation is modeled formally by reactions. The reaction rates and the constants  $E_\alpha^l$  are connected by the *Arrhenius law*

$$R_\alpha(\chi) = \exp \left( \frac{\chi E_\alpha^1 + (1 - \chi) E_\alpha^2 - E_A(\chi)}{k_B \theta} \right), \quad 1 \leq \alpha \leq \nu \quad (2.11)$$

for an activation or saddle point energy  $E_A(\chi)$  that has to be exceeded to start the nucleation.

Exploiting the Arrhenius law (2.11), with  $b_\chi$  as in (2.6), the free energy may be rewritten as

$$F(Z, \chi)(t) = \int_{\Omega} k_B \theta \sum_{\beta=1}^{\nu} Z_\beta \left[ b_\chi \ln \left( \frac{Z_\beta R_\beta^{1/b_\chi}}{N(Z)} \right) + \frac{E_A(\chi)}{k_B \theta} \right] + \theta \left( W(\chi) + \frac{\gamma}{2} |\nabla \chi|^2 \right) dx.$$

A direct computation yields

$$\begin{aligned}\frac{d}{dt}F(Z(t), \chi(t)) &= \sum_{\alpha=1}^{\nu} \frac{\partial F}{\partial Z_{\alpha}}(Z, \chi) \partial_t Z_{\alpha} + \frac{\partial F}{\partial \chi}(Z, \chi) \partial_t \chi \\ &= \int_{\Omega} k_B \theta \sum_{\alpha=1}^{\nu} \left[ b_{\chi} \ln \left( \frac{Z_{\alpha} R_{\alpha}^{1/b_{\chi}}}{N(Z)} \right) + \frac{E_A(\chi)}{k_B \theta} \right] \partial_t Z_{\alpha} dx - \frac{1}{\tau} \left( \frac{\partial F}{\partial \chi}(Z, \chi) \right)^2.\end{aligned}$$

Here we used that for any  $1 \leq \alpha \leq \nu$

$$\sum_{\beta=1}^{\nu} Z_{\beta} \frac{\partial}{\partial Z_{\alpha}} \left[ \ln \left( \frac{Z_{\beta} R_{\beta}^{1/b_{\chi}}}{N(Z)} \right) \right] = 0.$$

If we resolve  $\partial_t Z_{\alpha}(x, t)$  by the evolution law (2.2), this becomes

$$\begin{aligned}\frac{d}{dt}F(Z(t), \chi(t)) &= \int_{\Omega} k_B \theta \left\{ \left( - \sum_{\alpha=1}^{\nu} J_{\alpha}(Z, \chi) - J_1(Z, \chi) \right) \left[ b_{\chi} \ln \left( \frac{Z_1 R_1^{1/b_{\chi}}}{N(Z)} \right) + \frac{E_A(\chi)}{k_B \theta} \right] \right. \\ &\quad \left. + \sum_{\alpha=2}^{\nu} (J_{\alpha-1}(Z, \chi) - J_{\alpha}(Z, \chi)) \left[ b_{\chi} \ln \left( \frac{Z_{\alpha} R_{\alpha}^{1/b_{\chi}}}{N(Z)} \right) + \frac{E_A(\chi)}{k_B \theta} \right] \right\} dx \\ &\quad - \frac{1}{\tau} \left( \frac{\partial F}{\partial \chi}(Z, \chi) \right)^2 \\ &= \int_{\Omega} k_B \theta \sum_{\alpha=1}^{\nu} J_{\alpha}(Z, \chi) \left[ b_{\chi} \ln \left( \frac{Z_{\alpha+1} R_{\alpha+1}^{1/b_{\chi}}}{Z_{\alpha} R_{\alpha}^{1/b_{\chi}}} \frac{N(Z)}{Z_1 R_1^{1/b_{\chi}}} \right) - \frac{E_A(\chi)}{k_B \theta} \right] dx \\ &\quad - \frac{1}{\tau} \left( \frac{\partial F}{\partial \chi}(Z, \chi) \right)^2.\end{aligned}$$

In glance of the thermodynamic structure of reactive systems, this motivates to set

$$\frac{1}{K} \frac{Z_1 R_1^{1/b_{\chi}}}{N(Z)} = \exp \left( \frac{-E_A(\chi)/b_{\chi}}{k_B \theta} \right) \quad \text{in } \Omega_T. \quad (2.12)$$

For the existence proof of Section 4 we postulate the following two conditions:

