Karlsruhe Institute of Technology

IAM Institute for Applied Materials

Phase-field modeling of microstructure evolution during polymer solidification

Ahmed Elmoghazy[a], Andreas Prahs[a], Daniel Schneider[a,c], Britta Nestler[a,b,c]
[a]Institute for Applied Materials – Microstructure Modelling and Simulation (IAM-MMS), Karlsruhe Institute of Technology (KIT) , [b]Institute of Digital Materials Science (IDM), University of Applied Sciences (HKA), [c]Institute of Nanotechnology (INT), Karlsruhe Institute of Technology (KIT), *ahmed.elmoghazy@kit.edu

Motivation

- Coarse-grained model of crystallinity morphology on the meso-scale
- A semi-crystalline phase does not represent a singular spherulite but rather a collection of spherulites, quantified by the degree of crystallinity.

(a) $\theta = 205\,°C$ (b) $\theta = 188\,°C$ (c) $\theta = 173\,°C$

Polarized light microscopy images of a sample cooling down at 50 K/min.

Governing Equations

- Helmholtz free energy of the system

$$\psi^\alpha = \psi^\alpha_\chi(\chi_\alpha, \theta) + \psi^\alpha_\theta(\theta)$$

$$\psi^\alpha_\chi = \Delta h^{100}_{f,\alpha} \frac{\theta - \theta_{on,\alpha}}{\theta_{on,\alpha}} \chi_\alpha, \quad \psi^\alpha_\theta = \int_{\theta_{on}}^{\theta} c_{p,\alpha}(\chi_\alpha, \tilde{\theta}) \mathrm{d}\tilde{\theta} - \theta \int_{\theta_{on}}^{\theta} \frac{c_{p,\alpha}(\chi_\alpha, \tilde{\theta})}{\tilde{\theta}} \mathrm{d}\tilde{\theta}$$

- Multiphase-field model

$$\mathcal{F}[\phi, \nabla\phi, \chi] = \int_V f \, \mathrm{d}V = \int_V f_{\mathrm{grad}}(\phi, \nabla\phi) + f_{\mathrm{pot}}(\phi) + \bar{f}_{\mathrm{bulk}}(\phi, \chi) \, \mathrm{d}V$$

$$\frac{\partial \phi_\alpha}{\partial t} = -\frac{1}{\epsilon \mathrm{N}^*} \sum_{\alpha \neq \beta}^{\mathrm{N}^*} M_{\alpha\beta} \left(\frac{\delta \mathcal{F}}{\delta \phi_\alpha} - \frac{\delta \mathcal{F}}{\delta \phi_\beta} \right)$$

- Polymer Crystallization Model

$$\frac{\partial \chi_\alpha}{\partial t} = n_\alpha K_\alpha(\theta, \dot{\theta})(1 - \chi_\alpha) \left(\ln \left(\frac{1}{1 - \chi_\alpha} \right) \right)^{(n_\alpha - 1)/n_\alpha} \frac{\Delta h_{m,\alpha}(\dot{\theta})}{\Delta h^{100}_{f,\alpha}}$$

