Efficient Numerical Schemes for the Simulation of Adsorption Processes to Cyclic Steady State

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Efficient Numerical Schemes for the Simulation of Adsorption Processes to Cyclic Steady State

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Outline

Why do we need efficient simulation tools?

Benefits of simulation

- Interpretation of experimental results
  - Insight into different physical effects and device behaviour
  - Estimation of device and process parameters
- Predicting the behaviour of future experiments
  - Design of experiments
  - Optimisation of the process

Very complex problem

- 3D problem for pressure, temperature and concentration
- Different length scales: column, pellets, crystals
- Different time scales: convection, macro- and micropore diffusion, adsorption, heat transfer

Potential simplifications of the model

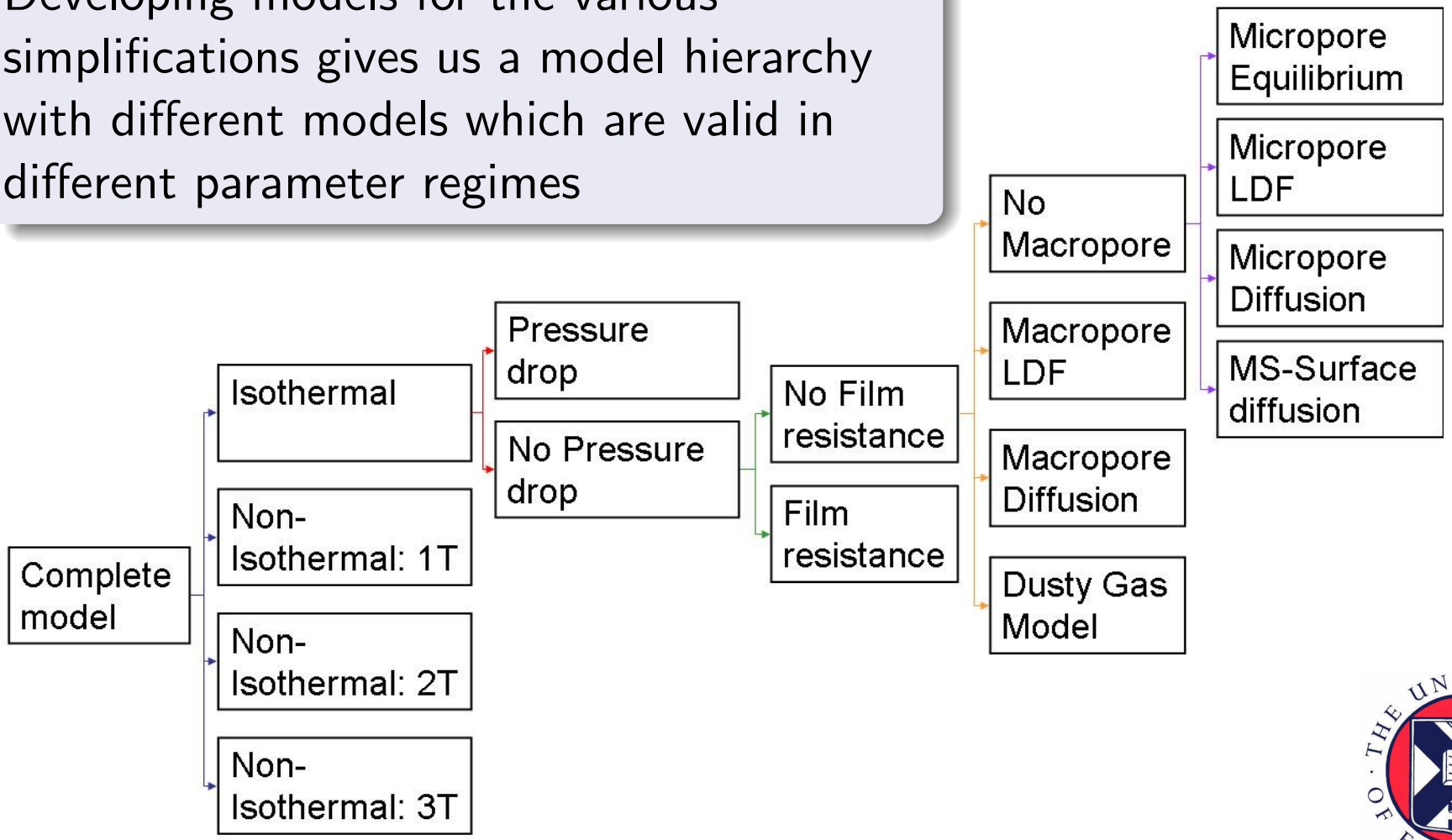
We can write the full, complex model but in many cases a simplified model will give the same results with much less effort

