Numerical Simulation and Control of Sublimation Growth of SiC Single Crystals

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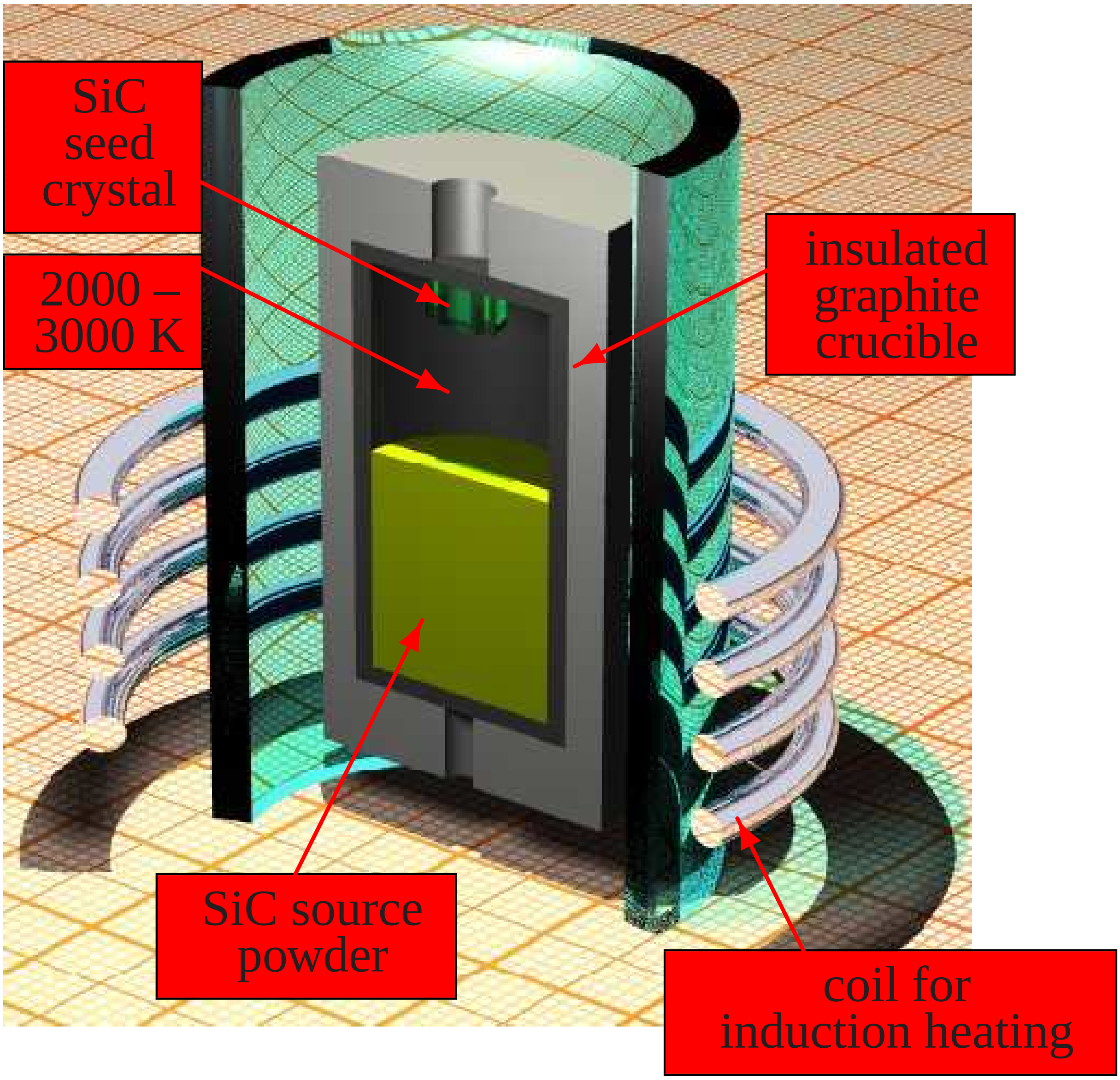
SiC growth by physical vapor transport (PVT)



- polycrystalline SiC powder sublimates inside induction-heated graphite crucible at 2000 – 3000 K and $\approx$ 20 hPa
- a gas mixture consisting of Ar (inert gas), Si, SiC_2, Si_2C, . . . 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:

$$\frac{\partial \varepsilon_j}{\partial t} + \operatorname{div} \mathbf{q}_j = f_j, \qquad \mathbf{q}_j = -\kappa_j \nabla T,$$

ε_j: internal energy, T: absolute temperature,

$\mathbf{q}_j$: heat flux, κ_j: thermal conductivity,

f_j: power density of heat sources (induction heating).

