

Microscopic Simulation of Solid State Sintering Regarding Irregularly Shaped Powder Particles

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Preface

1 Introduction

1.1 The RefraBund Project

1.1.1 Aim and Scope

1.1.2 Project Structure

1.1.3 Process Routes

1.2 State of the Art

1.2.1 Basic Theory of Sintering

1.2.2 Sintering Models with Sharp Interfaces

1.2.3 Sintering Models with Diffuse Interfaces

1.2.4 Monte-Carlo-Methods (MCM)

Classic Monte Carlo Methods

Kinetic Monte Carlo Methods

1.2.5 The Thermodynamic Extremal Principle (TEP)

General things about internal state and non-equilibrium

Applications

Classic Formulation

The classic formulation of the principle was basically formulated by Svoboda and Turek [4], however dependent on the works of Ziegler [5, 6] and Onsager [3]. It is based on the assumption, that the dissipation in the system is always maximized with respect to thermodynamic and kinetic constraints. This is equivalent to the statement, that the system always tries to approach the equilibrium as fast as possible.

The dissipation \mathcal{D} can be formulated as in Equation 1.1a in dependence on the vector of external state variables \mathbf{X} , the vector of internal state variables \mathbf{x} and the velocity vector of internal state variables $\dot{\mathbf{x}}$. It is a bilinear form in the velocity vector of internal state variables and the vector of thermodynamik forces. The latter can be expressed as $\partial G(\mathbf{X}, \mathbf{x}) / \partial \mathbf{x}$, where G is the Gibbs energy of the system. The Gibbs energy is as state function independent of $\dot{\mathbf{x}}$, thus the thermodynamic forces do not include any kinetic constraints. Since \mathcal{D} is linear in $\dot{\mathbf{x}}$, maximizing without further constraints is not meaningful. Therefore, an additional dissipation function \mathcal{Q} is introduced, which must be always equal to \mathcal{D} , since both describe the dissipation of the process (compare Equation 1.1b). However, \mathcal{Q} is formulated in terms of the kinetic conditions of the process and is generally and non-linear form in $\dot{\mathbf{x}}$, often a quadratic form. The actual form of

\mathcal{Q} heavily depends on the regarded process, but it commonly includes empirical kinetic material parameters such as diffusion coefficients or mobilities.

$$\mathcal{D}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) = \frac{\partial G(\mathbf{X}, \mathbf{x})}{\partial \mathbf{x}} \cdot \dot{\mathbf{x}} \rightarrow \max_{\dot{\mathbf{x}}} \quad (1.1a)$$

$$\mathcal{D}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) - \mathcal{Q}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) = 0 \quad (1.1b)$$

The constrained optimization problem in Equation 1.1 can be solved using the Lagrange formalism. The problem is reformulated as the Lagrange functional \mathcal{L} in Equation 1.2 with the additional parameter λ .

$$\mathcal{L} = \mathcal{D}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) + \lambda (\mathcal{D}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) - \mathcal{Q}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}})) \quad (1.2)$$

Setting the gradient of \mathcal{L} with respect to $\dot{\mathbf{x}}$ and λ equal zero gives the optimum of the dissipation \mathcal{D} under the given constraints. Note that the derivative with respect to λ is always identical to the constraint equation.

$$\mathcal{L}_{\dot{\mathbf{x}}} = (1 + \lambda) \frac{\partial G(\mathbf{X}, \mathbf{x})}{\partial \mathbf{x}} - \lambda \frac{\partial \mathcal{Q}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}})}{\partial \dot{\mathbf{x}}} \stackrel{!}{=} 0 \quad (1.3a)$$

$$\mathcal{L}_{\lambda} = \mathcal{D}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) - \mathcal{Q}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) \stackrel{!}{=} 0 \quad (1.3b)$$

For the simple case of \mathcal{Q} being quadratic in $\dot{\mathbf{x}}$ the solution of the system is given in Equation 1.4, which is a linear system of equations of the same size as $\dot{\mathbf{x}}$. Compare Svoboda and Turek [4] and Fischer, Svoboda, and Petryk [1] on this.

$$-\frac{\partial G(\mathbf{X}, \mathbf{x})}{\partial \mathbf{x}} = \frac{1}{2} \frac{\partial \mathcal{Q}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}})}{\partial \dot{\mathbf{x}}} \quad (1.4)$$

Note, that for this formulation it is required, that the dissipation \mathcal{D} and the dissipation function \mathcal{Q} must depend on the same kinetic variables $\dot{\mathbf{x}}$. Often the fluxes \mathbf{j} are used therein.

