
DTU

Systematic Modeling and Simulation of One-Dimensional Packed-bed Processes

Marcus Johan Schytt, John Bagterp Jørgensen

Technical University of Denmark
Department of Applied Mathematics and Computer Science

26th Nordic Process Control Workshop 2026

Project introduction

MISSION GREEN FUELS

MissionGreenFuels

Innovation Fund Denmark's 2nd Green Innomission

- P2X for shipping, trucking, & aviation
- **Danish Goal:** Reduce CO_2e emissions
  - by 70% in 2030
  - by 100% in 2050

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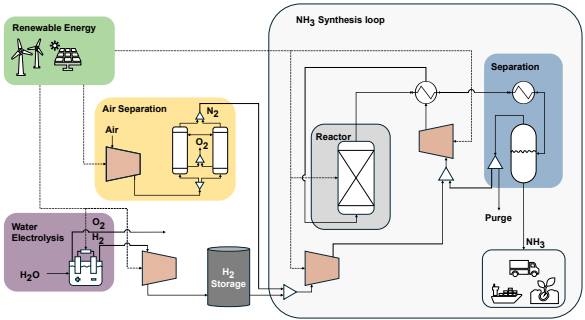

Figure: Power-to-Ammonia process overview

Fixed-bed processes

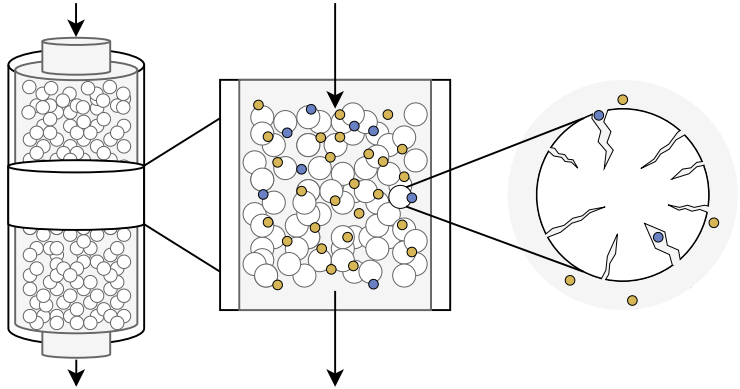

Figure: Transport phenomena present in a fixed-bed column

Column model

1D model

Axisymmetric column modeled on $\Omega = [0, L]$

- **Heterogenous** mass balances
- **Pseudo-homogeneous** energy balance

Variables

Given component set $\mathcal{C}$

- Mobile-phase concentrations $c = [c_\alpha]_{\alpha \in \mathcal{C}}$
- Solid-phase concentrations $q = [q_\alpha]_{\alpha \in \mathcal{C}}$
- Internal energy density u
- Temperature T
- Pressure P

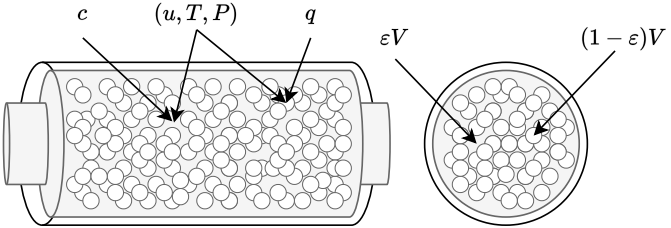

Figure: Heterogeneous and pseudo-homogeneous variables, and mobile/solid-phase volumes

Transport equations

First principles

The general principle of conservation states

$$\text{Acc} = \text{In} - \text{Out} + \text{Prod}$$

Differential form

For a density w with flux F and source term S

$$\partial_t w = -\partial_z F + S$$

Transport equations

First principles

The general principle of conservation states

$$\text{Acc} = \text{In} - \text{Out} + \text{Prod}$$

Mass balance

With molar flux N and production rate R

$$\partial_t c = -\partial_z N + R$$

Differential form

For a density w with flux F and source term S

$$\partial_t w = -\partial_z F + S$$

Energy balance

With energy flux E and rate of energy transfer Q

$$\partial_t u = -\partial_z E + Q$$

Thermodynamics
Bulk properties

We formulate our model using the thermodynamic properties

- Volume $\mathcal{V}(T, P, n)$
- Enthalpy $\mathcal{H}(T, P, n)$
- Internal energy $\mathcal{U}(T, P, n)$