Interface Conditions

Continuity of the heat flux:

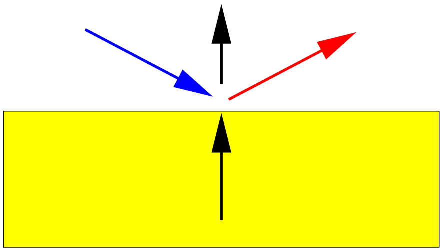
Between solids: $\mathbf{q}_{j_1} \bullet \mathbf{n}_{j_1} = \mathbf{q}_{j_2} \bullet \mathbf{n}_{j_1}$ on γ_{j_1,j_2}.

Between gas and solid j:

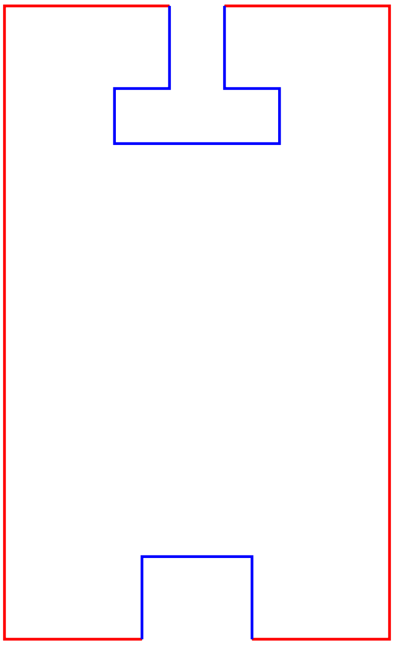
$$\mathbf{q}_{\text{gas}} \bullet \mathbf{n}_{\text{gas}} - R + J = \mathbf{q}_j \bullet \mathbf{n}_{\text{gas}} \text{ on } \gamma_{j,\text{gas}},$$

$\mathbf{n}_j$, $\mathbf{n}_{\text{gas}}$: outer unit normal, R: radiosity, J: irradiation.

Continuity of temperature throughout apparatus.



Outer Boundary Conditions



Emission according to Stefan-Boltzmann law:

$$-\left(\kappa_j \nabla T\right) \bullet \mathbf{n}_j = \sigma \epsilon_j(T) \left(T^4 - T_{\mathrm{room}}^4\right),$$

ϵ_j: emissivity, $T_{\mathrm{room}} = 293$ K.

On surfaces of open cavities:

$$\mathbf{q}_j \bullet \mathbf{n}_j - R + J = 0.$$

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 $p^{\text{eq}}_{\text{crystal}}$.

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

  mass flux from gas to crystal

  $$= \frac{s_{\text{crystal}}\, M_{\text{SiC}}^{1/2}}{(2\pi RT)^{1/2}} \left(p_{\text{SiC-gas}} - p^{\text{eq}}_{\text{crystal}} \right),$$

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

- Step 3: Include chemical reactions: "SiC-gas" actually consists of Si, SiC_2, Si_2C, Si_2, etc. Mass action laws yield relations between the different partial pressures in the equilibrium, e.g. for

$$
\begin{aligned}
2\mathrm{SiC} &\longrightarrow \mathrm{Si} + \mathrm{SiC}_2, \\
\mathrm{SiC} + \mathrm{Si} &\longrightarrow \mathrm{Si}_2\mathrm{C}, \\
\mathrm{SiC} + \mathrm{Si}_2 &\longrightarrow \mathrm{Si}_2\mathrm{C} + \mathrm{Si}
\end{aligned}
$$

involving solid SiC and some gas species:

$$
\begin{aligned}
p_{\mathrm{Si}}\; p_{\mathrm{SiC}_2} &= K_{\mathrm{I}}(T), \\
\frac{p_{\mathrm{Si}_2\mathrm{C}}}{p_{\mathrm{Si}}} &= K_{\mathrm{II}}(T), \\
\frac{p_{\mathrm{Si}_2\mathrm{C}}\; p_{\mathrm{Si}}}{p_{\mathrm{Si}_2}} &= K_{\mathrm{III}}(T)
\end{aligned}
$$

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 ϕ_A exists such that the heat sources are:

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

σ: electrical conductivity, ω: frequency of imposed voltage. ϕ_A is determined from complex, elliptic PDEs:

In insulators:

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

in conductors:

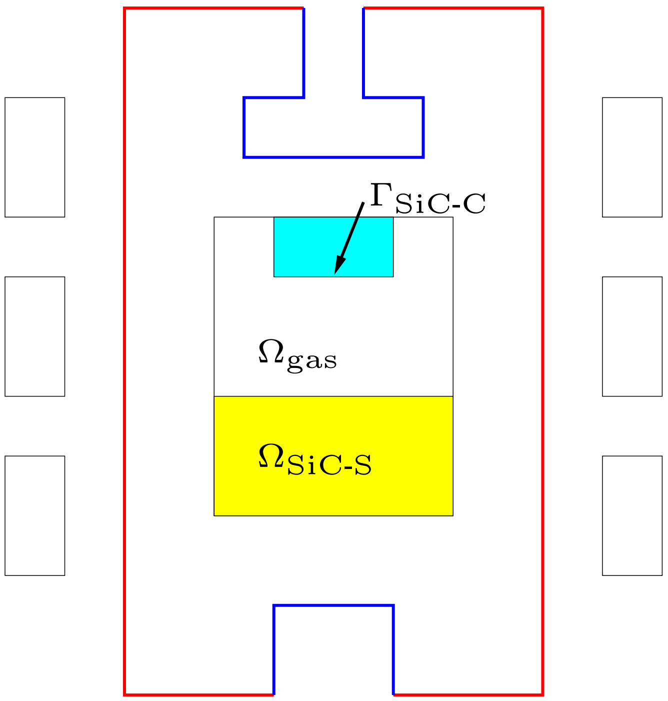
$$-\partial_r\left(\frac{\nu}{r}\partial_r(r\phi_A)\right) - \partial_z\left(\nu\partial_z\phi_A\right) + i\omega\sigma\phi_A = \frac{\sigma}{2\pi r}V,$$

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

ϕ_A and its flux are continuous at interfaces, ϕ_A vanishes on outer boundaries.

The correct voltage distribution to the coil rings is determined from a linear system.

Stationary optimal control problem for the temperature field



Known fact: Crystal surface forms along isotherms.

Goal: Radially constant isotherms during growth.

Control: $$\int_{\Omega_{\text{gas}}} w(z) \; \frac{\partial T}{\partial r}(r,z) \;^{2} \, \mathrm{d}(r,z) \longrightarrow \text{min.}$$

PDEs ($\mathbf{v}_{\text{gas}} = 0,\ f(x,T,P) = f(x,P)$):

$$-\operatorname{div}\ \kappa^{(\text{Ar})}(T)\,\nabla T = 0 \quad \text{in } \Omega_{\text{gas}},$$

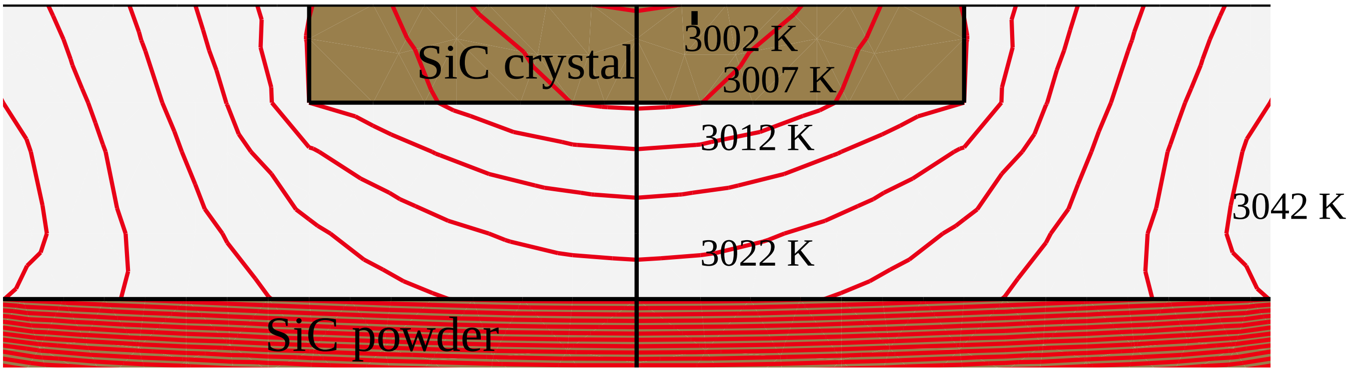
$$-\operatorname{div}\ \kappa(x,T)\,\nabla T = f(x,P) \quad \text{in } \Omega \setminus \Omega_{\text{gas}}.$$

Constraints:

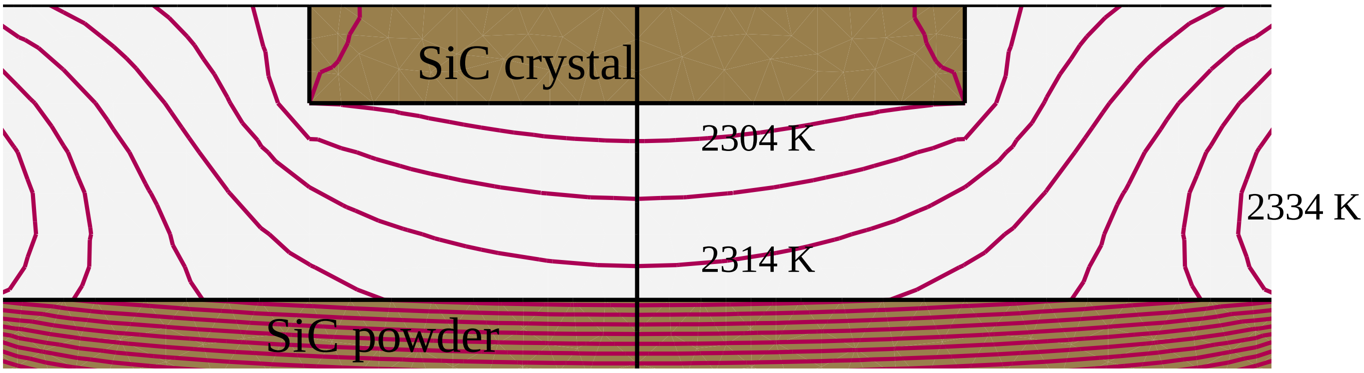
- $T_{\text{room}} \leq T \leq T_{\text{max}}$ in Ω,
- $T_{\text{min,SiC-C}} \leq T \leq T_{\text{max,SiC-C}}$ on $\Gamma_{\text{SiC-C}}$ (need right polytype),
- $T{\restriction}_{\Omega_{\text{SiC-S}}} \geq T{\restriction}_{\Gamma_{\text{SiC-C}}} + \delta, \quad \delta > 0$ (source temp. $\geq$ seed temp. $+\delta$),
- $0 \leq P \leq P_{\text{max}}$ (bounds for heating power P (control parameter)).

Numerical results: Optimization of temperature field

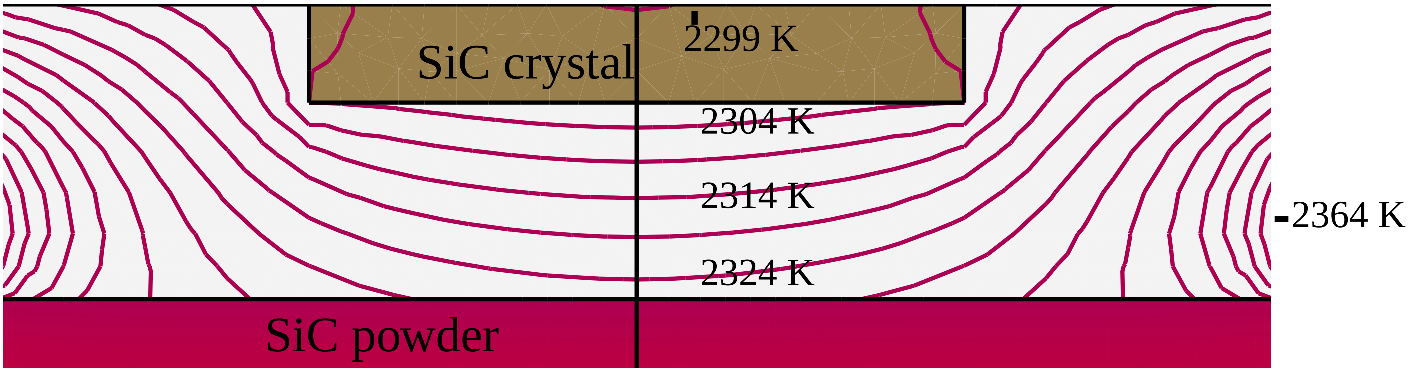
(a): $T(P = 10.0\ \mathrm{kW}, z_{\mathrm{rim}} = 24.0\ \mathrm{cm}, f = 10.0\ \mathrm{kHz})$



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



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



Selected Publications

- C. Meyer, P. Philip, F. Tröltzsch: *Optimal Control of a Semilinear PDE with Nonlocal Radiation Interface Conditions.* Preprint No. 2002 of the Institute for Mathematics and its Applications (IMA), Minneapolis, 2004. Submitted.
- C. Meyer, P. Philip: *Optimizing the temperature profile during sublimation growth of SiC single crystals: Control of heating power, frequency, and coil position.* Preprint No. 895 of the Weierstrass Institute for Applied Analysis and Stochastics (WIAS), Berlin, 2003. Submitted.
- O. Klein, P. Philip, J. Sprekels: *Modeling and simulation of sublimation growth of SiC bulk single crystals*, Interfaces and Free Boundaries 6 (2004), 295–314.

Further Publications / Information:

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

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

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