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Implicit time integration methods and inexact Newton methods: application to chemical vapor deposition

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Technical University Berlin, Germany

Outline

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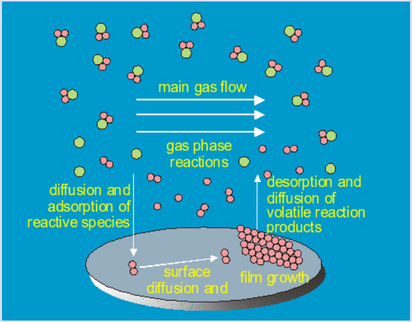
Chemical Vapor Deposition

- Synthesizes thin solid film from gaseous phase by chemical reaction on solid material
- Reactions driven by thermal energy



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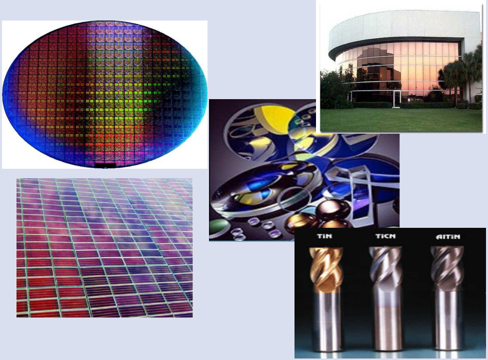
Chemical Vapor Deposition

Applications



- Semiconductors
- Solar cells
- Optical, mechanical and decorative coatings

Transport Model for CVD

Mathematical Model

- Continuity equation
- Navier-Stokes equations
- Energy equation in terms of temperature
- Species equation
- Ideal gas law

Species Equation

- Advection Diffusion Reaction Equation

$$\frac{\partial(\rho\omega)}{\partial t} = \nabla \cdot (\rho \mathbf{v} \omega) + \nabla \cdot (\mathbb{D}\nabla\omega) + m \sum_{k=1}^{\#\text{reactions}} \nu_k R_k^G$$

Transport Model for CVD

Reaction Rate

- Net molar reaction rate

$$R_k^G = k_{k,fw} \sum_{i=1}^{S} \|\nu_{i,s}\| c_i - k_{k,bw} \sum_{i=1}^{S} \| - \nu_{i,s}\| c_i$$

- Modified Law of Arrhenius $k_{k,fw} = A_k \cdot T^{\beta_k} e^{-\frac{E_k}{RT}}$
- $\max(k_{k,fw}) / \min(k_{k,bw}) = 10^{28}$

Properties Mathematical Model of CVD

- Consists of $(\#\text{species} - 1 + 3 + d)$ coupled PDEs
- Stiff nonlinear system of species equations

Numerical Methods

Properties

- Stiff Problem $\longrightarrow$ Stable Time Integration
- Positivity (= preservation of non-negativity):
  Negative Species can cause blow up of the solution
- Efficiency / Robustness
- Method of Lines approach

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Positivity

Mass fractions

A natural property for mass fractions is their non-negativity

Positivity of mass fractions should hold for . . .

1. Model equations
2. Spatial discretization: Hybrid scheme
   Introduces locally first order upwinding
3. Time integration
4. Iterative solvers:
   (Non)linear solver

Positivity for ODE systems

Euler Backward

- $w_{n+1} - w_n = \tau F(t_{n+1}, w_{n+1})$
- Unconditionally stable (*A*-stable/ stiffly stable)

Theorem (Hundsdorfer, 1996)

Euler Backward is positive for any step size τ

Theorem (Bolley and Crouzeix, 1970)

Any unconditionally positive time integration is at most first order accurate

Nonlinear Solvers

Inexact Newton to solve $F(x) = 0$

Let x_0 be given.
FOR $k = 1, 2, \ldots$ until 'convergence'
Find some $\eta_k \in [0, 1)$ and s_k that satisfy

$$\|F(x_k) + F'(x_k)s_k\| \leq \eta_k \|F(x_k)\|.$$

Set $x_{k+1} = x_k + s_k$.
ENDFOR

Nonlinear Solvers

Inexact Newton Condition

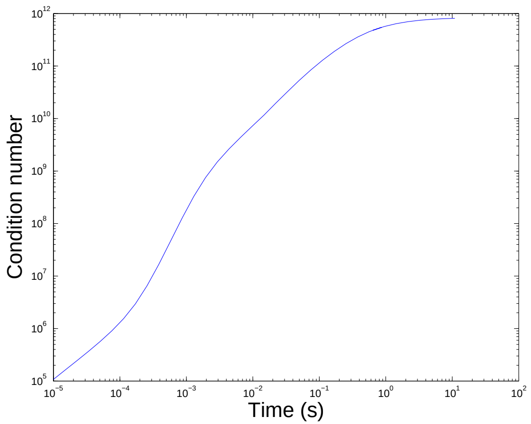
$$\|F(x_k) + F'(x_k)s_k\| \leq \eta_k \|F(x_k)\|$$