*Specified by ideal-gas correlations, **with** or without **cubic modifications***

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Relations

The internal energy is recovered from the first two

$$\mathcal{U} = \mathcal{H} - P\mathcal{V}$$

Extensive properties are first-order homogeneous

$$\mathcal{V}(T, P, \lambda n) = \lambda \mathcal{V}(T, P, n)$$

Closures

Act as algebraic constraints for T & P

$$0 = \mathcal{U}(T, P, c) - u$$
$$0 = \mathcal{V}(T, P, c) - 1$$

General formulation

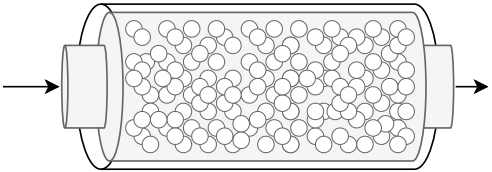


Figure: Fixed-bed adsorption column

Mass & energy balances

$$\partial_t c = -\partial_z N + R$$
$$\partial_t u = -\partial_z E + Q$$
$$0 = \mathcal{U}(T, P, c) - u$$
$$0 = \mathcal{V}(T, P, c) - 1$$

Constitutive laws

Molar fluxes

Advection & diffusion: $N = N^{adv} + N^{diff}$

$$N^{\text{adv}} = v(\partial_z P, c) \odot c$$
$$N^{\text{diff}} = -D(T, P, c) \odot \partial_z c$$

Energy flux

Convection & conduction: $E = E^{conv} + E^{cond}$

$$E^{\text{conv}} = \varepsilon \mathcal{H}(T, P, N^{\text{adv}})$$
$$E^{\text{cond}} = q^{\text{diff}} + q^{\text{therm}}$$

E^{cond} *splits into diffusive & Fourier contributions*

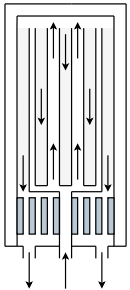
$$q^{\text{diff}} = -\partial_c \mathcal{H}(T, P, c)^T N^{\text{diff}}$$
$$q^{\text{therm}} = -\kappa(T, P, c)\partial_z T$$

Modeling case studies

Fixed-bed reactors

A Systematic Modeling Framework for Dynamic Simulation of Fixed-Bed Reactors

M. J. Schytt, J. B. Jørgensen
Comp. Chem. Eng. 2026

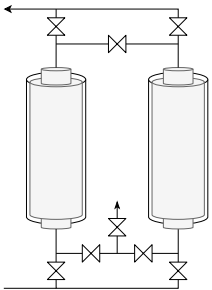


$$R = \frac{1-\varepsilon}{\varepsilon}\nu^T r, \qquad Q = aK(T' - T)$$

Adsorption columns

Modeling and Simulation of Nitrogen Generation by Pressure Swing Adsorption for Power-to-Ammonia

M. J. Schytt, L. T. Biegler, J. B. Jørgensen
Syst. Control Trans. 2026



$$R = -\frac{1-\varepsilon}{\varepsilon}\partial_t q, \qquad Q = -(1-\varepsilon)\sum_{\alpha\in\mathcal{C}} \Delta H_\alpha \partial_t q_\alpha$$

Semi-discrete model

PDAE system

Writing states $w = [c; q; u; T; P]$ *and BCs* p

$$M\dot{w} = -\partial_z F(w, \partial_z w, p) + S(w)$$

With mass matrix $M = \mathrm{diag}(1, 1, 1, 0, 0)$

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Finite volume discretization

Approximating $w \approx w_h^k$ *&* $\partial_z w \approx [w_h^{k+1} - w_h^k]/h$

$$\boldsymbol{M}_h \dot{w}_h^k = -\left[F_h^{k+\frac{1}{2}} - F_h^{k-\frac{1}{2}}\right]/h + S(w_h^k)$$

With mass matrix $\boldsymbol{M}_h = \mathrm{diag}(I, I, I, 0, 0)$

$$w_h^k \approx \frac{1}{h}\int_{z_h^{k-\frac{1}{2}}}^{z_h^{k+\frac{1}{2}}} w \, \mathrm{d}z$$