For each  $1 \leq \alpha \leq \nu$  and every  $(x, t) \in \Omega_T$  there exists  $0 < \gamma_{\alpha}(x, t) < \infty$  such that

$$\begin{aligned}\max \left\{ R_{\alpha}(\chi(x, t))^{1/b_1}, K(x) R_{\alpha}(\chi(x, t))^{1/b_1}, \right. \\ \left. R_{\alpha}(\chi(x, t))^{1/b_2}, K(x) R_{\alpha}(\chi(x, t))^{1/b_2} \right\} \leq \gamma_{\alpha}(x, t)\end{aligned} \quad (A2)$$

and for every  $(x, t) \in \Omega_T$  it holds

$$\gamma_{\alpha+1}(x, t) \leq \gamma_{\alpha}(x, t), \quad \lim_{\alpha \rightarrow \infty} \frac{\gamma_{\alpha}(x, t)}{\alpha} = 0. \quad (A3)$$

With (2.12) and (2.4), the final form of the free energy inequality is

$$\begin{aligned} \frac{d}{dt}F(Z(t), \chi(t)) &= \int_{\Omega} k_B \theta \sum_{\alpha=1}^{\nu} \left( KR_{\alpha}(\chi)^{1/b_{\chi}} Z_{\alpha} - R_{\alpha+1}(\chi)^{1/b_{\chi}} Z_{\alpha+1} \right) \\ &\quad \times b_{\chi} \ln \left( \frac{R_{\alpha+1}(\chi)^{1/b_{\chi}} Z_{\alpha+1}}{KR_{\alpha}(\chi)^{1/b_{\chi}} Z_{\alpha}} \right) dx - \frac{1}{\tau} \left( \frac{\partial F}{\partial \chi}(Z, \chi) \right)^2. \end{aligned} \quad (2.13)$$

Equality (2.13) immediately implies the validity of the second law of thermodynamics, as in general  $(B-A) \ln(A/B) \leq 0$  for arbitrary  $A > 0$  and  $B > 0$ . So we unconditionally infer  $\frac{d}{dt}F(Z(t), \chi(t)) \leq 0$  as desired. Eqn. (2.13) also justifies the ansatz (2.4).

### 3 The relationship to the non-standard Becker-Döring model

In the special case of  $b_1 = b_2 = 1$ , the system (2.2)-(2.5) corresponds to the non-standard Becker-Döring model by Dreyer and Duderstadt, [11], that in contrast to the original Becker-Döring system satisfies the laws of thermodynamics.

The condition  $b_1 = b_2 = 1$  generically holds for liquids and gases, but is also fulfilled in solids if all lattice sites are freely accessible to nucleation. In this case, we may choose  $\Gamma_{\alpha}^C, \Gamma_{\alpha+1}^E$  independent of  $\chi$  to obtain  $J_{\alpha} = J_{\alpha}(Z)$  for

$$J_{\alpha}(Z) = \Gamma_{\alpha}^C(\cdot)Z_{\alpha} - \Gamma_{\alpha+1}^E(\cdot)Z_{\alpha+1} \quad \text{in } \Omega_T.$$

The equations (2.2)-(2.5) decouple from  $\chi$  and form a system of ordinary differential equations for the family of unknowns  $(Z_{\alpha}(x, t))_{1 \leq \alpha \leq \nu}$  parameterized by  $x \in \Omega$ .

The traditional Becker-Döring model uses constant condensation and evaporation rates,

$$\Gamma_{\alpha}^C(x, t) \equiv \Gamma_{\alpha}^C > 0, \quad \Gamma_{\alpha+1}^E(x, t) \equiv \Gamma_{\alpha+1}^E > 0. \quad (3.1)$$

This constitutive assumption makes the number of nuclei independent of  $x$ , thus  $Z(t) = (Z_{\alpha}(t))_{1 \leq \alpha \leq \nu}$ . The equations (2.2)-(2.5) then coincide with the Becker-Döring system, with

$$\Gamma_{\alpha}^C = K\Gamma_{\alpha}^E \quad \text{for } 1 \leq \alpha \leq \nu,$$

which is a consequence of (2.4).

With these simplifications, the approach (2.10) relates to the choice

$$f(Z(t)) = k_B \theta \sum_{\alpha=1}^{\nu} Z_{\alpha}(t) \ln \left( \frac{Z_{\alpha}(t)}{q_{\alpha}(t)N(Z(t))} \right)$$

derived by Dreyer and Duderstadt in [11], where  $q_{\alpha} = \exp(-\frac{E_{\alpha}}{k_B \theta})$ . This is analogous to the formula for  $R_{\alpha}$  in (2.11) if the activation energy  $E_A$  is neglected. The condition (2.12) determines  $\Gamma_{\alpha}^C/\Gamma_{\alpha}^E$ , whereas the condition

$$\frac{N(Z(t))q_{\alpha}q_1}{Z_1(t)q_{\alpha+1}} = \frac{\Gamma_{\alpha+1}^E}{\Gamma_{\alpha}^C}$$

chosen in [11] to guarantee the thermodynamic correctness of the modified Becker-Döring system determines the ratio  $\Gamma_\alpha^C/\Gamma_{\alpha+1}^E$ .