Choices for Forcing Term

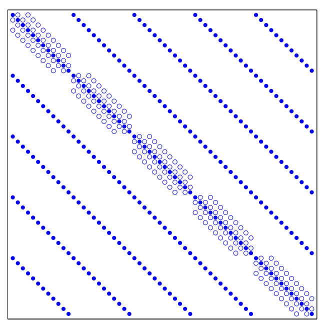
- $\eta_k = \dfrac{\left| \|F(x_k)\| - \|F(x_{k-1}) - F'(x_{k-1})s_{k-1}\| \right|}{\|F(x_{k-1})\|}$
- $\eta_k = \gamma \dfrac{\|F(x_k)\|^2}{\|F(x_{k-1})\|^2}$

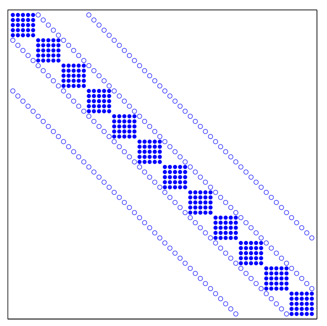
Properties

- Huge condition numbers due to chemistry terms



Lexicographic ordering (left) and Alternate blocking per grid point(right)





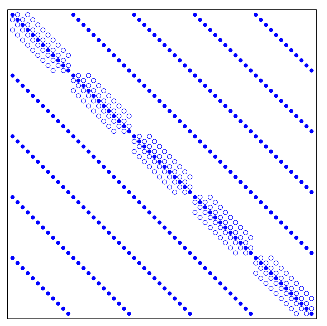
Iterative Linear Solver

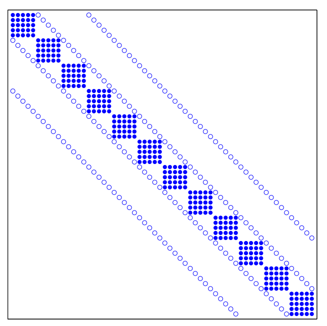
- Right preconditioned BiCGStab
- 'Heavy' chemistry terms → diagonal blocks
- Incomplete Factorization: ILU(0)

Number of	lexico graphic	alternate blocking per gridpoint
F	220	197
Newton iters	124	111
Linesearch	12	7
Rej. time steps	0	0
Acc. time steps	36	36
CPU Time	400	300
linear iters	444	346

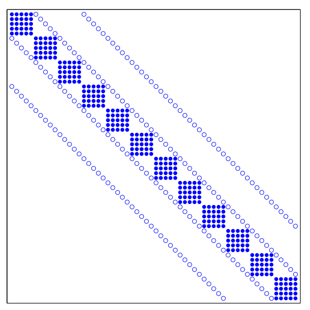
Preconditioners: Lumping

Important: Lumping per species





Preconditioners: Block Diagonal



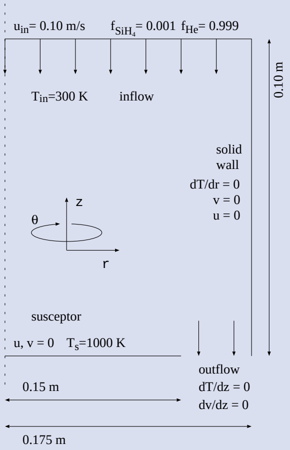
- ‘natural’ blocking over species
- series of uncoupled systems → LU factorization per subsystem

Preconditioners: Block D-ILU

- Block version of the matrix derived from central differences on a Cartesian product grid
- To compute: inverse of a diagonal block $\longrightarrow$ solve linear system directly
- Storage: factorization of diagonal blocks

Kleijn's Benchmark Problem

Computational Domain



- Axisymmetric
- 0.1 mole% SiH_4 at the inflow
- Rest is carrier gas helium He
- Susceptor does not rotate

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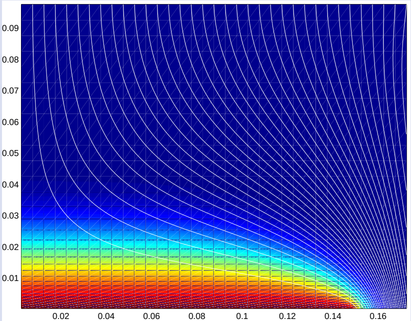
Kleijn's Benchmark Problem

Computational Domain



- grid sizes: 35 $\times$ 32 up to 70 $\times$ 82 grid points
- Temperature:
  Inflow 300 K
  Susceptor 1000 K
- Uniform velocity at inflow

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Kleijn's Benchmark Problem

Chemistry Model: 16 species, 26 reactions [1]

