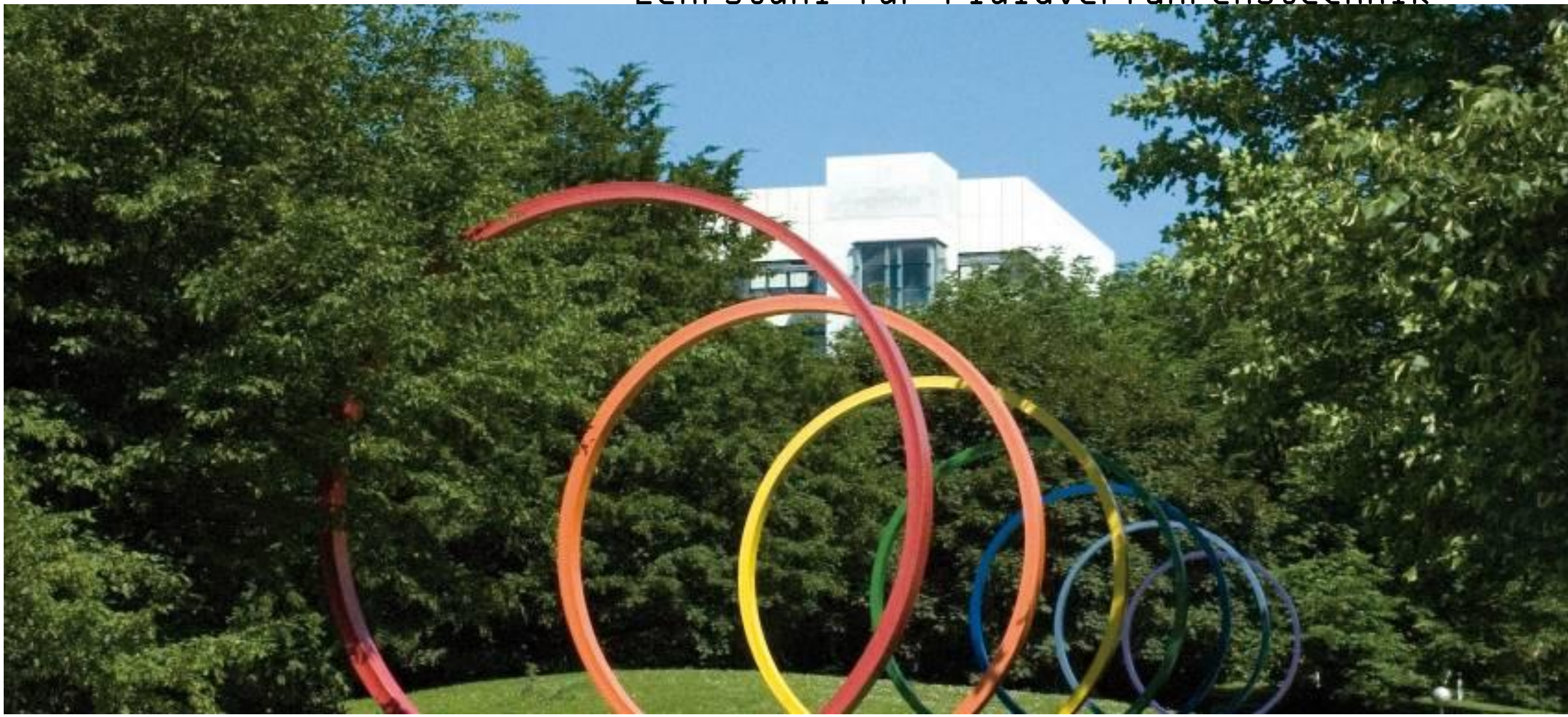
technische universität dortmund

Fakultät Bio- und Chemieingenieurwesen
Lehrstuhl für Fluidverfahrenstechnik

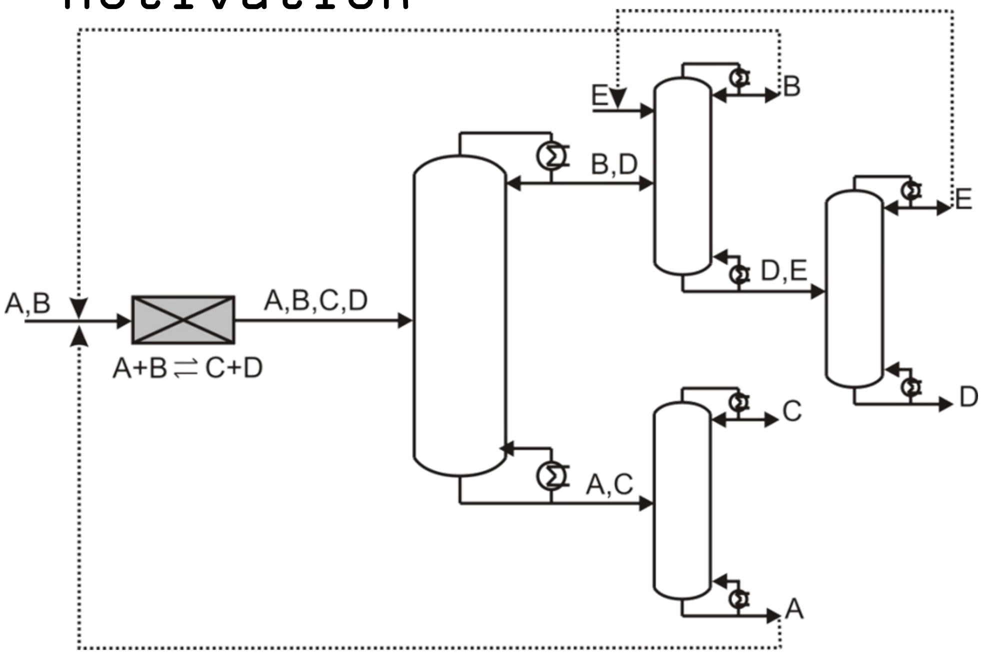


Experimental Investigation, Analysis and Optimisation of Hybrid Separation Processes

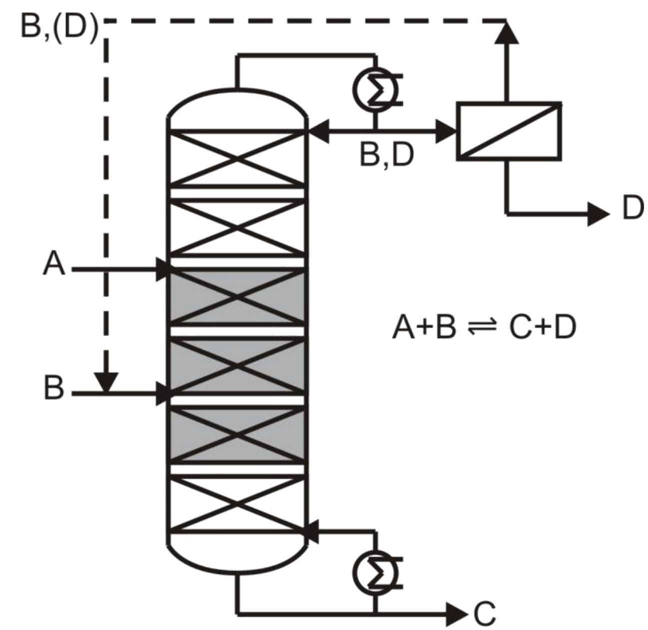
EFCE Excellence Award in Process Intensification 2009