Generalized Formulation

Recently, Hackl, Fischer, and Svoboda [2] published a generalized formulation of the principle breaking up the need to have the same kinetic variables in the dissipation \mathcal{D} and the dissipation function \mathcal{Q} . The following elaborations use a different notation than in the reference, which fits better to the needs of the application in section 4.6, but the meaning is generally equivalent. The dissipation \mathcal{D} is defined in the same way as before, but the dissipation function \mathcal{Q} does not include the velocities of internal state $\dot{\mathbf{x}}$, but instead the fluxes \mathbf{j} as in Equation 1.5b. Note, that the velocities $\dot{\mathbf{x}}$ are *not* required to be identical to the fluxes \mathbf{j} here, neither they must have the same size. The connections between the velocities $\dot{\mathbf{x}}$ and the fluxes \mathbf{j} are introduced by the constraints \mathbf{f} as in Equation 1.5c. These are required to be able to solve the problem, so they shall be called *required* constraints hereinafter. \mathbf{f} and \mathbf{j} must be of same size. But it is also possible to include several *additional* constraints \mathbf{g} . These may be used for other requirements on the validity of the model, for example geometric constraints. See section 4.6 for details on their application.

$$\mathcal{D}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) = \frac{\partial G(\mathbf{X}, \mathbf{x})}{\partial \mathbf{x}} \cdot \dot{\mathbf{x}} \rightarrow \max_{\dot{\mathbf{x}}} \quad (1.5a)$$

$$\mathcal{D}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) - \mathcal{Q}(\mathbf{X}, \mathbf{x}, \mathbf{j}) = 0 \quad (1.5b)$$

$$\mathbf{f}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}, \mathbf{j}) = 0 \quad (1.5c)$$

$$\mathbf{g}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}, \mathbf{j}) = 0 \quad (1.5d)$$

With this generalized formulation the Lagrange functional writes as in Equation 1.6. Here we have more Lagrange parameters. λ_1 is equivalent to the classic formulation. The vector parameters $\boldsymbol{\lambda}_2$ and $\boldsymbol{\lambda}_3$ for the required and additional constraints are of the same size as \mathbf{f} resp. \mathbf{g} .

$$\mathcal{L} = \mathcal{D}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) + (\mathcal{D}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) - \mathcal{Q}(\mathbf{X}, \mathbf{x}, \mathbf{j})) \lambda_1 + \mathbf{f}^T(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}, \mathbf{j}) \cdot \boldsymbol{\lambda}_2 + \mathbf{g}^T(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}, \mathbf{j}) \cdot \boldsymbol{\lambda}_3 \quad (1.6)$$

As before, the gradient of \mathcal{L} is set equal to zero to obtain the constrained optimum. Finding a simplified equation system as done above (Equation 1.4) is here not possible due to the constraints.

$$\mathcal{L}_{\dot{\mathbf{x}}} = \frac{\partial G(\mathbf{X}, \mathbf{x})}{\partial \mathbf{x}} (1 + \lambda_1) + \frac{\partial \mathbf{f}^T(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}, \mathbf{j})}{\partial \dot{\mathbf{x}}} \cdot \boldsymbol{\lambda}_2 + \frac{\partial \mathbf{g}^T(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}, \mathbf{j})}{\partial \dot{\mathbf{x}}} \cdot \boldsymbol{\lambda}_3 \stackrel{!}{=} 0 \quad (1.7a)$$

$$\mathcal{L}_{\mathbf{j}} = -\frac{\partial \mathcal{Q}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}})}{\partial \mathbf{j}} \lambda_1 + \frac{\partial \mathbf{f}^T(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}, \mathbf{j})}{\partial \mathbf{j}} \cdot \boldsymbol{\lambda}_2 + \frac{\partial \mathbf{g}^T(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}, \mathbf{j})}{\partial \mathbf{j}} \cdot \boldsymbol{\lambda}_3 \stackrel{!}{=} 0 \quad (1.7b)$$

$$\mathcal{L}_{\lambda_1} = \mathcal{D}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) - \mathcal{Q}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}) \stackrel{!}{=} 0 \quad (1.7c)$$

$$\mathcal{L}_{\boldsymbol{\lambda}_2} = \mathbf{f}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}, \mathbf{j}) \stackrel{!}{=} 0 \quad (1.7d)$$

$$\mathcal{L}_{\boldsymbol{\lambda}_3} = \mathbf{g}(\mathbf{X}, \mathbf{x}, \dot{\mathbf{x}}, \mathbf{j}) \stackrel{!}{=} 0 \quad (1.7e)$$