Model simplifications

- Axial dispersed plug flow
- No frictional pressure drop or Ergun equation
- Mass transfer: LDF, macro- or micropore diffusion, film resistance
- Equilibrium isotherm: linear, Langmuir, dual-site Langmuir
- Temperature: isothermal, non-isothermal, wall loss

Adsorption process model hierarchy

Developing models for the various simplifications gives us a model hierarchy with different models which are valid in different parameter regimes



Adsorption column: 1D, Linear Driving Force (LDF)

- Axial dispersed plug flow
- LDF mass transfer
- No frictional pressure drop
- Isothermal system
- Langmuir isotherm
- Ideal gas

Gas phase material balance

$$\frac{\partial c_i}{\partial t} + \frac{1-\epsilon}{\epsilon}\frac{\partial \bar{q}_i}{\partial t} + \frac{\partial (c_i u)}{\partial z} + \frac{\partial J_i}{\partial z} = 0$$

$$D\frac{\partial c_i}{\partial z}\bigg|_{z=0} = \frac{u+|u|}{2}\left(c_{i0+} - c_{i0-}\right)$$

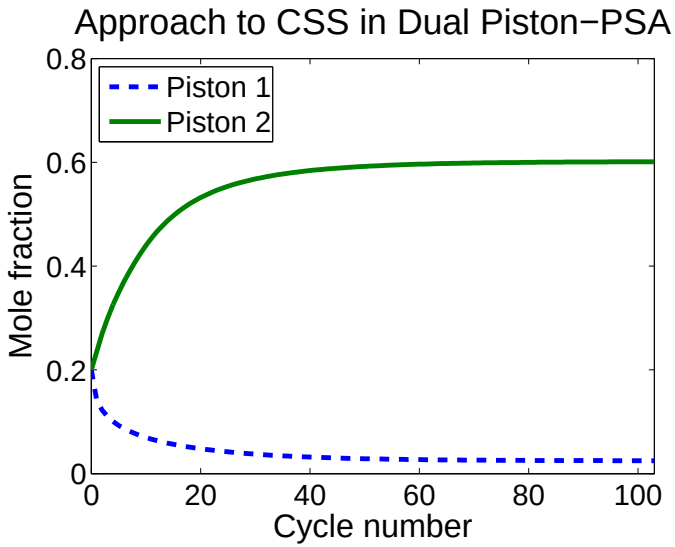
Solid phase material balance

$$\frac{\partial \bar{q}_i}{\partial t} = k_i\left(\frac{q_{si} b_p P x_i}{1 + b_p P x_i} - \bar{q}_i\right)$$

Difficulties in simulating adsorption processes

Difficulties

- Dynamic systems with many parameters and variables
- Many process configurations possible
- Large Peclet number and shock formation
- Convergence to cyclic steady state can be slow



Simulation tool requirements

- Conservation of mass, energy and momentum
- Fast simulation of one cycle
- Acceleration of convergence
- Robustness and ease of use

Solution strategy

Discretise only the spatial dimension: Method of Lines

- Gas phase discretised by the Finite Volume Method (FVM)
- Solid phase discretised by the Orthogonal Collocation on Finite Elements Method (OCFEM) when diffusion in the particles is included

Simulation to Cyclic Steady State

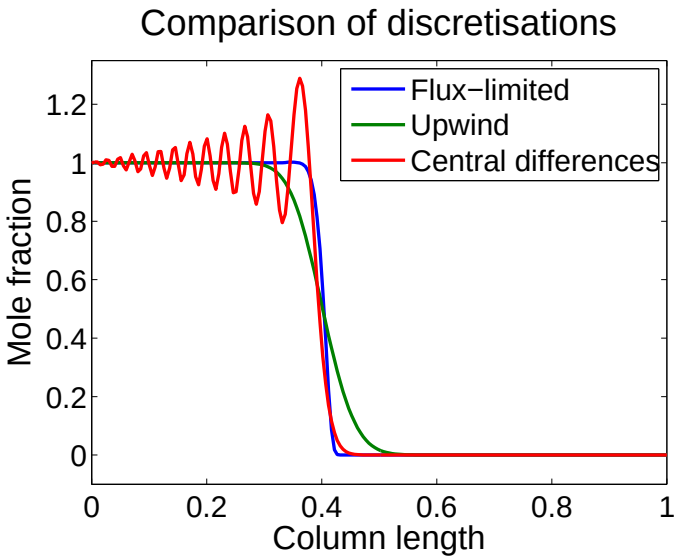
- Differential Algebraic Equations are solved by state-of-the-art solver SUNDIALS
- Accelerate the convergence to CSS with extrapolation