The ansatz (2.4) is general enough to cover any of the commonly used heuristic formulas for the evaporation and condensation rates like

$$\Gamma_\alpha^C(t) = \alpha^A Z_1(t), \quad \Gamma_\alpha^E(t) = \alpha^A \left( C + \frac{D}{\alpha^B} \right)$$

with constants  $0 \leq A < 1$ ,  $0 < B < 1$ ,  $C > 0$ ,  $D > 0$ .

Finally we mention that a formalism similar to (2.10) is used in [4], where the thermodynamics of reactions accompanied by phase transitions were studied and estimates similar to (2.13) are found. General reaction schemes and their asymptotic limits have also been studied in [5], where the reactions formally model the generation and annihilation of vacancies in a solid due to plastic effects accompanied by moving reconstitutive transition layers.

## 4 Existence and uniqueness of weak solutions

We proof existence and uniqueness of weak solutions to (2.2)-(2.4), (2.9) for the most general case  $\nu = \infty$ . By  $C^k(I; S)$  we denote the space of  $k$ -times continuously differentiable functions from an interval  $I \subset \mathbb{R}$  to a set  $S$  and  $H^{m,2}(\Omega)$  denotes the Sobolev space of  $m$ -times weakly differentiable functions in the Hilbert space  $L^2(\Omega)$ .

The first step is to decouple (2.2)-(2.4) and slice the solution  $Z$  in the  $x$ -variable. This allows to apply the methods and results of [2], [14] for the Becker-Döring system. For fixed  $\bar{x} \in \Omega$  and given  $\bar{\chi}(\bar{x}, \cdot) \in C^0([0, T]; [0, 1])$  we introduce the solution vector

$$z(t) \equiv (z_\alpha(t))_{\alpha \in \mathbb{N}} \equiv z_{\bar{x}}(t) := Z(\bar{x}, t).$$

For the sliced system we seek solutions  $t \rightarrow z_{\bar{x}}(t)$  in  $C^0([0, T]; X)$  with

$$X := \left\{ (z_\alpha)_{\alpha \in \mathbb{N}} \mid \|z\|_X < \infty \right\}, \quad \|z\|_X := \sum_{\alpha=1}^{\infty} \alpha |z_\alpha|.$$

We introduce the symbols

$$\begin{aligned} j_\alpha(z(t)) &:= J_\alpha(Z(\bar{x}, t), \bar{\chi}(\bar{x}, t)), \\ r_\alpha(t) &:= R_\alpha(\bar{\chi}(\bar{x}, t)), \\ k &:= K(\bar{x}), \\ b(t) &:= \bar{\chi}(\bar{x}, t)b_1 + (1 - \bar{\chi}(\bar{x}, t))b_2 \geq \min\{b_1, b_2\} =: b_0 > 0. \end{aligned}$$

The system (2.2)-(2.4) becomes

$$\frac{d}{dt} z_\alpha(t) = j_{\alpha-1}(z(t)) - j_\alpha(z(t)), \quad \alpha \geq 1, \quad (4.1)$$

$$j_0(z(t)) = - \sum_{\alpha=1}^{\infty} j_\alpha(z(t)), \quad (4.2)$$

$$j_\alpha(z(t)) = k r_\alpha(t) z_\alpha(t) - r_{\alpha+1}(t) z_{\alpha+1}(t), \quad \alpha \geq 1. \quad (4.3)$$

As initial conditions we impose

$$z(t=0) = \tilde{z}(0) \equiv \tilde{z}_{\bar{x}}(0) := \tilde{Z}(\bar{x}) = Z(\bar{x}, 0).$$

Assumption (A1) implies  $\rho_0 = \rho(\tilde{z}) > 0$  and  $\tilde{z}_\alpha \geq 0$  for all  $\alpha \in \mathbb{N}$  and  $\tilde{z}_1 > 0$ .

With the above notations, the free energy density  $f$  of the sliced system becomes

$$f(z)(t) := k_B \theta \sum_{\alpha=1}^{\infty} z_\alpha(t) \left[ b(t) \ln \left( \frac{z_\alpha(t) r_\alpha^{1/b(t)}}{N(z(t))} \right) + \frac{e_A(t)}{k_B \theta} \right] + s_M,$$

where  $e_A(t) := E_A(\bar{\chi}(\bar{x}, t))$  and  $s_M$  is an integrating entropic constant.