Carsten Buchaly

Motivation

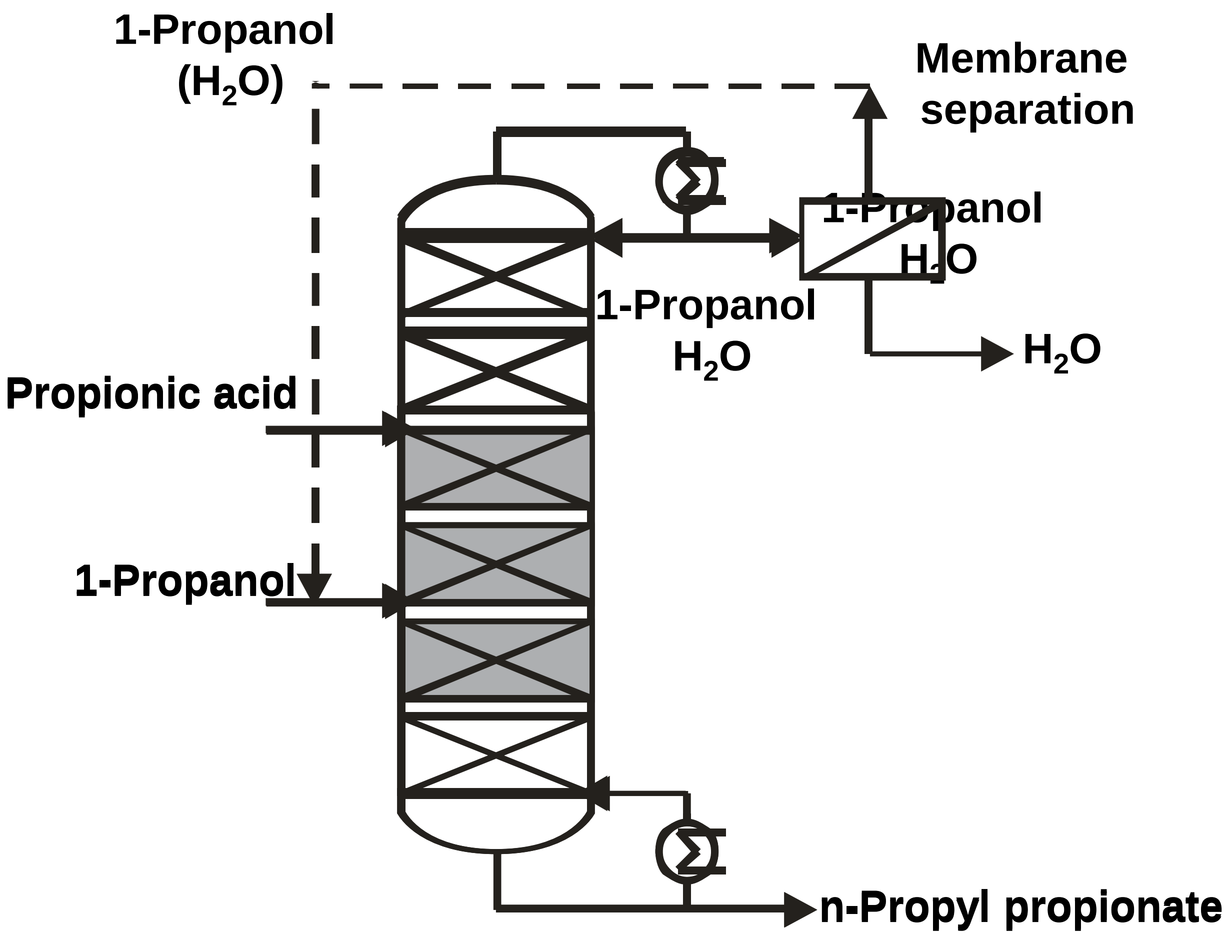


- Reduced volumina of apparatuses / capital costs
- Less energy consumption
- No auxiliary components required



- Strong interactions of both unit operations
- Detailed process know-how necessary
- Less experience existent

Process Description



Reactive Distillation

- Integration of reaction and separation
- Increased conversion and selectivity

Membrane Separation

- 1-Propanol recovery
- High selectivity
- Independent on VLE

Methodology

Hybrid Process

(reactive distillation + membrane separation)

Theory
- Experimental design
- Process analysis
- Optimisation

Experiments
- Pilot-scale
- Start-up behaviour
- Process know-how

Methodology

Hybrid Process

(reactive distillation + membrane separation)

Theory
- Experimental design
- Process analysis
- Optimisation

Experiments
- Pilot-scale
- Start-up behaviour
- Process know-how

Unit operation 1

(reactive distillation)

Unit operation 2

(membrane separation)