Benefits

- Spatial discretisations tailored to the corresponding phase
- SUNDIALS is robust and simplifies the development

Flux-limiting FVM for convective term (1/2)

- Sharp fronts require specialised discretisation schemes
- Use higher order methods in smooth regions
- First-order upwind methods close to the shock



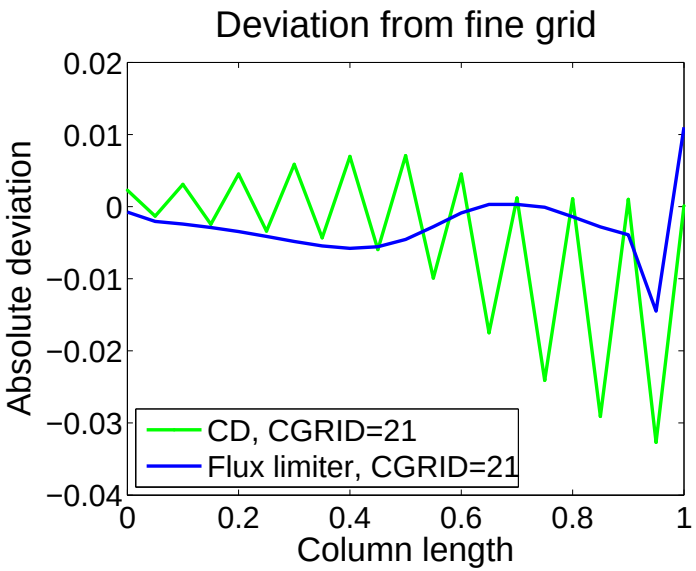
$$\frac{\partial y_i}{\partial t} + \frac{\left(F_{i+1/2} - F_{i-1/2}\right)}{\Delta z} = \frac{y_{i+1} - 2y_i + y_{i-1}}{\Delta z^2} + R_i$$

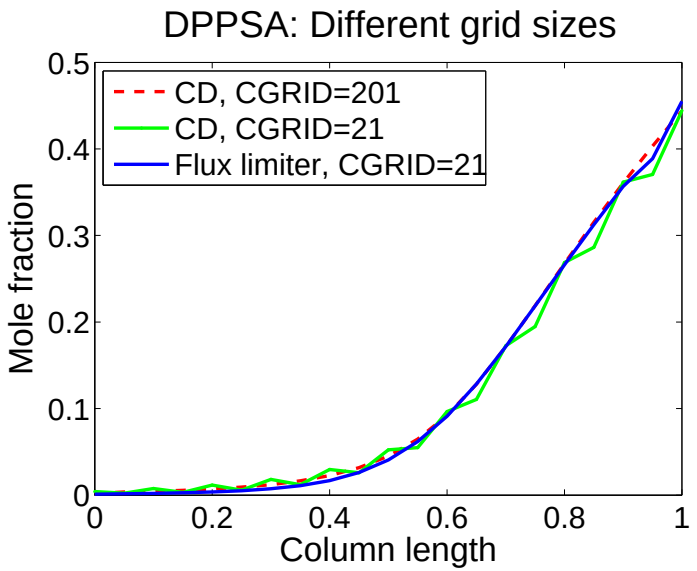
$$F_{i+1/2} = \frac{v}{2}\left[\left(y_i + y_{i+1}\right) + \left(1 - \sigma_i\right)\left(y_{i+1} - y_i\right)\right]$$

$$\sigma(\theta) = \frac{\theta + |\theta|}{1 + \theta} \qquad \theta = \frac{y_i - y_{i-1}}{y_{i+1} - y_i}$$

Flux-limiting FVM for convective term (2/2)

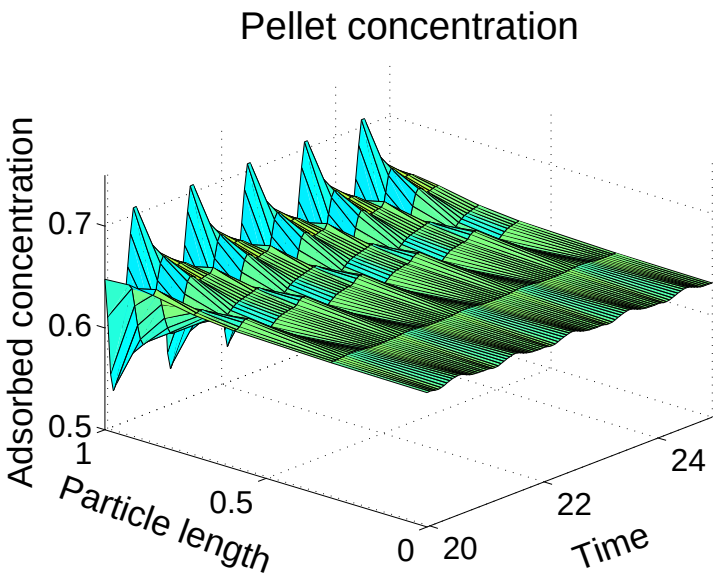
- Finite Volume Method is inherently conservative
- Simulations with a low number of grid points are possible
- Flux-limiting scheme guarantees the correct behaviour of the solution