Similar to [2], [14] we can show:

**Proposition 4.1.** *Let the assumptions (A2), (A3) hold. Then there exists a function  $z(t) = z_{\bar{x}}(t) \in C^0([0, T]; X)$  which is the unique weak solution of*

$$\begin{aligned} \frac{d}{dt} z_\alpha(t) &= j_{\alpha-1}(z(t)) - j_\alpha(z(t)), \quad \alpha \geq 2, \\ \frac{d}{dt} N_\alpha(z(t)) &= j_{\alpha-1}(z(t)), \quad \alpha \geq 2, \\ z_1(t_2) - z_1(t_1) &= \int_{t_1}^{t_2} j_0(z(t)) dt, \quad 0 \leq t_1, t_2 < T. \end{aligned} \quad (4.4)$$

Additionally it holds  $z_\alpha(t) \geq 0$  for all  $\alpha \geq 1$ ,  $t \in [0, T)$ , and

$$\rho(z(t)) = \rho(z(0)) = \rho_0, \quad 0 \leq t < T, \quad (4.5)$$

and for  $0 \leq t_1 \leq t_2 < T$  and a positive constant  $C = C(\theta)$

$$f(z(t_1)) - f(z(t_2)) \geq \frac{C}{\rho_0} \int_{t_1}^{t_2} \sum_{\alpha=1}^{\infty} |j_\alpha(z(t))|^2 dt \geq 0.$$

**Remark.** Due to (4.4),  $z$  is the *weak* and not the strong solution to (4.1)-(4.3).

The proof of Proposition 4.1 is divided into several steps. The equations (4.1)-(4.3) are approximated by a finite system of dimension  $n$  that results from the infinite system by neglecting all nuclei of size greater than  $n$ . So we consider

$$\frac{d}{dt} z_\alpha^{(n)}(t) = j_{\alpha-1}(z^{(n)}(t)) - j_\alpha(z^{(n)}(t)), \quad 2 \leq \alpha \leq n-1, \quad (4.6)$$

$$\frac{d}{dt} z_1^{(n)}(t) = -j_1(z^{(n)}(t)) - \sum_{\alpha=1}^{n-1} j_\alpha(z^{(n)}(t)), \quad (4.7)$$

$$\frac{d}{dt} z_n^{(n)}(t) = j_{n-1}(z^{(n)}(t)) \quad (4.8)$$

completed with the initial conditions

$$z_\alpha^{(n)}(0) = \tilde{z}_\alpha^{(n)} \quad \text{for } 1 \leq \alpha \leq n.$$

It holds  $\tilde{z}^{(n)} \rightarrow \tilde{z}$  in  $X$  for  $n \rightarrow \infty$ , where  $\tilde{z}$  is the initial datum of the infinite system.

On  $X$  we define weakstar-continuous functionals  $N_\alpha(z) := \sum_{\beta=\alpha}^{\infty} z_\beta$ . It clearly holds  $N(z) = N_1(z)$  and  $z_\alpha(t) = N_\alpha(z(t)) - N_{\alpha+1}(z(t))$ . One can formally show

$$\begin{aligned}\rho(z(t)) &= \sum_{\alpha=1}^{\infty} N_\alpha(z(t)), \\ \frac{d}{dt} N_\alpha(z(t)) &= j_{\alpha-1}(z(t)), \quad \alpha \geq 1.\end{aligned}$$

**Lemma 4.1.** *Let (A2), (A3) be satisfied. Then the following statements hold.*

- (i) For all  $n \in \mathbb{N}$  there exists a solution  $z^{(n)} \in C^\infty([0, T]; X)$  to (4.6)-(4.8).
- (ii) With  $N_\alpha^{(n)} := N_\alpha(z^{(n)})$ ,  $j_\alpha^{(n)} := j_\alpha(z^{(n)})$ ,  $\rho^{(n)} := \rho(z^{(n)})$ ,  $f^{(n)} := f(z^{(n)})$ , the following statements are valid:
  - (1)  $\rho^{(n)}(t) = \rho^{(n)}(0)$ .
  - (2)  $\frac{d}{dt} N_\alpha^{(n)} = j_{\alpha-1}^{(n)}(t)$  for all  $0 \leq t < T$  and all  $1 \leq \alpha \leq n$ .
  - (3) There exists a constant  $C = C(\theta) > 0$  independent of  $n$  such that

$$f^{(n)}(t_1) - f^{(n)}(t_2) \geq \frac{C}{\rho^{(n)}} \int_{t_1}^{t_2} \sum_{\alpha=1}^{n-1} |j_\alpha^{(n)}(t)|^2 dt \quad (4.9)$$

for  $0 \leq t_1 \leq t_2 < T$ .

- Proof.* (i) This follows from the Picard-Lindelöf theorem.  
(ii) We show only (4.9). The proof of the other statements is similar.

A calculation as in (2.13) yields

$$\frac{d}{dt} f(z^{(n)})(t) = -k_B \theta b(t) \sum_{\alpha=1}^{n-1} (c_\alpha - d_\alpha) (\ln(c_\alpha) - \ln(d_\alpha))$$

with  $c_\alpha := r_{\alpha+1}^{1/b(t)} z_{\alpha+1}^{(n)}(t)$ ,  $d_\alpha := k r_\alpha^{1/b(t)} z_\alpha^{(n)}(t)$ .