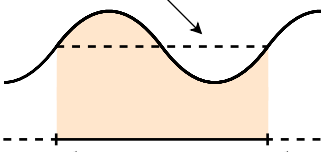

Figure: Volume-averaged state approximation

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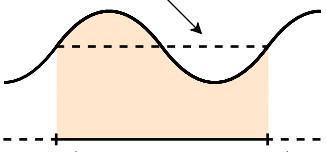

Figure: Volume-averaged state approximation

Index-1 DAE system

Writing $\boldsymbol{x} = [\boldsymbol{c}_h; \boldsymbol{q}_h; \boldsymbol{u}_h]$ *and* $\boldsymbol{y} = [\boldsymbol{T}_h; \boldsymbol{P}_h]$

$$\dot{\boldsymbol{x}} = f(\boldsymbol{x}, \boldsymbol{y}, p), \qquad \boldsymbol{x}(t_0) = \boldsymbol{x}_0$$
$$0 = g(\boldsymbol{x}, \boldsymbol{y}, p)$$

Alternatively $\boldsymbol{M}\dot{\boldsymbol{w}} = \boldsymbol{F}(\boldsymbol{w}, p)$ *with* $\boldsymbol{w} = [\boldsymbol{x}; \boldsymbol{y}]$

Steady-state simulation

Nonlinear system

At steady-state $\dot{\boldsymbol{w}} = 0$ the DAE reduces to

$$0 = \boldsymbol{F}(\boldsymbol{w}, p)$$

Newton's method

Compute sparse Jacobian $\partial \boldsymbol{F}$ via combined analytic and automatic differentiation

$$\partial \boldsymbol{F} \Delta \boldsymbol{w}^{\ell+1} = -\boldsymbol{F}(\boldsymbol{w}^{\ell})$$
$$\boldsymbol{w}^{\ell+1} = \boldsymbol{w}^{\ell} + \Delta \boldsymbol{w}^{\ell+1}$$

Perhaps using a trust-region method for ill-conditioned problems

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Ammonia synthesis

Isothermal direct-cooled reactor case study

$$\mathrm{N_2} + 3\mathrm{H_2} \leftrightharpoons 2\mathrm{NH_3}, \quad \Delta H_{298\mathrm{K}} \approx -92 \text{ [kJ/mol]}$$

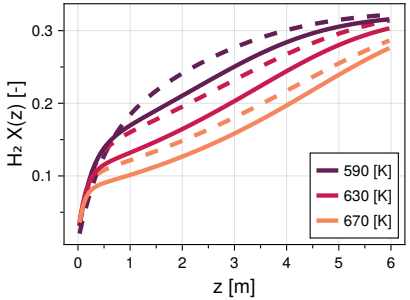

Figure: H_2 conversion profiles. Ideal (- -), SRK (–).

Parametric sensitivity analysis

Ammonia synthesis

Isothermal direct-cooled reactor case study

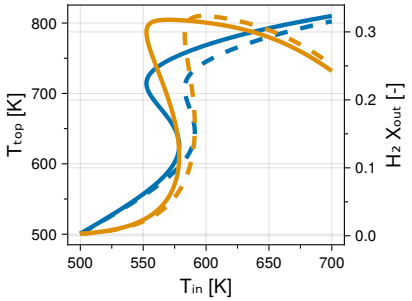

Figure: H_2 conversion and top temperature folds.
Ideal (- -), SRK (–).

Solving $w(p)$ repeatedly for different parameters p

$$0 = \boldsymbol{F}(\boldsymbol{w}, p), \qquad p \in [p_{\text{min}}, p_{\text{max}}]$$

Parametric sensitivity analysis

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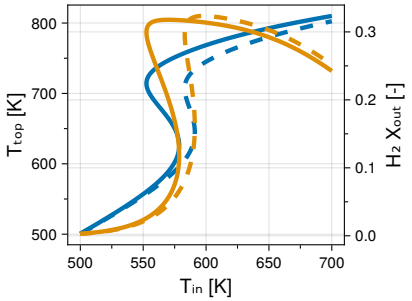

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$$0 = \boldsymbol{F}(\boldsymbol{w}, p), \qquad p \in [p_{\text{min}}, p_{\text{max}}]$$

