Numerical Simulation and Control of Sublimation Growth of SiC Single Crystals

Olaf Klein¹, Jürgen Geiser¹, Christian Meyer², Peter Philip³, Jürgen Sprekels¹, Fredi Tröltzsch²

¹Weierstrass Institute for Applied Analysis and Stochastics (WIAS)
Berlin, Germany

²TU Berlin
Department of Mathematics
Berlin, Germany

³University of Minnesota
Institute for Mathematics and its Applications (IMA)
Minneapolis, USA

IMA Workshop:
Future Challenges in Multiscale Modeling and Simulation
Minneapolis, November 18, 2004
SiC growth by physical vapor transport (PVT)

- polycrystalline SiC powder sublimes inside induction-heated graphite crucible at 2000 – 3000 K and ≈ 20 hPa
- a gas mixture consisting of Ar (inert gas), Si, SiC₂, Si₂C, . . . is created
- an SiC single crystal grows on a cooled seed
Goal:

Stationary and transient optimal control of process, using mathematical modeling, numerical simulation.

Heat Transport Model

Nonlinear heat conduction in material $j$:

$$\frac{\partial \varepsilon_j}{\partial t} + \text{div } q_j = f_j, \quad q_j = -\kappa_j \nabla T,$$

$\varepsilon_j$: internal energy, $T$: absolute temperature,
$q_j$: heat flux, $\kappa_j$: thermal conductivity,
$f_j$: power density of heat sources (induction heating).

Interface Conditions

Continuity of the heat flux:

Between solids: $q_{j1} \cdot n_{j1} = q_{j2} \cdot n_{j1}$ on $\gamma_{j1,j2}$.

Between gas and solid $j$:

$$q_{\text{gas}} \cdot n_{\text{gas}} - R + J = q_j \cdot n_{\text{gas}} \text{ on } \gamma_{j,\text{gas}},$$

$n_j, n_{\text{gas}}$: outer unit normal, $R$: radiosity, $J$: irradiation.

Continuity of temperature throughout apparatus.
Outer Boundary Conditions

Emission according to Stefan-Boltzmann law:

\[-(\kappa_j \nabla T) \cdot \mathbf{n}_j = \sigma \varepsilon_j(T) (T^4 - T_{room}^4),\]

\(\varepsilon_j: \) emissivity, \(T_{room} = 293 \, \text{K}.\)

On surfaces of open cavities:

\[\mathbf{q}_j \cdot \mathbf{n}_j - R + J = 0.\]
Crystal Growth and Source Sublimation

Consider the crystal surface; the modeling at the source’s surface proceeds analogous.

- **Step 1:** Assume growth is transport-limited, neglect growth kinetics: The “SiC-gas” pressure $p_{\text{SiC-gas}}$ at the surface is identical to the corresponding equilibrium pressure $p_{\text{crystal}}^{eq}$.

- **Step 2:** Include growth kinetics via a Hertz-Knudsen formula:

  \[
  \text{mass flux from gas to crystal} = s_{\text{crystal}} \frac{M_{\text{SiC}}^{1/2}}{(2\pi RT)^{1/2}} \left( p_{\text{SiC-gas}} - p_{\text{crystal}}^{eq} \right),
  \]

  $s_{\text{crystal}}$: probability of colliding molecule to be absorbed by surface, $M_{\text{SiC}}$: molar mass, $R$: universal gas constant, $T$: absolute Temperature.

- **Step 3:** Include chemical reactions: “SiC-gas” actually consists of Si, SiC$_2$, Si$_2$C, Si$_2$, etc. Mass action laws yield relations between the different partial pressures
in the equilibrium, e.g. for

$$2\text{SiC} \rightarrow \text{Si} + \text{SiC}_2,$$
$$\text{SiC} + \text{Si} \rightarrow \text{Si}_2\text{C},$$
$$\text{SiC} + \text{Si}_2 \rightarrow \text{Si}_2\text{C} + \text{Si}$$

involving solid SiC and some gas species:

$$p_{\text{Si}} \, p_{\text{SiC}_2} = K_I(T),$$
$$\frac{p_{\text{Si}_2\text{C}}}{p_{\text{Si}}} = K_{II}(T),$$
$$\frac{p_{\text{Si}_2\text{C}} \, p_{\text{Si}}}{p_{\text{Si}_2}} = K_{III}(T)$$

with appropriate functions $K_I$, $K_{II}$, and $K_{III}$.