1.2.6 Miscellaneous

2 Aim and Scope

3 Powder Analysis and Representation

3.1 Classic Methods of Powder Characterization

3.2 Particle Description by Parametrized Shape Functions

3.3 Characterization of Powders and Powder Mixtures

4 Model Development

4.1 A Discrete Model of Powder Particles

- Particles, Coordinate System
- Node Types
- Multi-Scale Considerations, Matrix

4.2 Considerations on Free Surfaces

$$\delta_{\perp} = \pi - \frac{1}{2} (\alpha_u + \alpha_l) \quad (4.1)$$

$$\Delta G_{\perp} = (a_u' - a_u + a_l' - a_l) \gamma_S \quad (4.2a)$$

with

$$a_u' = \sqrt{a_u^2 + \Delta s_{\perp}^2 - 2a_u \Delta s_{\perp} \cos \delta_{\perp}} \quad (4.2b)$$

$$a_l' = \sqrt{a_l^2 + \Delta s_{\perp}^2 - 2a_l \Delta s_{\perp} \cos \delta_{\perp}} \quad (4.2c)$$

$$\frac{\partial G}{\partial s_{\perp}} = \lim_{\Delta s_{\perp} \rightarrow 0} \frac{\Delta G_{\perp}}{\Delta s_{\perp}} = -2\gamma_S \cos \delta_{\perp} \quad (4.3)$$

$$\frac{\partial V}{\partial s_{\perp}} = \frac{1}{2} (a_u + a_l) \sin \delta_{\perp} \quad (4.4)$$

$$\delta_{\parallel} = \delta_{\perp} - \frac{\pi}{2} \quad (4.5)$$

$$\Delta G_{\parallel} = (a_u' - a_u + a_l' - a_l) \gamma_S \quad (4.6a)$$

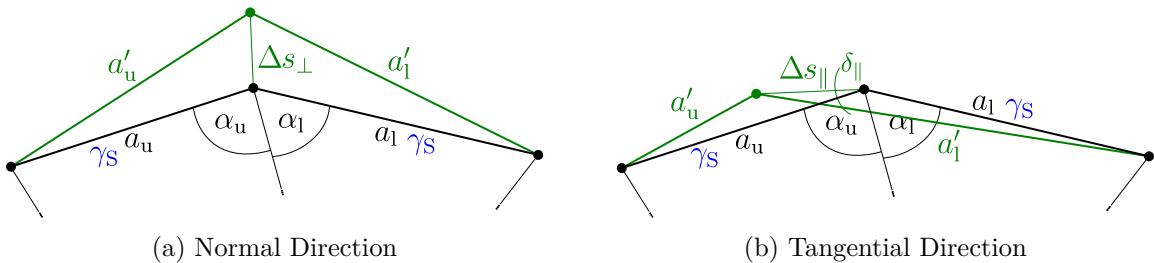


Figure 4.1: Shifting of Surface Nodes

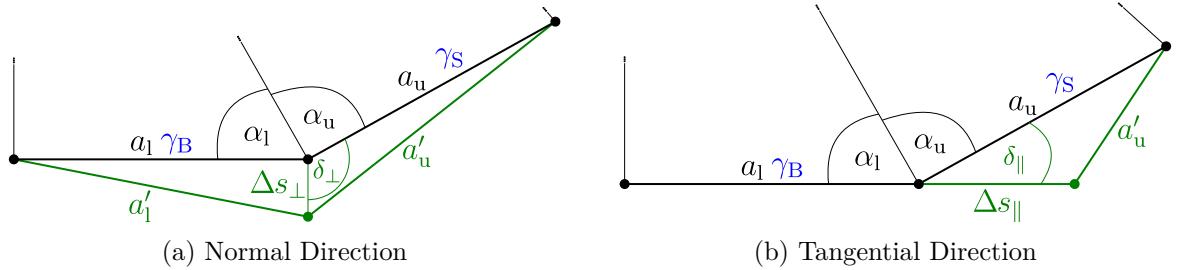


Figure 4.2: Shifting of Neck Nodes

with (note the sign before cosine term)