Let  $\gamma_\alpha = \gamma_\alpha(\bar{x}, t)$ . It holds  $z_\alpha^{(n)} \leq \rho^{(n)}/\alpha$  and with the assumptions (A2), (A3) we find  $\max\{c_\alpha, d_\alpha\} \leq \rho^{(n)}\gamma_\alpha/\alpha$  and therefore

$$\begin{aligned}k_B \theta b(t) (c_\alpha - d_\alpha) (\ln(c_\alpha) - \ln(d_\alpha)) &\geq k_B \theta b_0 \frac{(c_\alpha - d_\alpha)^2}{\rho^{(n)}} \frac{\alpha}{\gamma_\alpha} \\ &\geq \frac{k_B \theta b_0 C_1}{\rho^{(n)}} |j_\alpha^{(n)}|^2.\end{aligned}$$

In the last line we used (A3) which implies  $\alpha/\gamma_\alpha \geq C_1 > 0$ . Setting  $C := k_B \theta b_0 C_1$ , the proof of (4.9) is complete.  $\square$

**Lemma 4.2.** *The following functions introduced in Lemma 4.1 are uniformly, i.e. independently of  $n$ , bounded in  $C^0([0, T])$ .*

- (1)  $z_\alpha^{(n)}, N_\alpha^{(n)}, j_\alpha^{(n)}, \dot{z}_\alpha^{(n)}, \dot{N}_\alpha^{(n)}$  for  $1 \leq \alpha \leq n$ .
- (2)  $\dot{z}_\alpha^{(n)}, \dot{N}_\alpha^{(n)}, j_\alpha^{(n)}$  for  $2 \leq \alpha \leq n$ .
- (3)  $j_0^{(n)}, j_1^{(n)}$ .

*Proof.* We demonstrate only (1). We assume for simplicity  $z_\alpha^{(n)} > 0$  for all  $\alpha$ . By direct computations we find

$$\begin{aligned}
\frac{d}{dt} \sum_{\alpha=1}^n \alpha z_\alpha^{(n)}(t) &= \sum_{\alpha=1}^n \alpha \dot{z}_\alpha^{(n)}(t) \\
&= - \sum_{\alpha=1}^n j_\alpha^{(n)}(t) - j_1^{(n)}(t) + \sum_{\alpha=2}^n \alpha (j_{\alpha-1}^{(n)}(t) - j_\alpha^{(n)}(t)) \\
&= - \sum_{\alpha=1}^n j_\alpha^{(n)}(t) - j_1^{(n)}(t) + \sum_{\alpha=2}^n j_\alpha^{(n)}(t) + 2j_1^{(n)}(t) \\
&= 0.
\end{aligned} \tag{4.10}$$

After integrating (4.10) w.r.t.  $t$ , we get

$$z_\alpha^{(n)}(t) \leq \sum_{\alpha=1}^n \alpha z_\alpha^{(n)}(t) = \sum_{\alpha=1}^n \alpha z_\alpha^{(n)}(0) = \sum_{\alpha=1}^n \alpha \tilde{z}_\alpha.$$

The term on the right is bounded independently of  $n$ .

The bounds on  $\dot{z}_\alpha^{(n)}(t)$  can be derived directly,

$$\begin{aligned}
|\dot{z}_\alpha^{(n)}(t)| &= |j_{\alpha-1}^{(n)}(t) - j_\alpha^{(n)}(t)| \leq |j_{\alpha-1}^{(n)}(t)| + |j_\alpha^{(n)}(t)|, \quad 2 \leq \alpha \leq n, \\
|\dot{z}_1^{(n)}(t)| &= \left| -j_1^{(n)}(t) - \sum_{\alpha=1}^n j_\alpha^{(n)}(t) \right| \leq |j_1^{(n)}(t)| + \left| \sum_{\alpha=1}^n j_\alpha^{(n)}(t) \right|.
\end{aligned}$$

The bounds on  $j_\alpha^{(n)}(t)$  can be derived as in Lemma 4.1. □

The uniform bounds of Lemma 4.2 as a consequence of the Arzelà-Ascoli theorem permit to pass to the limit  $n \rightarrow \infty$ . The solution  $z$  in Proposition 4.1 is the limit of  $(z^{(n)})_{n \in \mathbb{N}}$ .