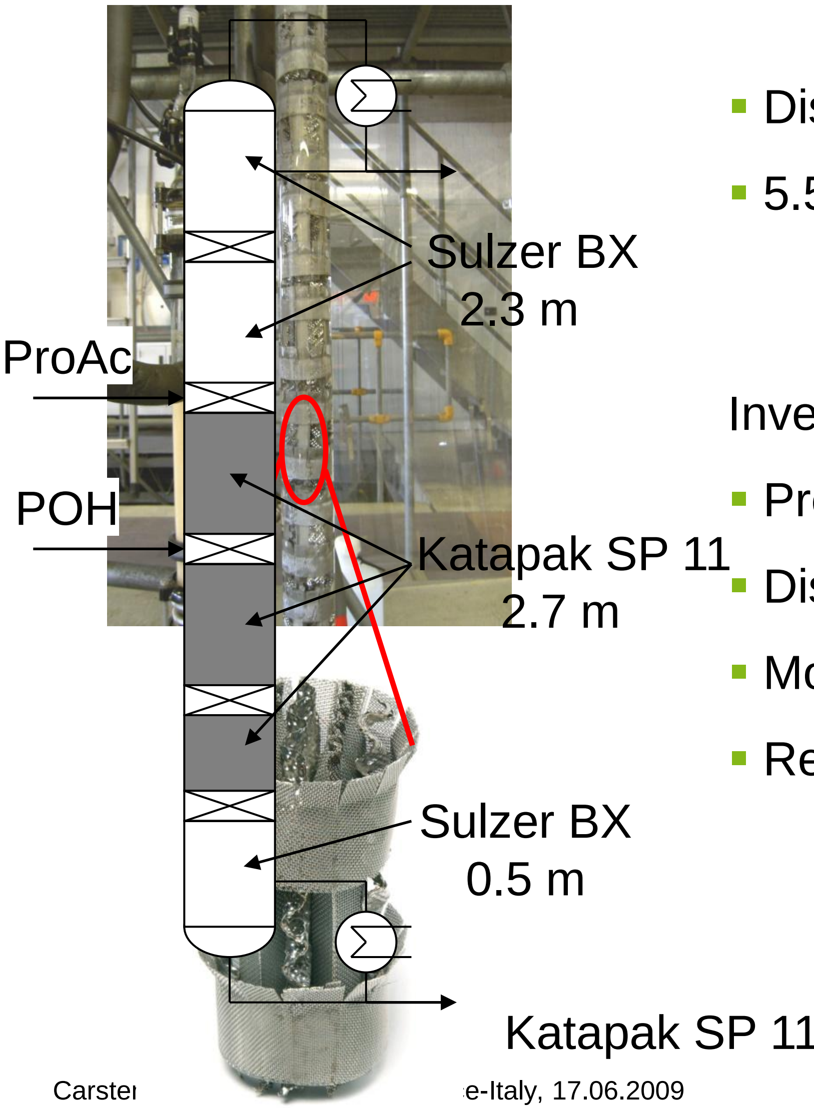
Experiments
- Pilot-scale
- Operational parameters
- Model validation

Theory
- Experimental design
- Process analysis

Experiments
- Lab-scale
- Separation characteristics
- Model parameters

Experiments
- Pilot-scale
- Scale-up

Reactive distillation: pilot-scale plant

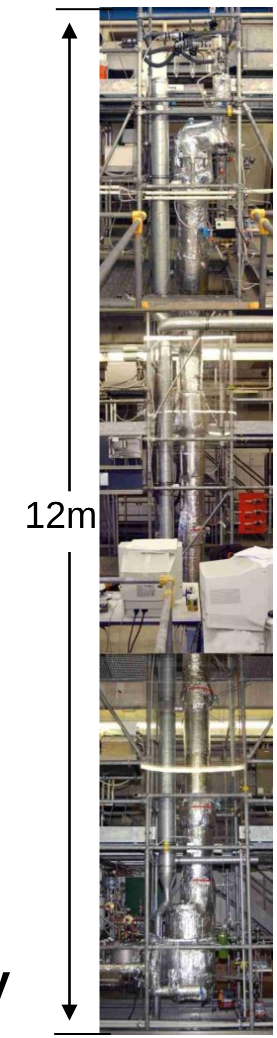


- Distillation column (DN 50)
- 5.5 m packing height

Investigated operating parameters

- Pressure: atmospheric
- Distillate-to-feed ratio: 0.33 - 0.45
- Molar feed ratio (χ): 2.1 - 2.5
- Reflux ratio (RR): 2.0 - 4.0

Database of 15 successfully realised RD experiments



Vapour permeation: pilot-scale plant

- Steady state vapour permeation experiments
- Connection with reactive distillation column to hybrid separation process
- Sulzer Pervap 2201(D) with A_{Memb} = 0.5m²

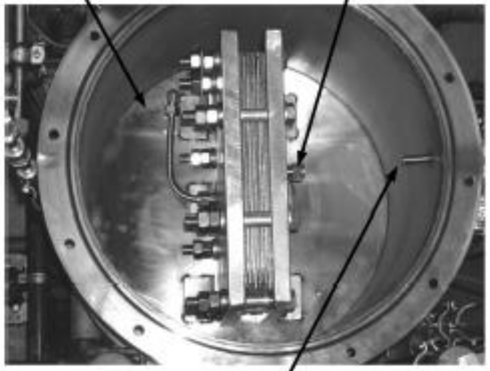


Plate-and-frame module

Buchaly, Kreis, Górak (2007): Chemical Engineering and Processing, 46, p. 790-799

Vapour permeation: pilot-scale plant

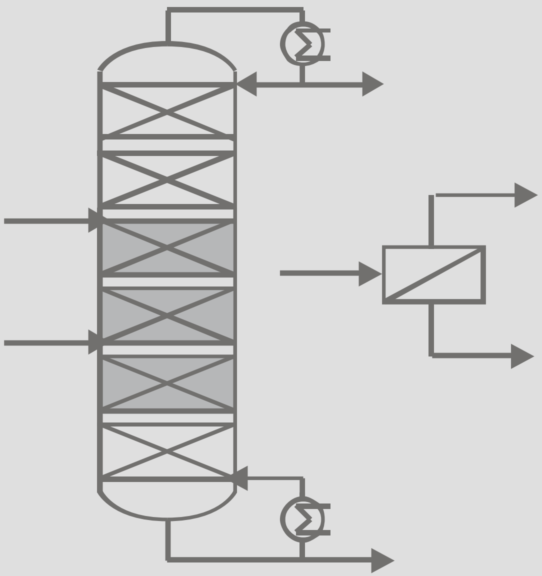
- Steady state vapour permeation experiments
- Connection with reactive distillation column to hybrid separation process
- Sulzer Pervap 2201(D) with A_{Memb} = 0.5m²

Binary system 1-propanol/water

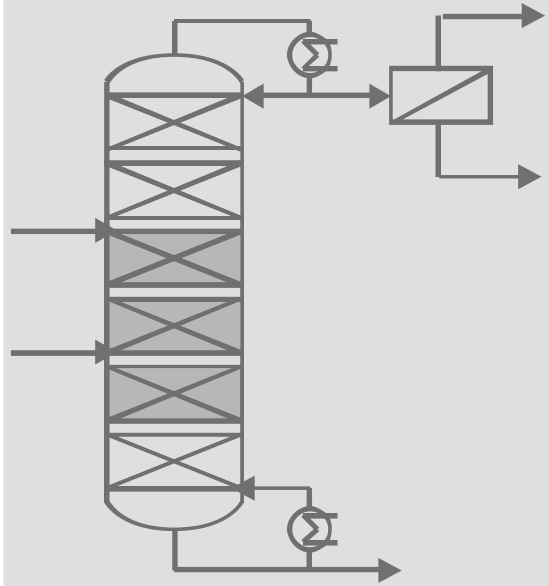
- Water concentration: 13 - 30 wt.-%
- Feed temperature: 92.0 - 98.7 °C
- Feed pressure: atmospheric
- Permeate pressure: 30 - 70 mbar

Buchaly, Kreis, Górak (2007): Chemical Engineering and Processing, 46, p. 790-799