- Above heated wafer SiH_4 decomposes into SiH_2 and H_2
- Chain of 25 homogeneous gas phase reactions
- Including the carrier gas the gas mixture contains 17 species, of which 14 contain silicon atoms
- Irreversible surface reactions at the susceptor leads to deposition of solid silicon

[1] M.E. Coltrin, R.J. Kee and G.H. Evans, A Mathematical Model of the Fluid Mechanics and Gas-Phase Chemistry in a Rotating Chemical Vapor Deposition Reactor, J. Electrochem. Soc., 136, (1989)

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Numerical Results

Integration statistics: 35×32 grid

	ILU(0)	Lumped Jac	block DILU	block diag	direct solver
F	197	310	210	3181	190
Newton	111	185	112	1239	94
linesearch	7	20	13	0	11
Rej. time step	0	3	0	459	1
Acc. time step	36	41	36	774	38
lin iters	346	3693	676	3315	
CPU	300	590	380	3250	6500

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Numerical Results

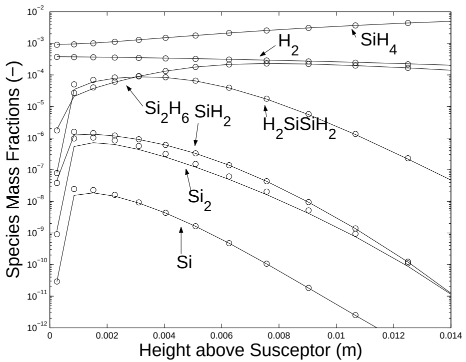
Integration statistics: 70 × 82 grid

	ILU(0)	Lumped Jac	block DILU	block diag
F	869	nf	613	nf
Newton	476	nf	327	nf
linesearch	127	nf	106	nf
Rej. time step	15	nf	0	nf
Acc. time step	62	nf	37	nf
lin iters	8503	nf	2036	nf
CPU	7400	nf	5300	nf

Direct solver is not feasable.

Kleijn's Benchmark Problem

Validation: Species mass fraction along the symmetry axis



- solid: Kleijn's solutions
- circles: our solutions

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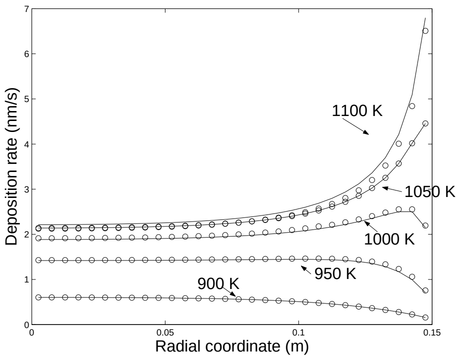
Kleijn's Benchmark Problem

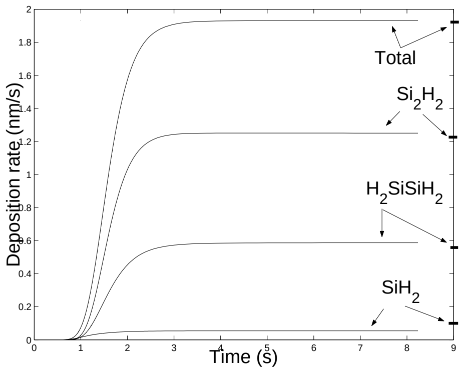
Validation: Radial profiles of total steady state deposition rate



- wafer temperature from 900 K up to 1100 K
- solid: Kleijn's solutions
- circles: our solutions

Kleijn's Benchmark Problem

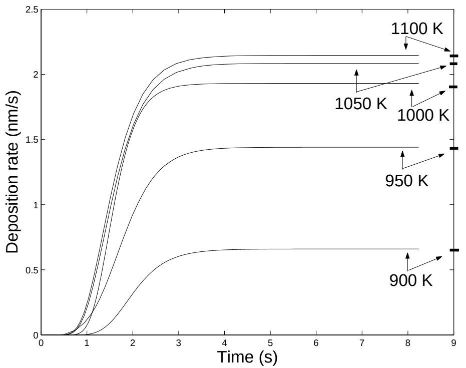
Transient behavior of deposition rates



- along symmetry axis
- wafer 1000 K

Kleijn's Benchmark Problem

Transient behavior of deposition rates



- along symmetry axis
- wafer temperatures from 900 up to 1100 K

Conclusions and Future Research

Conclusions

- Euler Backward is unconditionally positive, but Inexact Newton does not preserve this property
- Alternate blocking per grid point is more effective
- Easy preconditioners are effective for 2D problem
- Chemistry source terms should be in the preconditioner

Conclusions and Future Research

Future Research

- 3D transient simulations
- How to preserve positivity when iterative linear solvers are used ?
- More realistic chemistry/surface chemistry models
- Steady state solver

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References and Contact Information

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References and Contact Information

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