$$a_u' = \sqrt{a_u^2 + \Delta s_{\parallel}^2 - 2a_u \Delta s_{\parallel} \cos \delta_{\parallel}} \quad (4.6b)$$

$$a_l' = \sqrt{a_l^2 + \Delta s_{\parallel}^2 + 2a_l \Delta s_{\parallel} \cos \delta_{\parallel}} \quad (4.6c)$$

$$\frac{\partial G}{\partial s_{\parallel}} = \lim_{\Delta s_{\parallel} \rightarrow 0} \frac{\Delta G_{\parallel}}{\Delta s_{\parallel}} = 0 \quad (4.7)$$

$$\frac{\partial V}{\partial s_{\parallel}} = \frac{1}{2} (a_l - a_u) \sin \delta_{\parallel} \quad (4.8)$$

4.3 Considerations on Grain Boundaries

$$\frac{\partial G}{\partial s_{\perp}} = \lim_{\Delta s_{\perp} \rightarrow 0} \frac{\Delta G_{\perp}}{\Delta s_{\perp}} = -2\gamma_{GB} \cos \delta_{\perp} \quad (4.9)$$

$$\frac{\partial G}{\partial s_{\parallel}} = \lim_{\Delta s_{\parallel} \rightarrow 0} \frac{\Delta G_{\parallel}}{\Delta s_{\parallel}} = 0 \quad (4.10)$$

4.4 Considerations on Sinter Necks

$$\delta_{\perp} = \frac{3}{2}\pi - \alpha_u - \alpha_l \quad (4.11)$$

$$\Delta G_{\perp} = (a_u' - a_u) \gamma_s + (a_l' - a_l) \gamma_{GB} \quad (4.12a)$$

with

$$a_u' = \sqrt{a_u^2 + \Delta s_{\perp}^2 - 2a_u \Delta s_{\perp} \cos \delta_{\perp}} \quad (4.12b)$$

$$a_l' = \sqrt{a_l^2 + \Delta s_{\perp}^2} \quad (4.12c)$$

$$\frac{\partial G}{\partial s_{\perp}} = \lim_{\Delta s_{\perp} \rightarrow 0} \frac{\Delta G_{\perp}}{\Delta s_{\perp}} = -\gamma_s \cos \delta_{\perp} \quad (4.13)$$

$$\delta_{\parallel} = \pi - \alpha_u - \alpha_l \quad (4.14)$$

$$\Delta G_{\parallel} = (a_u' - a_u) \gamma_S + \frac{1}{2} \gamma_{GB} \Delta s_{\parallel} \quad (4.15a)$$

with

$$a_u' = \sqrt{a_u^2 + \Delta s_{\parallel}^2 - 2a_u \Delta s_{\parallel} \cos \delta_{\parallel}} \quad (4.15b)$$

$$\frac{\partial G}{\partial s_{\parallel}} = \lim_{\Delta s_{\parallel} \rightarrow 0} \frac{\Delta G_{\parallel}}{\Delta s_{\parallel}} = \frac{1}{2} \gamma_{GB} - \gamma_S \cos \delta_{\parallel} \quad (4.16)$$

Note that Δs_{\parallel} points in the opposite direction if the free surface is at the lower side of the neck node.

4.5 Considerations on Grain-Matrix Interfaces

4.6 Application of the Thermodynamic Extremal Principle (TEP)

In the following the generalized extremal principle is applied on the current model of particles and nodes, see subsubsection 1.2.5 for details on the approach. As before, an index of the currently regarded particle or node is neglected where appropriate for brevity. The indices \square_u and \square_l are used to denote the upper resp. lower neighbor of a node.

4.6.1 Time Scale Definition

The application of the TEP results in ordinary differential equations in time which can be solved analytically for simple problems. Here, however, the problem is too complex to be solved analytically, so numerical time integration is required. Usually, the transition from infinitesimal description to finite time steps would be performed as a last step, but doing this step at the beginning opens possibilities of simplifying the equations and clarifies the following elaborations. At this point, it does not matter if a simple Eulerian integration is used or more complex methods like Runge-Kutta. The key feature is, that we use a finite time step width Δt , which is fixed in each time step calculation, but may change from step to step. So, the internal state velocities \dot{x} are directly linked linearly to finite changes using the time step as follows:

$$\Delta x = \dot{x} \cdot \Delta t \quad (4.17)$$

4.6.2 Choice of Variables

First, the internal state variables x , the internal state velocities \dot{x} and the fluxes j must be chosen. The feasibility of the approach heavily depends on this choice.