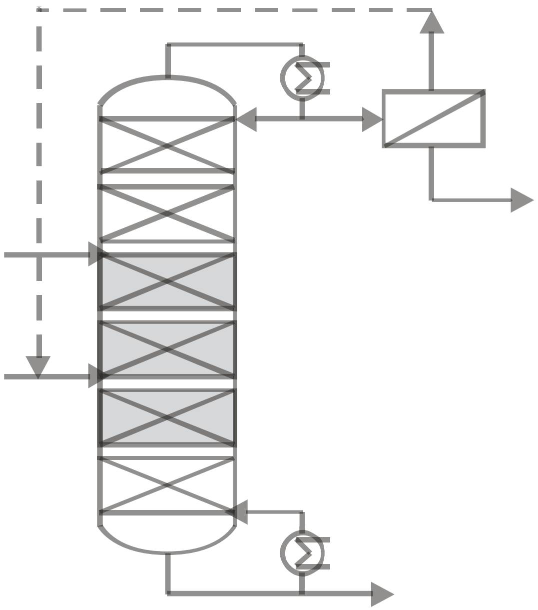
Hybrid separation process: experimental investigations



RD and VP separated

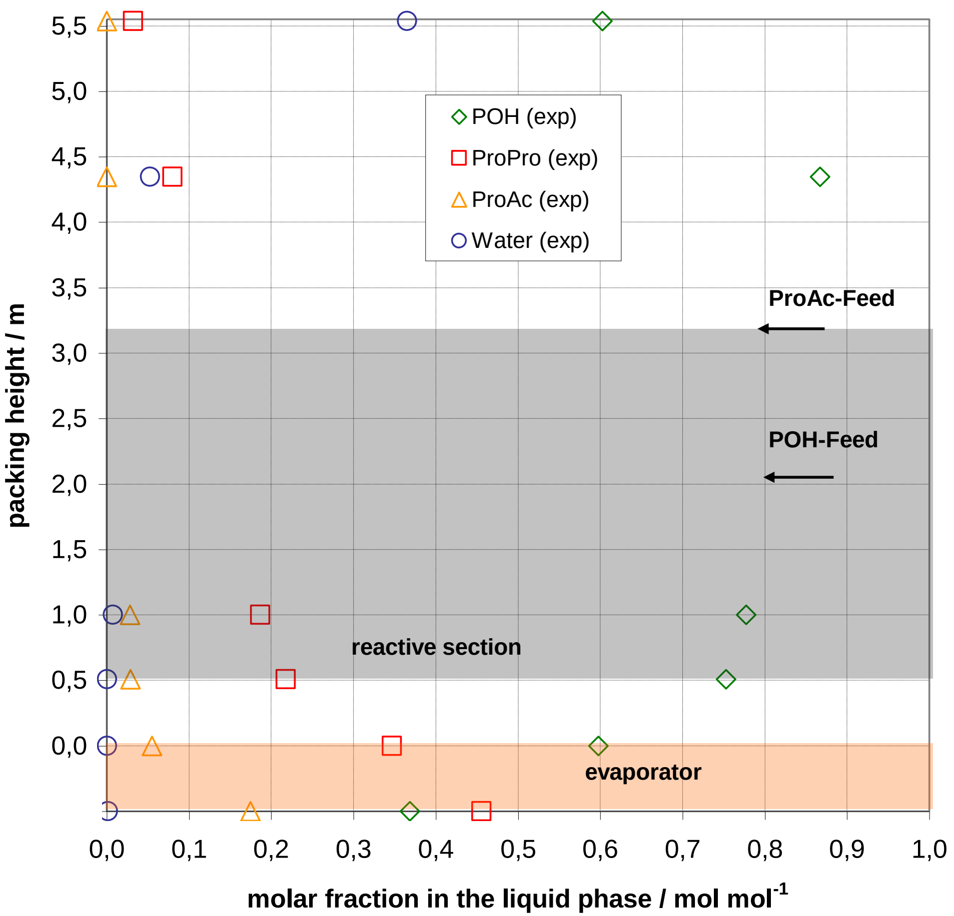


sequential configuration



„closed recycle“

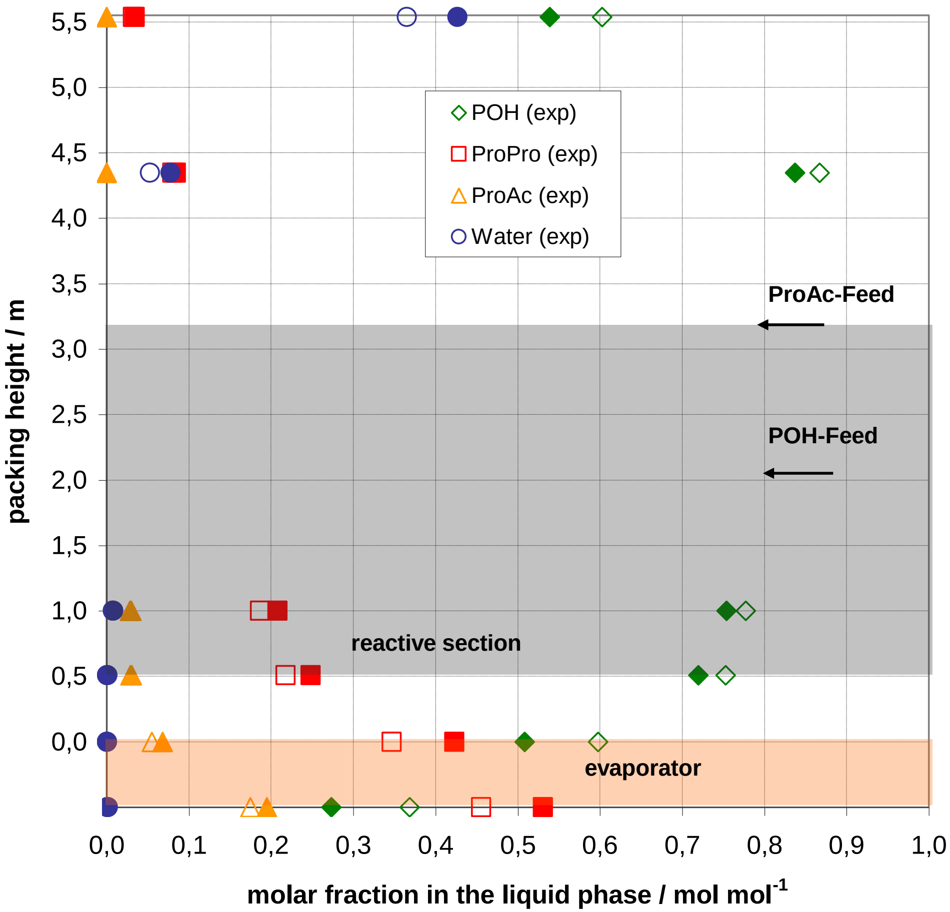
Hybrid separation process: experimental investigations