Pseudo arc-length continuation

Trace curve through folds $(\boldsymbol{w}(s), p(s))$ by solving

$$0 = \boldsymbol{F}(\boldsymbol{w}^{m+1}, p^{m+1}),$$
$$0 = (\boldsymbol{w}^{m+1} - \boldsymbol{w}^{m})^T \xi_{\boldsymbol{w}}^{m} + (p^{m+1} - p^{m})\xi_{p}^{m} - \Delta s^{m}$$

orthogonal to the approximate tangent obtained by

$$(\boldsymbol{w}^{m}, p^{m}) = (\boldsymbol{w}^{m-1}, p^{m-1}) + \Delta s^{m-1}\xi^{m}$$

Dynamic simulation

Ammonia synthesis

Isothermal direct-cooled reactor case study

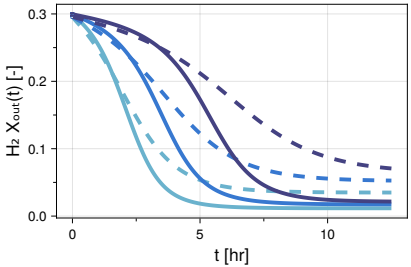

Figure: Steps $-\Delta T = 5, 10, 20$ [K] from extinction.
Ideal (- -), SRK (–).

Dynamic simulation

Ammonia synthesis

Isothermal direct-cooled reactor case study

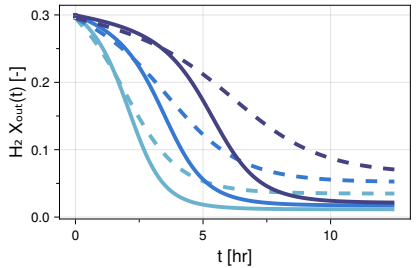

Figure: Steps $-\Delta T = 5, 10, 20$ [K] from extinction. Ideal (- -), SRK (–).

Diagonally implicit Runge-Kutta

With s stages and Butcher tableau $(a_{i,j}, b_i)$

$$\boldsymbol{M}\boldsymbol{W}_{n,i} = \boldsymbol{M}\boldsymbol{w}_n + h_n \sum_{j=1}^{s} a_{i,j} F(\boldsymbol{W}_{n,j}, p)$$

$$\boldsymbol{M}\boldsymbol{w}_{n+1} = \boldsymbol{M}\boldsymbol{w}_n + h_n \sum_{i=1}^{s} b_i F(\boldsymbol{W}_{n,i}, p)$$

*DIRK methods are **very** efficient for stiff problems*

- Solve sequentially for each stage
- High-order, A,L-stable, stiffly accurate
- Adaptivity using embedded methods
- Efficient direct sparse linear algebra

Comprises e.g., implicit Euler and trapezoidal rule

Nitrogen generation

Pressure swing adsorption air (N_2, O_2, Ar) separation case study

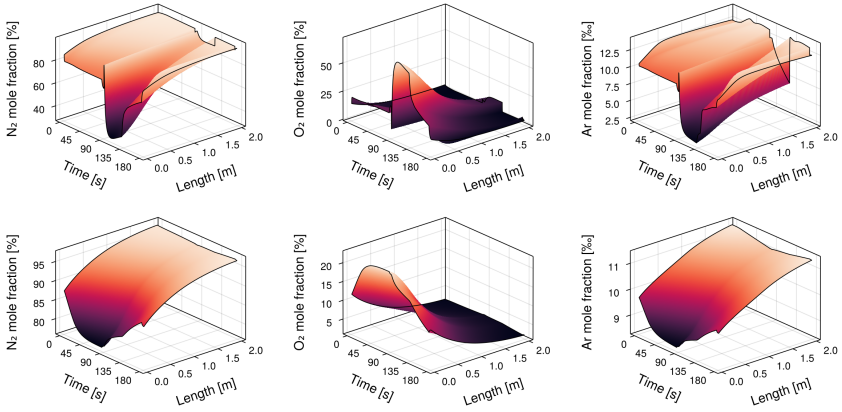

Figure: Fraction profiles (top: mobile, bottom: stationary) through the 8-step PSA cycle at CSS

Dynamic simulation performance

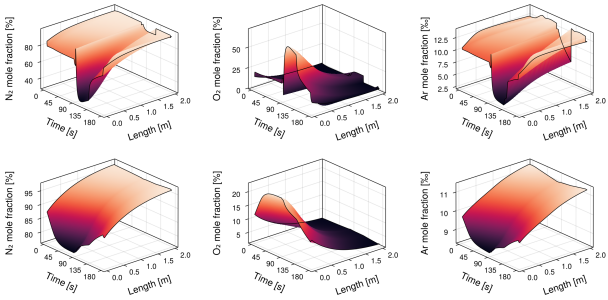

Figure: Fraction profiles (top: mobile, bottom: stationary) through the 8-step PSA cycle at CSS