Orthogonal collocation on finite element method

The adsorbed concentration in an adsorbent pellet changes considerably over one cycle but only a small part of the pellet actively takes part in the process.



- Uniquely suited to stationary gradients in the solid phase
- Split the domain into small elements to handle steep gradients
- High order of accuracy for a small number of grid points

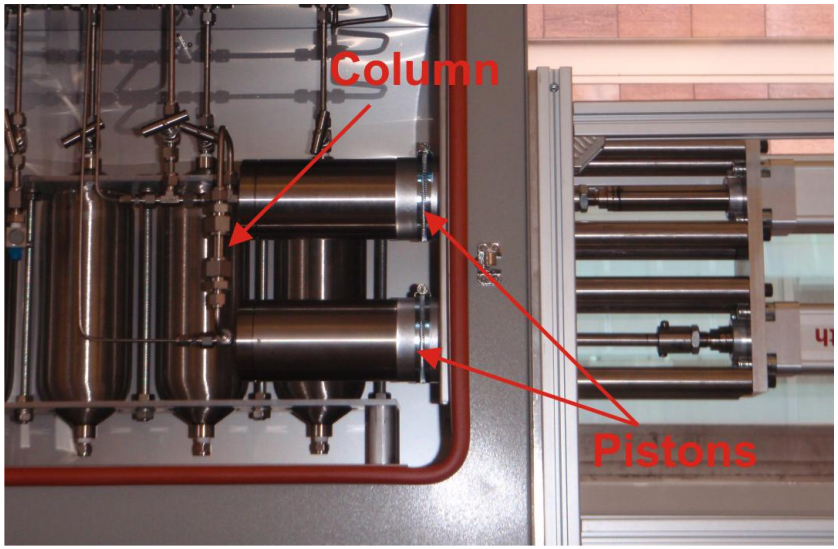
SUNDIALS

- Solver for Differential/Algebraic equation systems $F(t, y(t), \dot{y}(t)) = 0$
- Based on variable-order, variable-step size Backward Differentiation Formulas
- Nonlinear systems are solved by Newton iteration
- Linear systems are solved by direct or iterative solvers
- Forward and adjoint sensitivity analysis
- https://computation.llnl.gov/casc/sundials/main.html

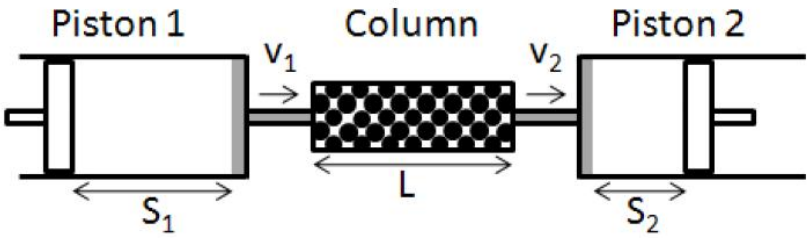
Benefits

- Based on robust and efficient algorithms
- Modular implementation in C
- User control over most aspects of the integration

Dual Piston Pressure Swing Adsorption (DP-PSA)



- Low sample requirement
- Rapid testing of adsorbents
- Many different configurations