The uniqueness of  $z$  follows from a Gronwall estimate and the conservation of mass (4.5) as outlined in [14]. This completes the proof of Proposition 4.1. □

**Proposition 4.2.** *Under the assumption (A1) there exists a unique solution  $\chi$  to the regularized Allen-Cahn equation (2.9) that satisfies*

- (i)  $\chi \in C^{0, \frac{1}{4}}([0, T]; L^2(\Omega))$ ,
- (ii)  $\partial_t \chi \in L^2(\Omega_T)$ ,
- (iii)  $\ln(\chi) \in L^1(\Omega_T)$  and  $0 < \chi < 1$  almost everywhere.

*Proof.* The statements (i)-(iii) follow from well-established existence and uniqueness results of the Allen-Cahn equation, see for instance [6], provided we can show that the convolution (2.8) is well defined, i.e. if

$$Z(\cdot, t) \in L^1(\Omega) \quad \text{for } 0 \leq t < T.$$

For  $t = 0$ , this follows from (A1). For  $t > 0$ , it is sufficient to show that there exists a function  $g \in L^1(\Omega)$  such that  $Z(\cdot, t)$  is measurable and

$$|Z_\alpha(x, t)| \leq g(x) \quad \text{for almost every } x \text{ in } \Omega$$

and any  $\alpha \geq 1$ . But the last follows from (4.5) and (A1), since

$$Z_\alpha(x, t) \leq \rho(z_x(t)) = \rho(\tilde{z}_x) = \rho(\tilde{Z}(x)) \leq C.$$

As it is evident that  $Z(\cdot, t)$  is measurable, the proof is complete.  $\square$

With the Propositions 4.1 and 4.2, the subsequent theorem is now evident.

**Theorem 4.3.** *Let the assumptions (A1)-(A3) be fulfilled. Then the system (2.2)-(2.9) possesses a unique weak solution  $(Z, \chi)$ , where  $Z(x, \cdot) \in C^0([0, T]; X)$  for almost every  $x \in \Omega$  fulfills the properties stated in Proposition 4.1 and where  $\chi \in C^0([0, T]; L^2(\Omega))$  satisfies the properties stated in Proposition 4.2.*

## 5 Characterization of the equilibrium states

In this section we characterize the equilibrium states  $(\bar{Z}, \bar{\chi})$ . Stationarity in  $\chi$  requires

$$\frac{\partial F}{\partial \chi}(\bar{Z}, \bar{\chi}) = 0$$

and due to the gradient term  $\frac{\gamma}{2} |\nabla \chi|^2$  in  $F$  this yields  $\bar{\chi} \equiv \text{const}$  in  $\Omega$ . This in turn shows  $\bar{Z}_\alpha \equiv \text{const}$  in  $\Omega$  for every  $\alpha$ . Clearly,  $(\bar{Z} \equiv 0, \bar{\chi})$  is always an equilibrium state. For  $\bar{Z}$  not completely vanishing, we prescribe the local mass  $\bar{\rho} > 0$  and want to prove that there exists an equilibrium state with  $\bar{\rho}(\bar{Z}) = \bar{\rho} > 0$ . Stationarity in  $Z$  implies  $J_\alpha(\bar{Z}, \bar{\chi}) = 0$ . With (2.4) we find

$$\bar{K} \Gamma_\alpha^E \bar{Z}_\alpha = \Gamma_{\alpha+1}^E \bar{Z}_{\alpha+1}, \quad (5.1)$$

where  $K(x) \equiv \bar{K}$  in  $\Omega$  is assumed.

The rates  $\Gamma_\alpha^E, \Gamma_\alpha^C$  are always positive numbers. Here we make the stronger assumption that they are bounded away from zero uniformly in  $\alpha$ , i.e. for a constant  $c_0 > 0$

$$\Gamma_\alpha^E(\bar{\chi}) = R_\alpha^E(\bar{\chi}) \geq c_0 \quad \text{for all } \alpha \in \mathbb{N}. \quad (A4)$$

Iterating (5.1) yields

$$\bar{Z}_\alpha = \bar{N}_s (\Gamma_\alpha^E)^{-1} \bar{K}^\alpha. \quad (5.2)$$

Here,  $s = s(\bar{\chi}) = \exp(-E_A(\bar{\chi}) / (k_B \theta b \bar{\chi}))$ , since by (2.12) it holds  $\bar{Z}_1 \Gamma_1^E(\bar{\chi}) = \bar{N} K s(\bar{\chi})$ .