Buchaly, Kreis, Górak (2008): Chemie Ingenieur Technik, 80, p. 145-156

- Successful experiment with coupled unit operations

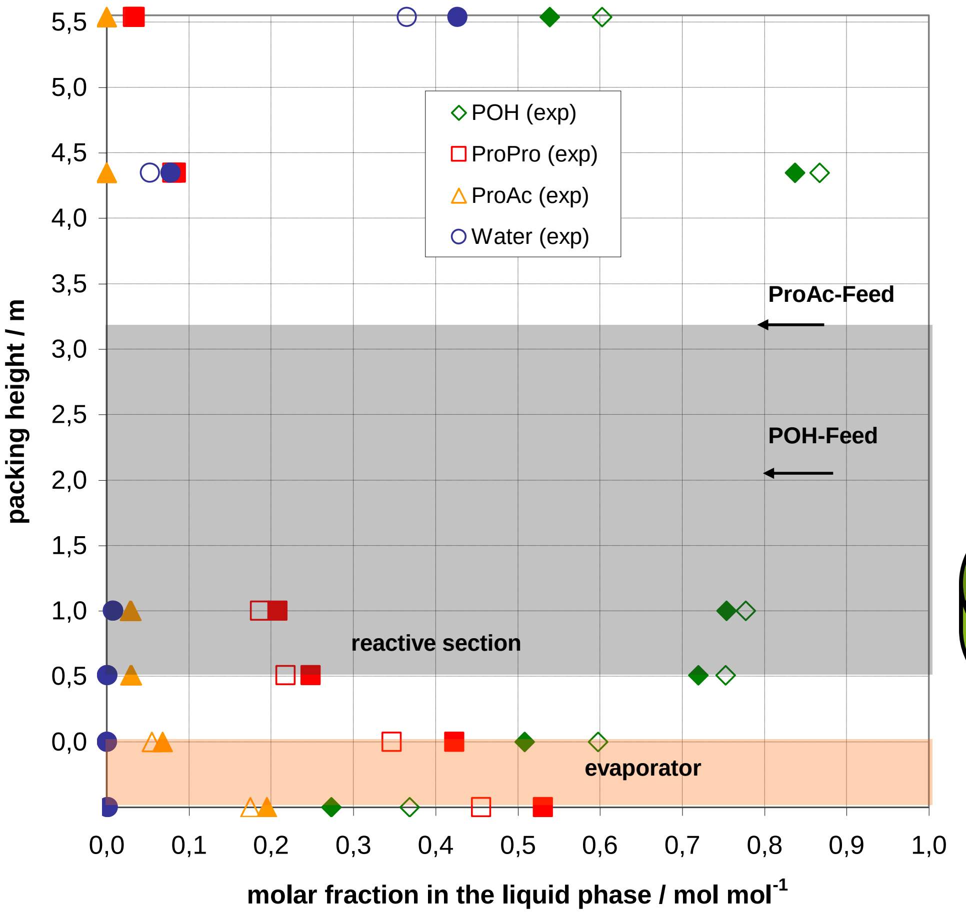
Hybrid separation process: experimental investigations



Buchaly, Kreis, Górak (2008): Chemie Ingenieur Technik, 80, p. 145-156

- Successful experiment with coupled unit operations
- Increase of ester concentration in the bottom
- Change of distillate composition

Hybrid separation process: experimental investigations



Buchaly, Kreis, Górak (2008): Chemie Ingenieur Technik, 80, p. 145-156

- Successful experiment with coupled unit operations
- Increase of ester concentration in the bottom
- Change of distillate composition

Strong interactions between both unit operations

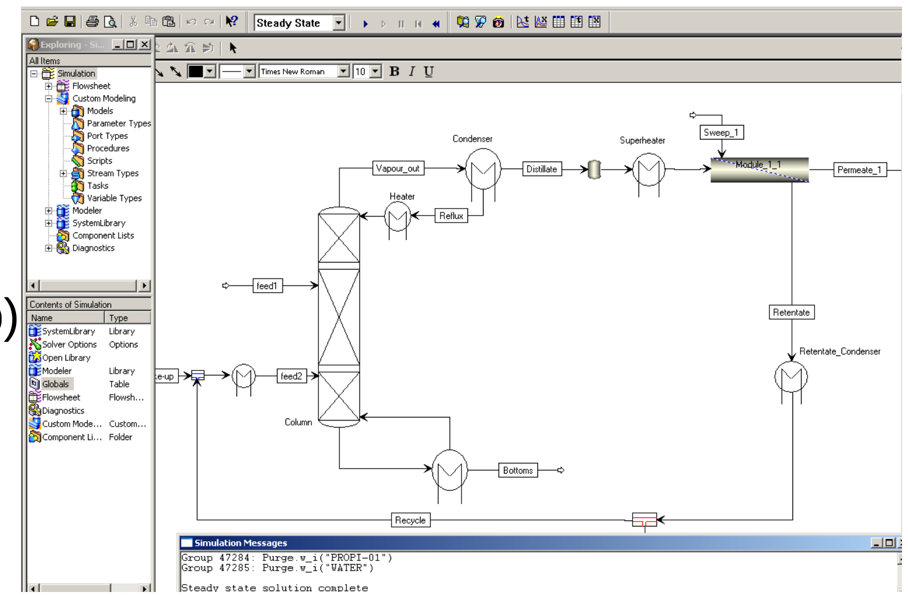
Modelling approach

Reactive distillation based on non-equilibrium stage model

- Multicomponent mass transfer (Stefan-Maxwell)
- Packing specific correlations (hold-up, Δp, $k_g a$, $k_l a$)
- Steady state and dynamic process simulation

Vapour permeation based on „Solution-Diffusion-Model“

- Polarisation effects (c,T)
- Hydrodynamics (co-, counter-current; Δp)
- Membrane materials



Implemented in Aspen Custom Modeler^{TM} (ACM)