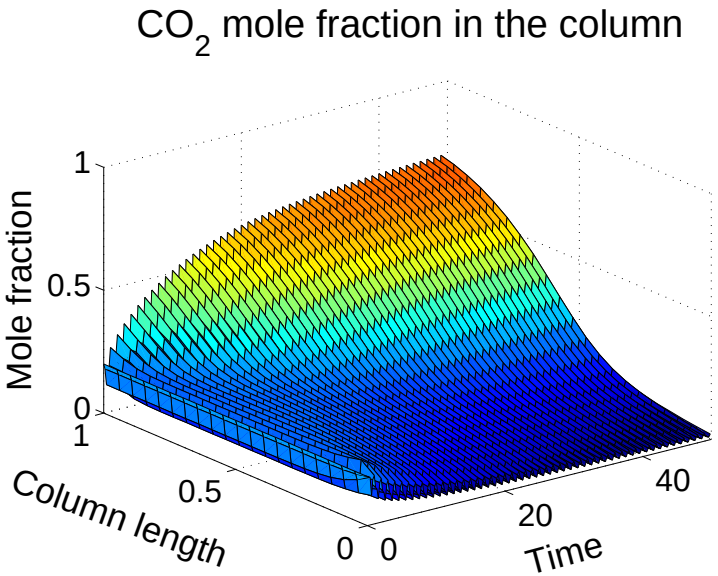


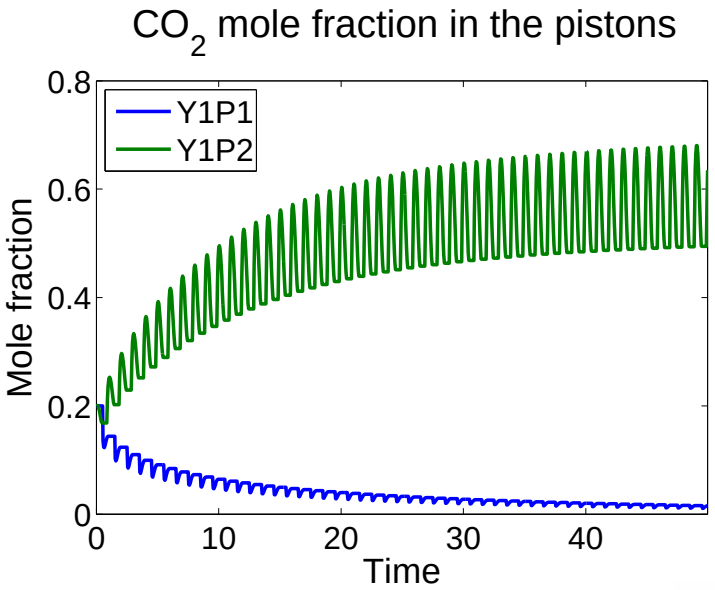
Efficient numerical simulation tool required

- Closed system: needs conservative scheme
- Dynamic system with many parameters and variables
- Large Peclet number: shock formation and propagation
- Long computation to cyclic steady state

Simulation of the DP-PSA system

Lots of data from this experiment so need a fast and efficient dynamic simulator for the analysis of the experiments and to estimate adsorbent parameters





Test case starting with uniform CO_2/N_2 mixture and inducing a CO_2 concentration gradient along the column length

Acceleration to Cyclic Steady State

Cyclic Steady State

- Many adsorption systems will reach Cyclic Steady State
- At CSS the system state is not changing between cycles, i.e. $y(kt_c) = y((k+1)t_c)$
- Sequential approach to CSS can be very slow, i.e. 1000 s of cycles

Acceleration schemes

1. Non-sequential cycle calculation: extrapolation algorithms
2. Model and discretisation switch
3. Interpolation of the starting conditions

ϵ extrapolation

Extrapolate the next solution from the previous $2m$ solutions

1. Set $x_0 = y_i$
2. Generate $x_{i+1} = F(x_i)$
3. Apply ϵ algorithm
4. Set $y_{i+1} = \epsilon_{2m}^{(0)}$

ϵ algorithm:

$$\epsilon_{-1}^{r} = 0$$

$$\epsilon_{0}^{r} = x_r$$

$$\epsilon_{s+1}^{r} = \epsilon_{s-1}^{r+1} + \left(\epsilon_{s}^{(r+1)} - \epsilon_{s}^{(r)}\right)^{-1}$$

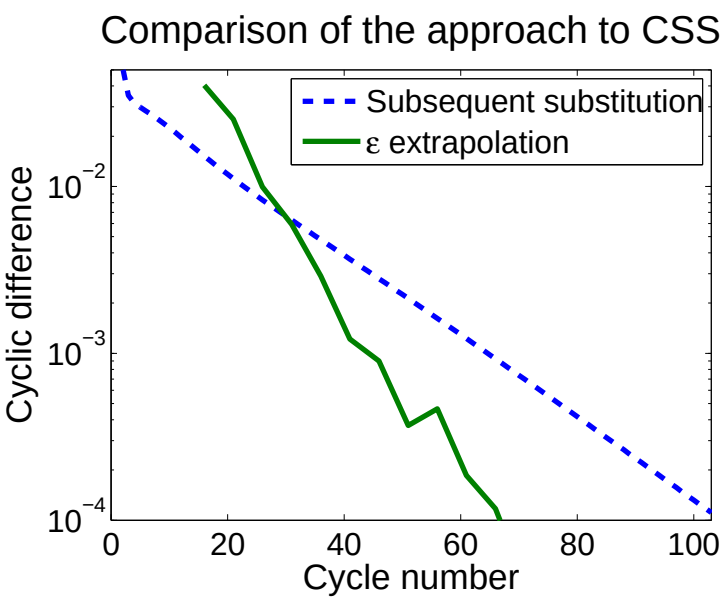
$$\begin{array}{ccccc}
\epsilon_0^0 & & & & \\
& \epsilon_1^0 & & & \\
\epsilon_0^1 & & \epsilon_2^0 & & \\
& \epsilon_1^1 & & \epsilon_3^0 & \\
\epsilon_0^2 & & \epsilon_2^1 & & \epsilon_4^0 \\
& \epsilon_1^2 & & \epsilon_3^1 & \\
\epsilon_0^3 & & \epsilon_2^2 & & \\
& \epsilon_1^3 & & & \\
\epsilon_0^4 & & & &
\end{array}$$