The choice of the fluxes is straightforward with the diffusional fluxes. One has two diffusional fluxes on each node j_u and j_l , however the flux of the upper node to the lower and the flux from the lower to the upper are always equal due to constance of mass. Additional fluxes are to be found in the fluxes from the particle to the matrix j_M , if a matrix is present in the inter-particle spaces.

The internal state variables are the coordinates of the particles $[r_P, \varphi_P, \omega_P]$ and their nodes $[r, \varphi]$ in space, since they determine the thermodynamic forces and the main aim of the simulation is to follow the time evolution of the particles and nodes.

The straightforward choice of the internal state velocities are the finite changes of internal state variables (e.g. Δr and $\Delta\varphi$). However, for the changes in node coordinates this is not feasible, since they do not depend linearly on the fluxes due to triangular function relations. A more appropriate way is to define the node shifts Δs_{\perp} and Δs_{\parallel} as internal state velocities, so that the required constraints f become linear (see subsection 4.6.5). As they are determined, translation into new coordinates is directly possible using the relations obtained in the previous sections.

The external state variables (such as temperature T and pressure p) are not considered in the following, since they are considered constant.

4.6.3 Dissipation \mathcal{D}

The dissipation at one node is given by the product node shifts and the respective Gibbs energy derivatives as in Equation 4.18. The Gibbs energy derivatives were determined in previous sections for the different node types. The dissipation of the whole system is the sum of all node dissipations, since all thermodynamic forces occur at nodes. Shifting of particles alone does not follow a thermodynamic force, but is determined by the evolution of the related notes.

$$\mathcal{D} = \frac{\partial G}{\partial s_{\perp}} \Delta s_{\perp} + \frac{\partial G}{\partial s_{\parallel}} \Delta s_{\parallel} \quad (4.18)$$

4.6.4 Dissipation Function \mathcal{Q}

$$\mathcal{Q} = \frac{1}{2} \frac{RT}{V_m \xi^o} \left(\frac{a_u j_u^2}{D_u} + \frac{a_l j_l^2}{D_l} \right) \Delta t \quad (4.19)$$

4.6.5 Required Constraints f

Relation between fluxes resp. volume change and shift.

$$\frac{\partial V}{\partial s_{\perp}} \Delta s_{\perp} + \frac{\partial V}{\partial s_{\parallel}} \Delta s_{\parallel} = \Delta V = (j_u + j_l + j_M) \Delta t \quad (4.20)$$

4.6.6 Additional Constraints g

Keeping contacted nodes together.

$$\Delta s_{\perp L} + \Delta s_{\perp R} = \frac{\partial s_{\perp}}{\partial r_P} \Delta r_P + \frac{\partial s_{\perp}}{\partial \varphi_P} \Delta \varphi_P + \frac{\partial s_{\perp}}{\partial \omega_P} \Delta \omega_P \quad (4.21a)$$

$$\Delta s_{\parallel L} + \Delta s_{\parallel R} = \frac{\partial s_{\parallel}}{\partial r_P} \Delta r_P + \frac{\partial s_{\parallel}}{\partial \varphi_P} \Delta \varphi_P + \frac{\partial s_{\parallel}}{\partial \omega_P} \Delta \omega_P \quad (4.21b)$$

Constance of mass.

$$j_{u_i} = j_{l_{i+1}} \quad (4.22)$$

5 Software Implementation of the Model

Reference to open source code

5.1 Representation of Particles and Nodes

- Classes
- Tree Structure
- Coordinate Systems

5.2 Numerical Solution Procedure

- TEP Solution
- Time Step
- Monte-Carlo Drawing

5.3 Calculation and Extraction of Key Features

- Volume Cell, Shrinkage
- Neck Measures

6 Model Validation

6.1 Investigations on Simple Test Cases

- 6.1.1 A Single Particle Free in Space**
- 6.1.2 A Particle Pair Free in Space**
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6.2 Experimental Validation Counter Bulk Sintering Trials

7 Summary and Outlook

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