From  $\bar{N}(\bar{Z}) = \bar{N}$ ,  $\rho(\bar{Z}) = \bar{\rho}$  we obtain

$$\bar{N}s \sum_{\alpha=1}^{\infty} (\Gamma_{\alpha}^E(\bar{\chi}))^{-1} \bar{K}^{\alpha} = \bar{N}, \quad \bar{N}s \sum_{\alpha=1}^{\infty} \alpha (\Gamma_{\alpha}^E)^{-1} \bar{K}^{\alpha} = \bar{\rho}.$$

In order to derive a condition on  $\bar{K}$ , this is re-written in the form

$$\tilde{f}(\bar{K}) = 1, \quad \bar{N} = \frac{\bar{\rho}}{\tilde{g}(\bar{K})}$$

with

$$\tilde{f}(K) := s \sum_{\alpha=1}^{\infty} (\Gamma_{\alpha}^E)^{-1} K^{\alpha}, \quad \tilde{g}(K) := s \sum_{\alpha=1}^{\infty} \alpha (\Gamma_{\alpha}^E)^{-1} K^{\alpha}.$$

Assumption (A4) ensures the convergence of both series with radius of convergence 1. The function  $\tilde{f}$  is continuous, strictly increasing in  $[0, 1]$ , and fulfills  $\tilde{f}(K) \geq s(\Gamma_1^E)^{-1}K$ . Therefore  $\bar{K} \in [0, \min\{1, \Gamma_1^E/s\}]$  if and only if  $\tilde{f}(1) \geq 1$ . To fulfill the second condition  $\rho(\bar{Z}) = \bar{\rho} > 0$ , we require  $\tilde{g}(\bar{K}) < \infty$ . But from  $\tilde{f}(1) > 1$  we infer  $\bar{K} < 1$  and thus  $\tilde{g}(\bar{K}) < \infty$ . So we arrive at the following sufficient condition of an equilibrium state:

$$\tilde{f}(1) > 1, \quad \text{or } (\tilde{f}(1) = 1 \text{ and } \tilde{g}(1) < \infty). \quad (EQ)$$

**Proposition 5.1.** *Let (EQ) and Assumption (A4) hold. Then, for any  $\bar{\rho} > 0$ , there exists an equilibrium state  $(\bar{Z}, \bar{\chi})$  of (2.2)–(2.4), (2.9) with  $\rho(\bar{Z}) = \bar{\rho}$ . Furthermore, (a)  $(\bar{Z}_{\alpha})_{\alpha \geq 1}$  and  $\bar{\chi}$  are constant functions in  $\Omega$  which satisfy*

$$\begin{aligned} \frac{\partial F}{\partial \chi}(\bar{Z}, \bar{\chi}) &= 0, \\ J_{\alpha}(\bar{Z}, \bar{\chi}) &= 0 \quad \text{for } \alpha \geq 1. \end{aligned}$$

(b) *The function  $K$  fulfills  $K \equiv \bar{K}$  in  $\Omega$  with a unique constant  $\bar{K} \in (0, 1]$  such that*

$$\tilde{f}(\bar{K}) = 1.$$

(c)  *$\bar{Z}_{\alpha}$  is given by (5.2) for all  $\alpha \in \mathbb{N}$ , i.e.  $\bar{Z}_{\alpha} = \bar{N}s(\Gamma_{\alpha}^E)^{-1}\bar{K}^{\alpha}$ , where  $\bar{N} = N(\bar{Z}) = \bar{\rho}/\tilde{g}(\bar{K})$ .*

## 6 Concluding remarks

In this article we derived a generalized Becker-Döring model for nucleation in the presence of phase transitions which respects the second law of thermodynamics, as opposed to the original formulation [3]. A key feature of the proposed model relies on the prediction that condensation and evaporation rates depend on the phase parameter  $\chi$ . The mathematical formulation of this model led us to prove existence and uniqueness of weak solutions, and to characterize the equilibrium states of the system.

A limitation of the developed theory, as for the Becker-Döring equations in general, is that it is a system of *ordinary* differential equations. In addition, the dependence on

the spatial coordinate  $x$  is not known. We plan to further investigate this issue through future work, which will also be aimed at numerically studying the dependence on the parameters and their spatial variation in more detail. Such a study will employ both deterministic and statistical approaches, and will be focused on the analysis of time-dependent properties and metastability, since available literature results for Becker-Döring models are still incomplete in terms of convergence rates, coarsening effects and evolution of large clusters as time goes to infinity (refer, e.g., to [15] and references therein).