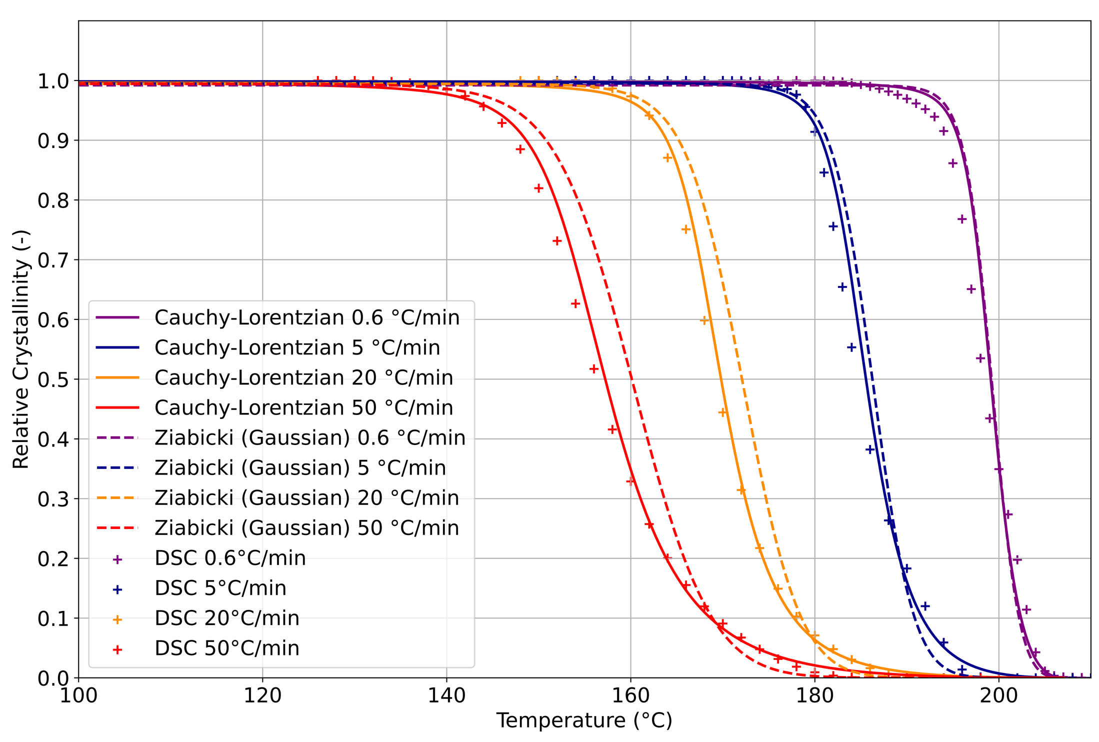
$$K_\alpha(\theta, \dot{\theta}) = K_{max,\alpha}(\dot{\theta}) \exp\left(-4 \ln(2) \frac{(\theta - \theta_{max,\alpha})^2}{D_\alpha^2} \right)$$

- Heat conduction equation

$$\overline{\rho c_p} \dot{\theta} = -\operatorname{div}(\overline{\boldsymbol{q}}) + \underbrace{\sum_{\alpha}^{N^*} \rho_\alpha \Delta h^{100}_{f,\alpha} \dot{\chi}_\alpha \phi_\alpha}_{A} + \underbrace{\sum_{\alpha}^{N^*} \rho_\alpha \Delta h^{100}_{f,\alpha} \dot{\phi}_\alpha \chi_\alpha}_{B} - \underbrace{\sum_{\alpha}^{N^*} \dot{\phi}_\alpha \rho_\alpha \int_{\theta_{on}}^{\theta} c_{p,\alpha}(\chi_\alpha, \tilde{\theta}) \mathrm{d}\tilde{\theta}}_{C}$$