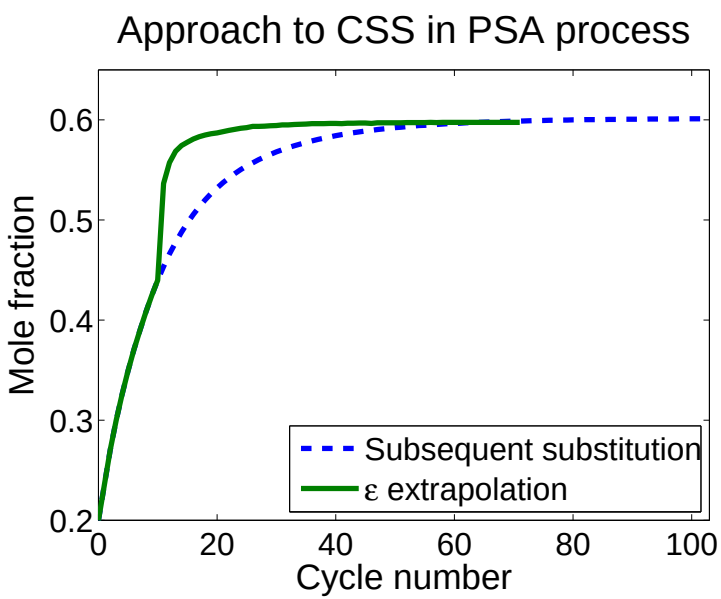
- m depends on the problem eigenvalues
- For the DP-PSA $m = 2$ or 3
- Switch to subsequent substitution close to CSS
- Skelboe, IEEE Transactions on Circuits and Systems, 27, 3, 1980

ϵ extrapolation: Convergence to CSS

Convergence to CSS

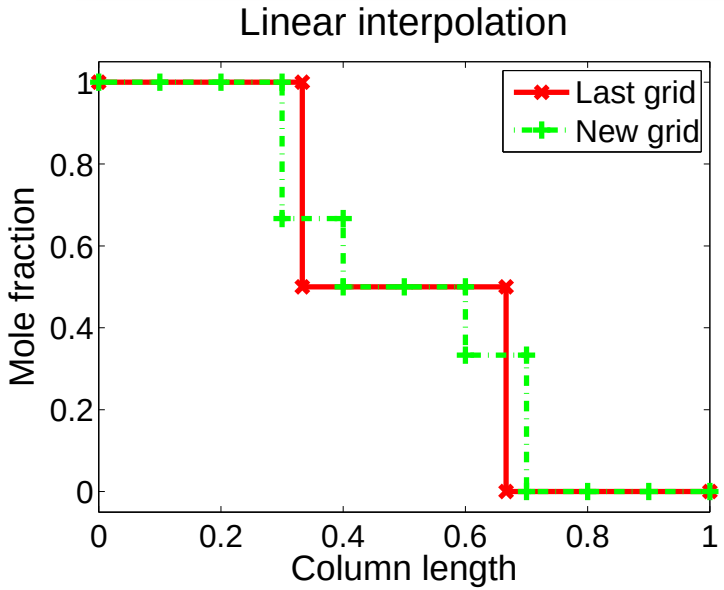
- Subsequent substitution: linear convergence
- ϵ extrapolation: quadratic convergence
- In the studied cases 35% faster convergence





Model and discretisation switch

- Start with the simplest case from the model hierarchy, e.g. isothermal and LDF, and coarse discretisation
- At CSS switch to the model and discretisation which accurately describe the problem
- Interpolate the coarse solution to the accurate description
- Simulate accurate model to CSS