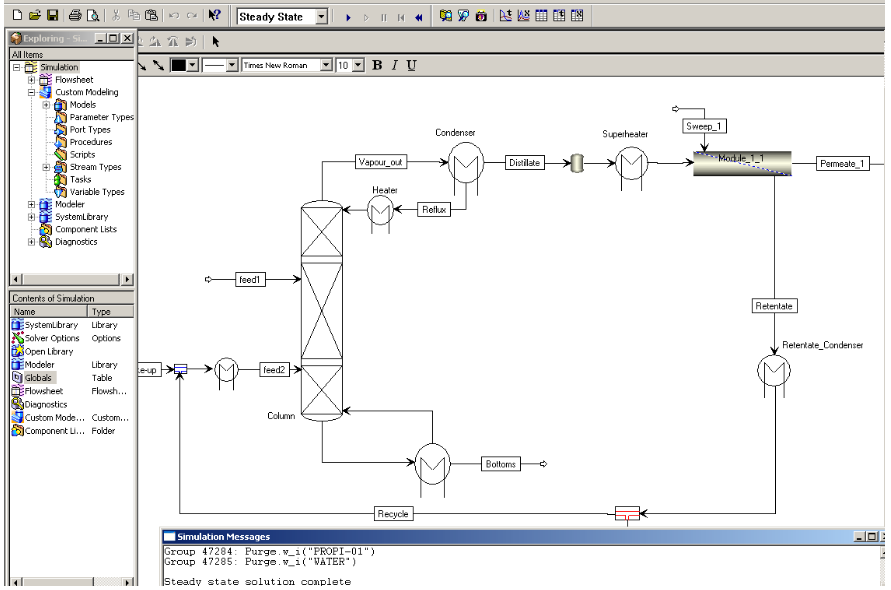
Modelling approach

Reactive distillation based on non-equilibrium stage model

- Multicomponent mass transfer (Stefan-Maxwell)
- Packing specific correlations (hold-up, Δp, $k_g a$, $k_l a$)
- Steady state and dynamic process simulation

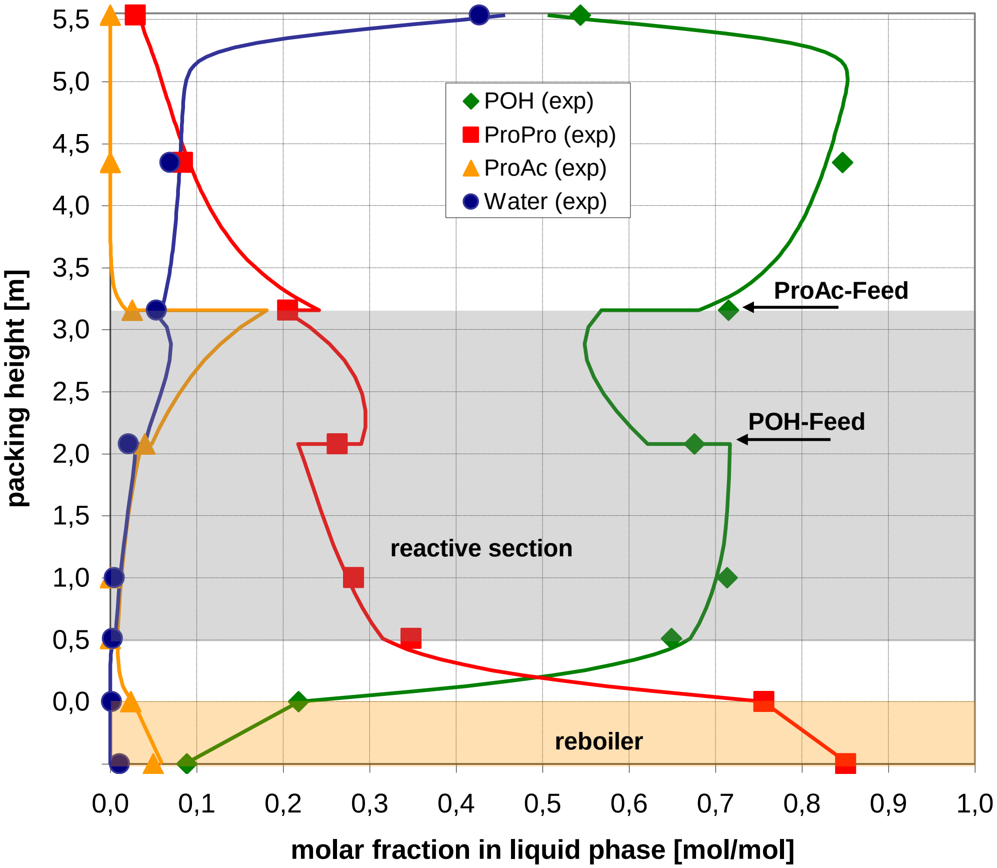
Vapour permeation based on „Solution-Diffusion-Model“

- Polarisation effects (c,T)
- Hydrodynamics (co-, counter-current; Δp)
- Membrane materials



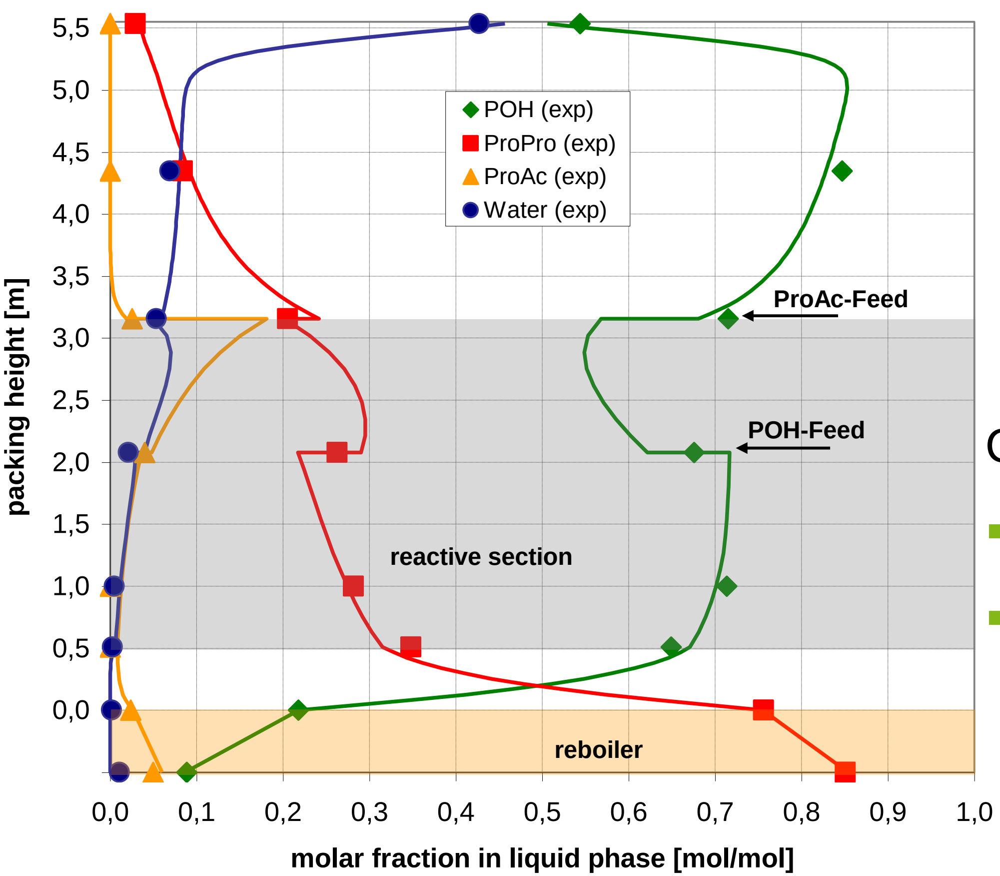
Implemented in Aspen Custom Modeler™ (ACM)