Method	Statistic	Tol=10^{-2}	Tol=10^{-4}	Tol=10^{-6}	Tol=10^{-8}
ESDIRK2(1)3L[2]SA	Steps [#]	178	266	1589	14542
2nd order 3 stages	Time [min:sec]	**00:04**	**00:06**	00:31	04:43
ESDIRK3(2)5L[2]SA	Steps [#]	143	163	282	725
3rd order 5 stages	Time [min:sec]	00:05	**00:06**	**00:10**	00:27
ESDIRK4(3)7L[2]SA	Steps [#]	**129**	**144**	**233**	**377**
4th order 7 stages	Time [min:sec]	00:06	00:07	00:11	**00:20**

Cyclic steady-state simulation

Fixed-point problem

CSS is computed as a fixed-point

$$\Phi(\boldsymbol{x}, p) = \boldsymbol{x}$$

of the Runge-Kutta integrator Φ

Root-finding problem

Reformulated as a nonlinear system

$$\underbrace{\Phi(\boldsymbol{x}, p) - \boldsymbol{x}}_{R(\boldsymbol{x},p)} = 0$$

Solve by fixed-point or Newton iterations?

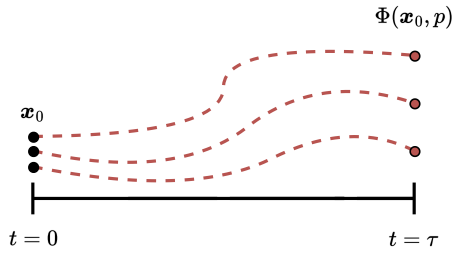

Figure: Shooting of the initial state $\boldsymbol{x}_0$ via Φ

Cyclic steady-state simulation performance

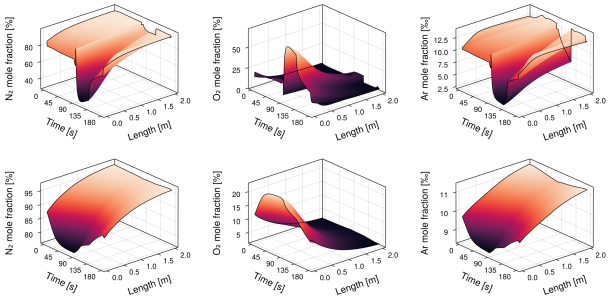

Figure: Fraction profiles (top: mobile, bottom: stationary) through the 8-step PSA cycle at CSS

Statistic	Picard	AA	Newton
Iterations [#]	1123	416	**3**
Time [hr:min]	01:19	00:23	**00:02**

Parametric sensitivity analysis: Revisited

Solve CSS repeatedly (using continuation) for different BCs p tracking performance metrics

$$R(\boldsymbol{x}, p) = 0$$

Cycle performance

Track N_2 purity and recovery as a function of feed velocity

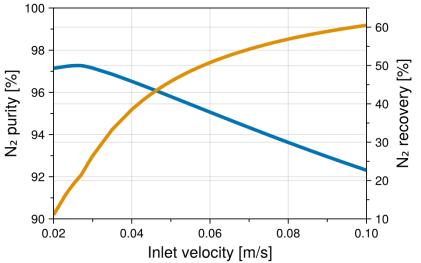

Figure: N_2 purity and recovery versus inlet velocity

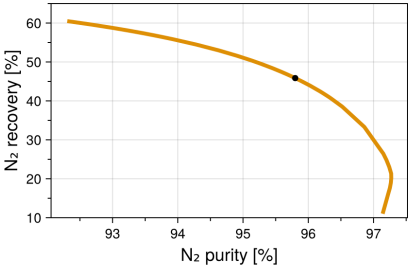

Figure: N_2 purity-recovery trade-off

Acknowledgements

Funded by MissionGreenFuels project DynFlex (no. 1150-00001B)

In Collaboration with Lorenz Biegler, Carnegie Mellon University

Thank you for your attention!

Questions?