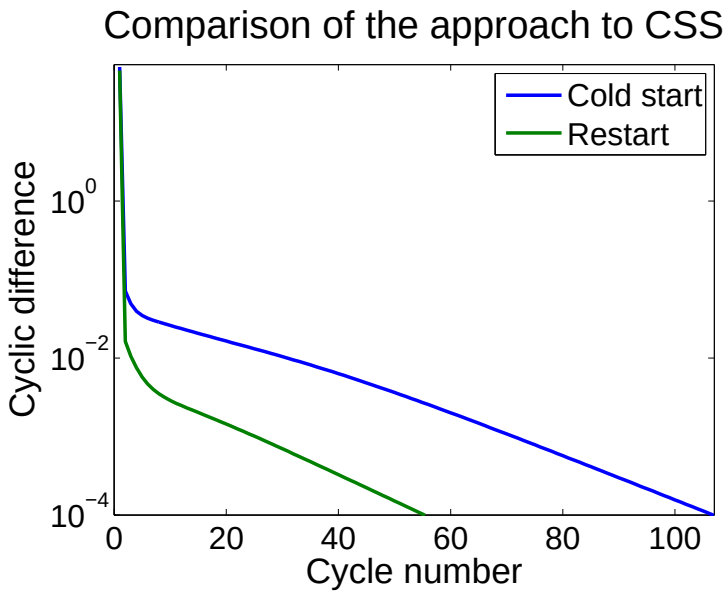


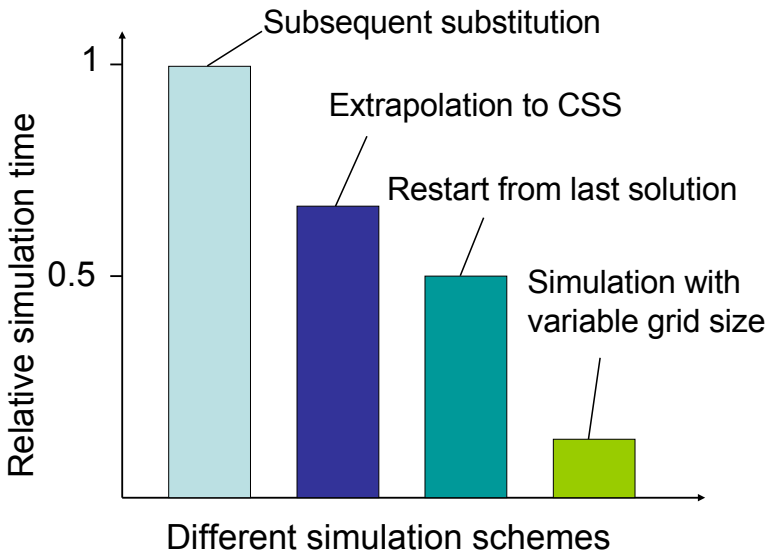
- Reduces simulation time by at least an order of magnitude
- Simpler than an adaptive scheme