Reactive distillation: model validation



Feed:	2,0 kg/h
$\chi_{POH/ProAc}$:	2,068
RR:	2,49
Distillate:	0,85 kg/h

Reactive distillation: model validation



Feed:	2,0 kg/h
$\chi_{POH/ProAc}$:	2,068
RR:	2,49
Distillate:	0,85 kg/h

Correlations for column internals:

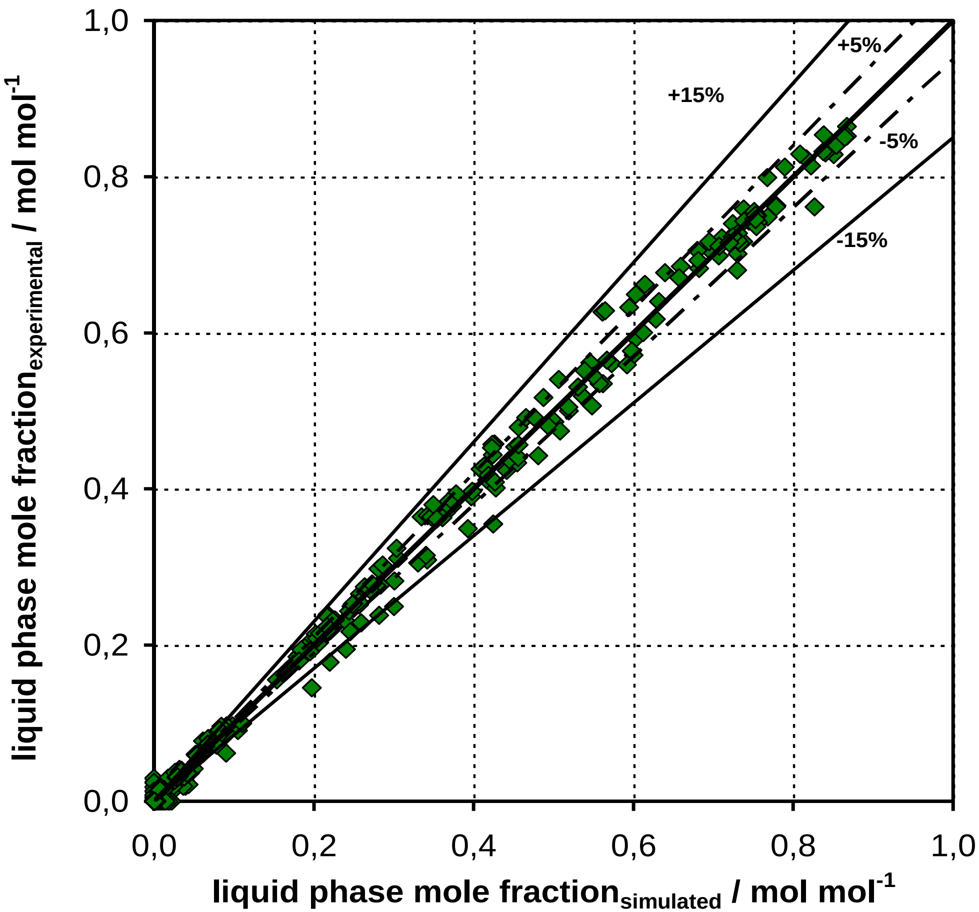
- Sulzer BX -> Bravo et al.*; Rocha et al.**
- Katapak-SP -> Brunazzi***

* Bravo et al. (1985): Hydr. Proc., 1, p. 91–95

** Rocha et al. (1993): Ind. Eng. Chem. Res., 32, p. 641–651

*** Brunazzi, Viva (2006): Distillation & Absorption 2006

Reactive distillation: model validation



Feed:	2,0 kg/h
$\chi_{POH/ProAc}$:	2,068
RR:	2,49
Distillate:	0,85 kg/h

Correlations for column internals:

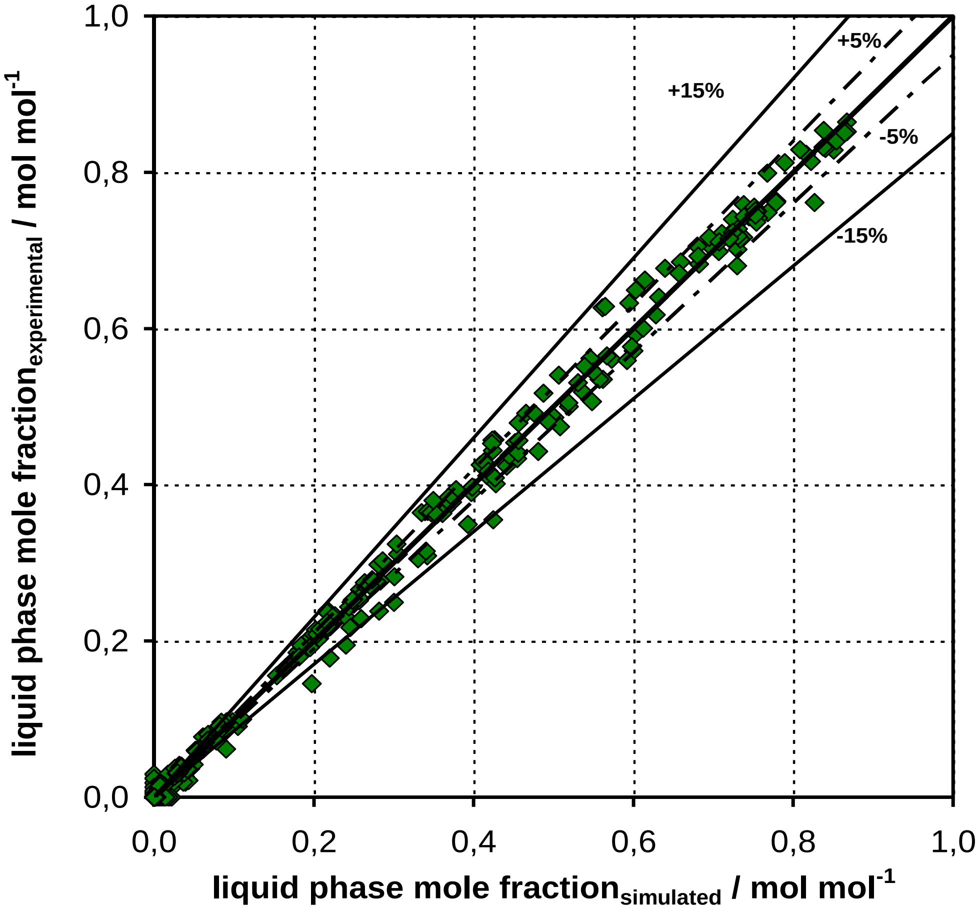
- Sulzer BX -> Bravo et al.*; Rocha et al.**
- Katapak-SP -> Brunazzi***