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## References

- [1] Ask, A. and Forest, S. and Appolaire, B. and Ammar, K.: A Cosserat phase-field theory of crystal plasticity and grain boundary migration at finite deformation. *Continuum Mech Therm*, 1–33 (2018)
- [2] Ball, J.M., Carr, J., Penrose, O.: The Becker-Döring cluster equations: Basic properties and asymptotic behaviour of solutions. *Comm. Math. Phys.* **104**, 657–692 (1986)
- [3] Becker, R., Döring, W.: Kinetische Behandlung der Keimbildung in übersättigten Dämpfen. *Annalen der Physik* **24**, 719–752 (1935)
- [4] Blesgen, T.: A Revised Model for Diffusion Induced Segregation Processes. *Journal of Mathematical Physics* **46**, 022702 (2005)
- [5] Blesgen, T., Luckhaus, S.: On the Role of Lattice Defects close to Phase Transitions. *Math. Meth. Appl. Sci.* **29**, 525–536 (2006)
- [6] Blesgen, T., Weikard, U.: Multi-Component Allen-Cahn Equation For Elastically Stressed Solids. *Electron. J. Diff. Equ.* **89**, 1–17 (2005)
- [7] Blesgen, T.: A variational model for dynamic recrystallization based on Cosserat plasticity, *Composites B: Engineering* **115**, 236–243 (2017)
- [8] Burton, J.J.: Nucleation theory, In: Berne B.J. (eds) *Statistical Mechanics, Modern Theoretical Chemistry Vol.5*, Springer Boston, 195–234 (1977)
- [9] De Masi, A., Dirr, N., Presutti, E.: Interface Instability under Forced Displacements. *Ann. Henri Poincaré* **7**, 471–511 (2006)

- [10] Dreyer, W., Duderstadt, F.: Towards the thermodynamic modeling of nucleation and growth of liquid droplets in single crystals. *Internat. Ser. Num. Math.* **147**, 113–130 (2004)
- [11] Dreyer, W., Duderstadt, F.: On the Becker/Döring theory of nucleation of liquid droplets in solids. *J. Stat. Phys.* **123**, 55–87 (2006)
- [12] Frenkel, J.I.: A general theory of heterogeneous fluctuations and pretransition phenomena. *J. Chem. Phys.* **7**, 538–547 (1939)
- [13] Haumesser, P.-H.: *Nucleation and Growth of Metals: From Thin Films to Nanoparticles*. Elsevier, 1–194 (2016)
- [14] Herrmann, M., Naldzhieva, M., Niethammer B.: On a thermodynamically consistent modification of the Becker-Döring equations. *Physica D* **222**, 116–130 (2006)
- [15] Hingant, E., Yvinec, R.: Deterministic and stochastic Becker-Dring equations: Past and recent mathematical developments. *Stochastic Processes, Multiscale Modeling and Numerical Methods for Computational Cellular Biology*, 175–204 (2017)
- [16] Hong, B.Z., Keong, L.K., Shariff, A.M.: CFD modelling of most probable bubble nucleation rate from binary mixture with estimation of components' mole fraction in critical cluster. *Continuum Mech. Therm.* **28(3)**, 655–668 (2016)
- [17] Kashchiev, D., Rosmalen, G.M.: Review: Nucleation in solutions revisited, *Crystal Research and Technology* **38**, 555–574 (2003)
- [18] Kirkaldy, J.S., Young D.J.: *Diffusion in the Condensed State*. The Institute of Metals, London (1987)
- [19] Niethammer, B.: On the dynamics of the Becker-Döring equations. Habilitation thesis, University of Bonn (2002)
- [20] Niethammer, B.: On the evolution of large clusters in the Becker-Döring model. *Journal Nonlin. Science* **13**, 115–155 (2003)
- [21] Pathria, R.K.: *Statistical Mechanics*, Butterworth-Heinemann Pub., 2nd Edition, Oxford (1996)
- [22] Penrose, O.: Metastable states for the Becker-Döring cluster equations. *Comm. Math. Phys.* **124**, 515–541 (1989)
- [23] Penrose, O.: The Becker-Döring equations at large times and their connection with the lsw theory of coarsening. *Comm. Math. Phys.* **189**, 305–320 (1997)
- [24] Puglisi, G.: Nucleation and phase propagation in a multistable lattice with weak nonlocal interactions. *Continuum Mech. Therm.* **19(5)**, 299–319 (2007)
- [25] Roters, F., Raabe, D., Gottstein, G.: Work hardening in heterogeneous alloys - a microstructural approach based on three internal state variables. *Acta Materialia* **48**, 4181–4189 (2000)

- [26] Shi, R., Shen, C., Dregia, S.A., Wang, Y: Form of critical nuclei at homo-phase boundaries, *Scripta Materialia* **146**, 276–280 (2018)
- [27] Tutcuoglu, A.D., Vidyasagar, A., Bhattacharya, K., Kochmann, D.M.: Stochastic modeling of discontinuous dynamic recrystallization at finite strains in hcp metals, *Journal of the Mechanics and Physics of Solids* **122** (2019), 590–612 (2019)
- [28] Tutyshkin, N.D., Lofink, P., Mller, W.H., Wille, R., Stahn, O.: Constitutive equations of a tensorial model for strain-induced damage of metals based on three invariants. *Continuum Mech. Therm.* **29**, 251–269 (2017)
- [29] Zecevic, M., Lebensohn, R.A., Mc Cabe, R.J., Knezevic, M.: Modelling recrystallization textures driven by intragranular fluctuations implemented in the viscoplastic self-consistent formulation, *Acta Materialia* **164**, 530–546 (2019)