Interpolation of the starting conditions

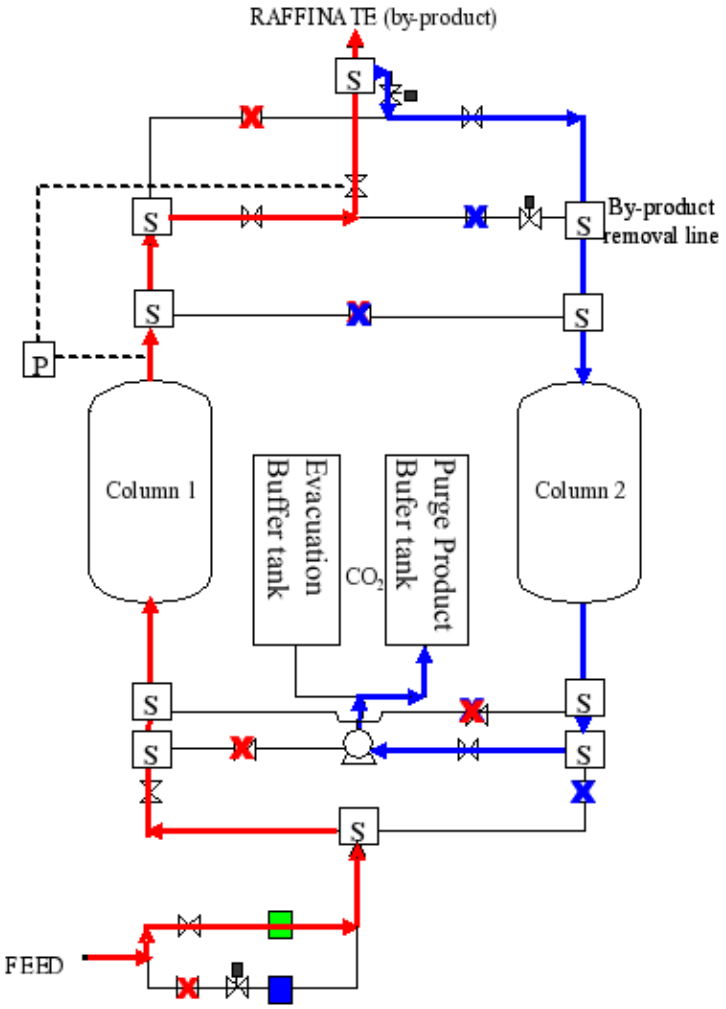
Restart from old solutions

- Interpolate the initial conditons from previous runs
- Can reduce the simulation time to CSS by $\sim 50\%$
- Especially important for parameter estimation because many runs of the simulation tool have to be performed

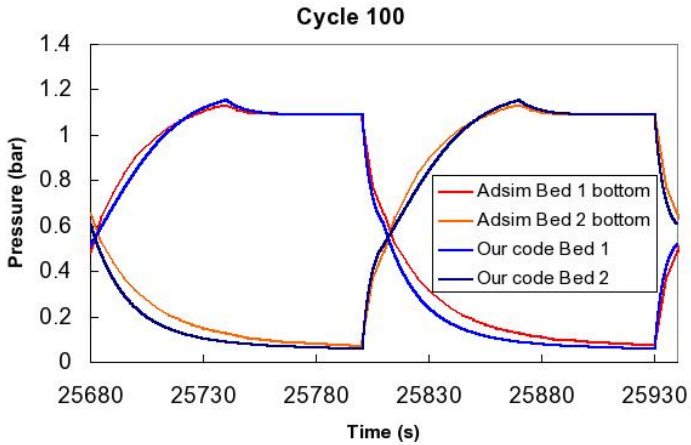




Preliminary results for the cycle simulator



- Column code is integrated into a cycle simulator
- Arbitrary sequence of steps
- Preliminary results are comparable with Adsim



Conclusion

- Model hierarchy describing the adsorption process in different levels of detail
- Simulation tool for cyclic adsorption processes with tailored discretisation schemes for the fluid and solid phase
- Several acceleration schemes which accelerate the convergence to CSS by at least an order of magnitude
- Applied the simulation tool to the simulation of the Dual Piston - PSA system

Future work

- Implement all models from the model hierarchy
- Automated choice of the correct model
- Parameter estimation from experimental data

Acknowledgement

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- EPSRC grant 'Innovative Gas Separations for Carbon Capture', EP/G062129/1
- EPSRC Science and Innovation Award 'Carbon Capture from Power Plant and Atmosphere', EP/F034520/1

Experimental work

275d, Total Reflux Dual Piston-PSA: a Novel Apparatus for Testing Adsorbent Materials
Wenli Dang, Daniel Friedrich, Maria-Chiara Ferrari and Stefano Brandani

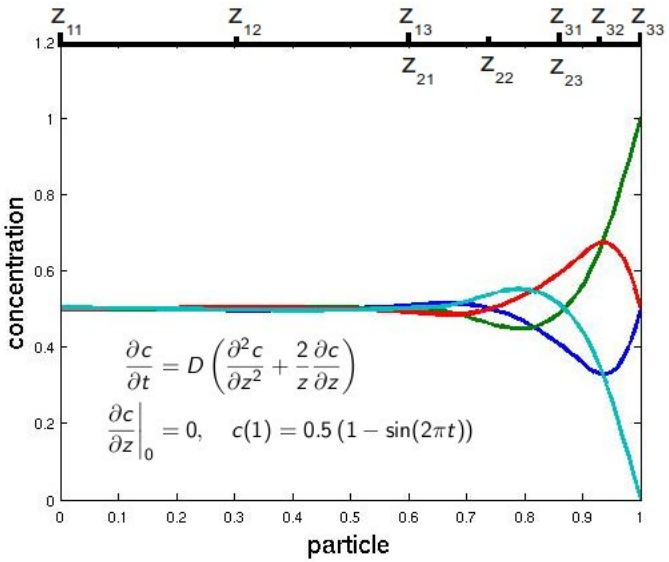
Questions?

Thank you for your attention!

Questions?

Orthogonal collocation on finite element method

- Approximate solution by trial functions $y_i = \sum_{k=0}^{N} y_k^{(i)} z^k$
- Substitute trial solution into original equations $R = F(y_i)$
- Set residual R to zero at the roots of orthogonal polynomials
- Split the domain into small elements to handle steep gradients in the solution



Implementation:

- Arbitrary number of elements
- Element order between 3 and 7
- Arbitrary position of element boundary