- **Step 4:** Formulate mass action laws for reactions changing the composition of the surface.

- **Step 5:** Model the kinetics of the chemical reactions.
Modeling Induction Heating

Assumptions: Axisymmetry, sinusoidal time dependence.

Then a complex magnetic scalar potential $\phi_A$ exists such that the heat sources are:

$$\mu = \frac{\sigma \omega^2}{2} |\phi_A|^2,$$

$\sigma$: electrical conductivity, $\omega$: frequency of imposed voltage. $\phi_A$ is determined from complex, elliptic PDEs:

In insulators:

$$\partial_r \left( \frac{\nu}{r} \partial_r (r \phi_A) \right) + \partial_z (\nu \partial_z \phi_A) = 0,$$

in conductors:

$$-\partial_r \left( \frac{\nu}{r} \partial_r (r \phi_A) \right) - \partial_z (\nu \partial_z \phi_A) + i \omega \sigma \phi_A = \frac{\sigma}{2\pi r} V,$$

$\nu$: reciprocal of magnetic permeability, $i$: imaginary unit, $V$: imposed voltage (non-zero only in coil rings).

$\phi_A$ and its flux are continuous at interfaces, $\phi_A$ vanishes on outer boundaries.

The correct voltage distribution to the coil rings is determined from a linear system.
Known fact: Crystal surface forms along isotherms.
Goal: Radially constant isotherms during growth.

Control: \[ \int_{\Omega_{gas}} w(z) \left( \frac{\partial T}{\partial r}(r, z) \right)^2 d(r, z) \longrightarrow \min. \]

PDEs (\(v_{gas} = 0\), \(f(x, T, P) = f(x, P)\)):
\[ - \text{div} \ \kappa^{(Ar)}(T) \nabla T = 0 \quad \text{in} \ \Omega_{gas}, \]
\[ - \text{div} \ \kappa(x, T) \nabla T = f(x, P) \quad \text{in} \ \Omega \setminus \Omega_{gas}. \]

Constraints:
- \(T_{room} \leq T \leq T_{max}\) in \(\Omega\),
- \(T_{min, SiC-C} \leq T \leq T_{max, SiC-C}\) on \(\Gamma_{SiC-C}\) (need right polytype),
- \(T|_{\Omega_{SiC-S}} \geq T|_{\Gamma_{SiC-C}} + \delta, \ \delta > 0\) (source temp. \(\geq\) seed temp. \(+\delta\)),
- \(0 \leq P \leq P_{max}\) (bounds for heating power \(P\) (control parameter)).
Numerical results: Optimization of temperature field

(a): \( T(P = 10.0 \text{ kW}, z_{\text{rim}} = 24.0 \text{ cm}, f = 10.0 \text{ kHz}) \)

(b): \( T(P = 7.98 \text{ kW}, z_{\text{rim}} = 22.7 \text{ cm}, f = 165 \text{ kHz}) \), Nelder-Mead res. for \( \mathcal{F}_{r,2}(T) \)

(c): \( T(P = 10.3 \text{ kW}, z_{\text{rim}} = 12.9 \text{ cm}, f = 84.9 \text{ kHz}) \), Nelder-Mead res. for \( \frac{\mathcal{F}_{r,2}(T) - \mathcal{F}_{z,2}(T)}{2} \)
Selected Publications


Further Publications / Information:

http://www.ima.umn.edu/~philip/sic/#Publications

http://www.ima.umn.edu/~philip/sic/

Funding:

Supported by the DFG Research Center “Matheon: Mathematics for Key Technologies” in Berlin, by the IMA in Minneapolis, and by the WIAS in Berlin.