Comparison of different formulations for K_α

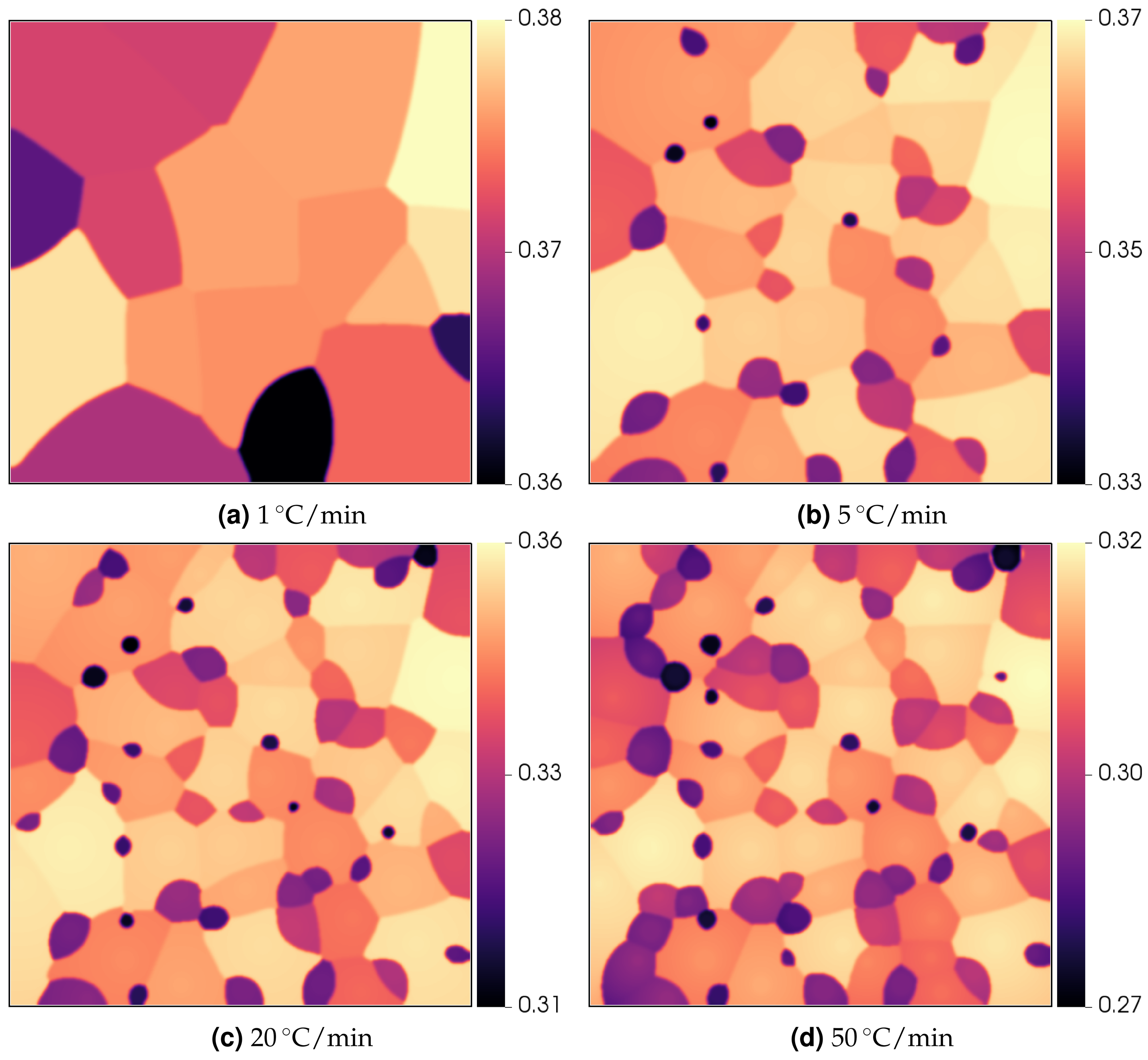
- Cauchy-Lorentzian formulations offers a better fit for this sample



Simulation of crystallinity evolution in a small domain

Initial and Boundary conditions

- 2D domain grid of 500×500 cells with $\Delta x = \Delta y = 1\,\mu$m
- 100 randomly dispersed grains within an amorphous matrix, radii: 4–10 μm
- Initial crystallinity: randomly assigned between 0.6 % and 7 %
- Dirichlet boundaries with cooling rates $R \in \{1, 5, 20, 50\}$ °C/min



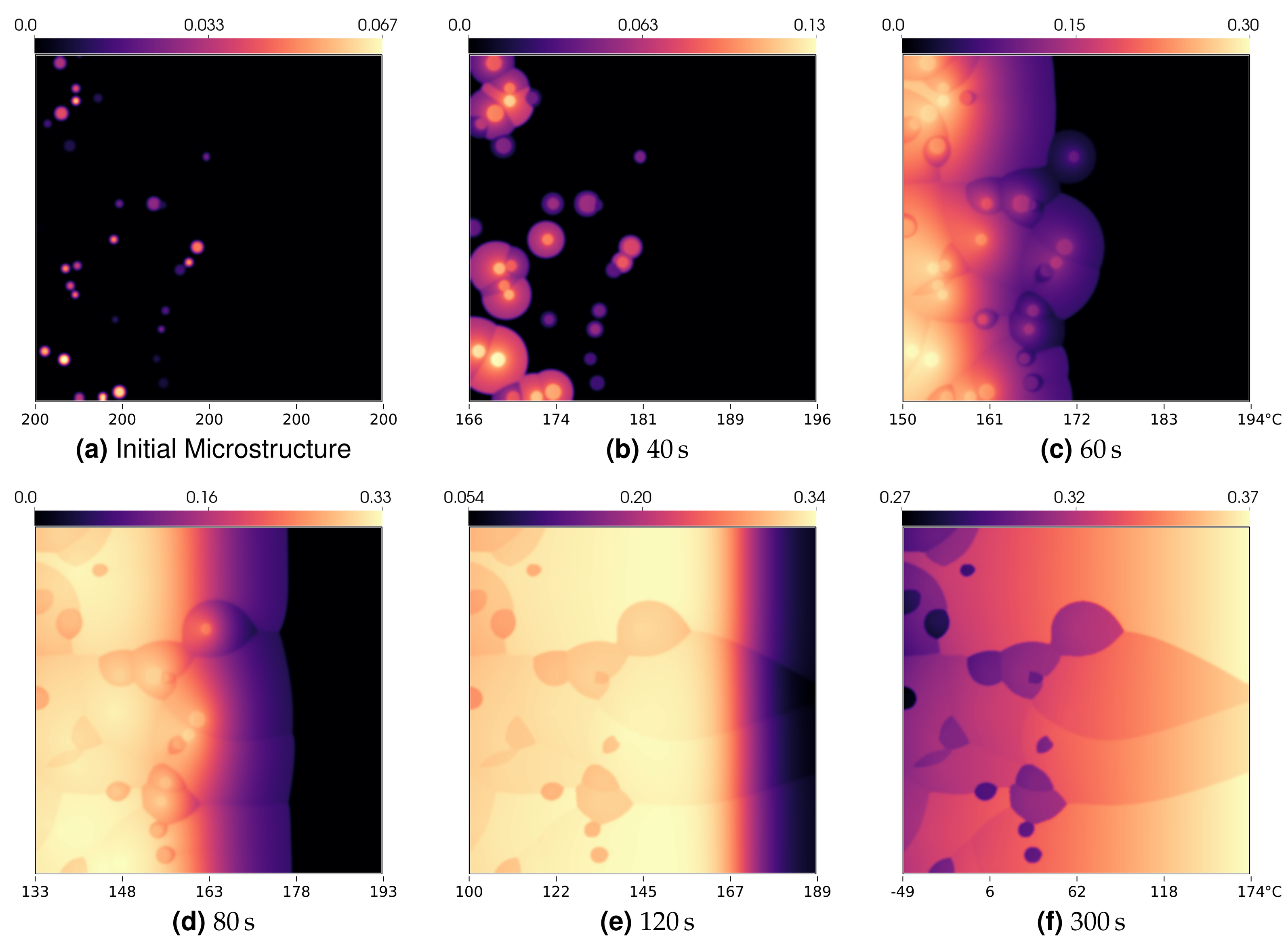
(a) 1 °C/min (b) 5 °C/min (c) 20 °C/min (d) 50 °C/min

Final morphology of the microstructure showing the degree of crystallinity.

Effect of temperature and cooling rate gradients

Initial and boundary conditions

- 2D domain grid of 500×500 cells with $\Delta x = \Delta y = 1\,\mu$m
- Grains are initialized on the left half of the domain
- Left boundary: 50 °C/min, Right boundary: 5 °C/min



(a) Initial Microstructure (b) 40 s (c) 60 s (d) 80 s (e) 120 s (f) 300 s

Acknowledgements

The financial support of the MSE programme (no. 43.31.01) of Helmholtz association, Federal Ministry of Education and Research of Germany in the framework of "DASEA-4-SOFC" (no. 05M2022) is gratefully acknowledged.I also would like to thank my team, my supervisors and the project partners for contributing to this work.