* Bravo et al. (1985): Hydr. Proc., 1, p. 91–95

** Rocha et al. (1993): Ind. Eng. Chem. Res., 32, p. 641–651

*** Brunazzi, Viva (2006): Distillation & Absorption 2006

Reactive distillation: model validation



Feed:	2,0 kg/h
$\chi_{POH/ProAc}$:	2,068
RR:	2,49
Distillate:	0,85 kg/h

Correlations for column internals:

- Sulzer BX -> Bravo et al.*; Rocha et al.**
- Katapak-SP -> Brunazzi***

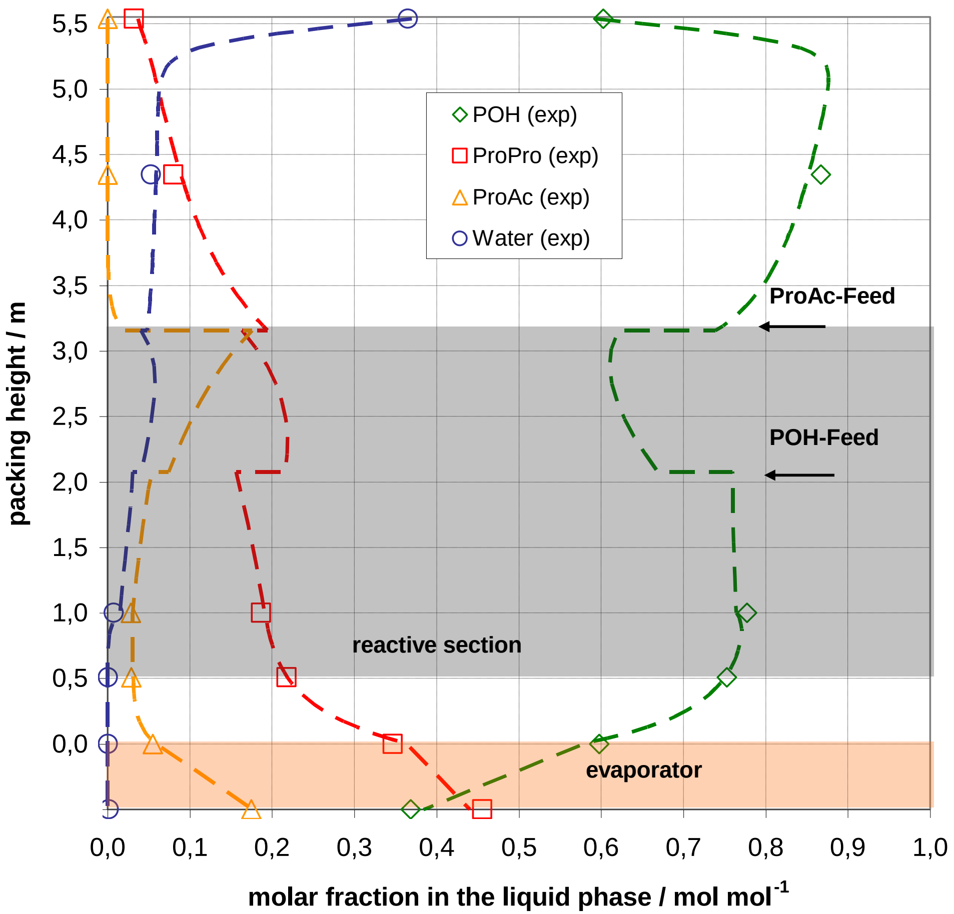
Model validation successful

* Bravo et al. (1985): Hydr. Proc., 1, p. 91–95

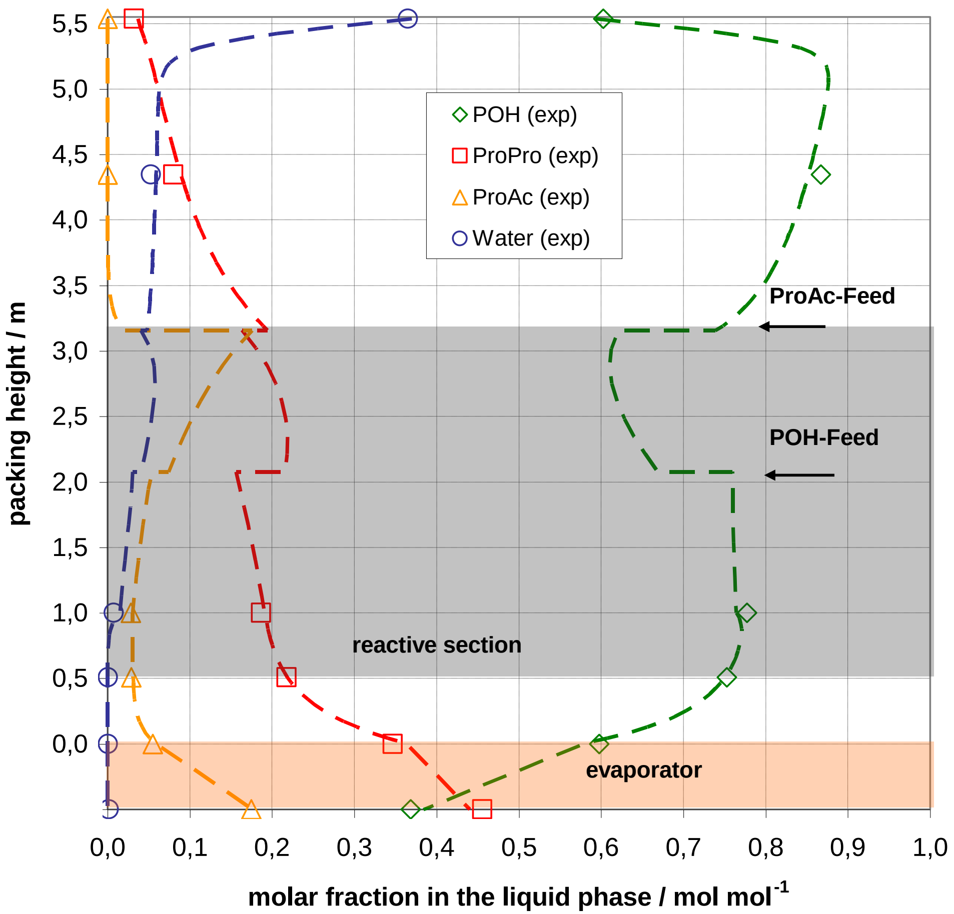
** Rocha et al. (1993): Ind. Eng. Chem. Res., 32, p. 641–651

*** Brunazzi, Viva (2006): Distillation & Absorption 2006

Hybrid separation process: model validation



Hybrid separation process: model validation

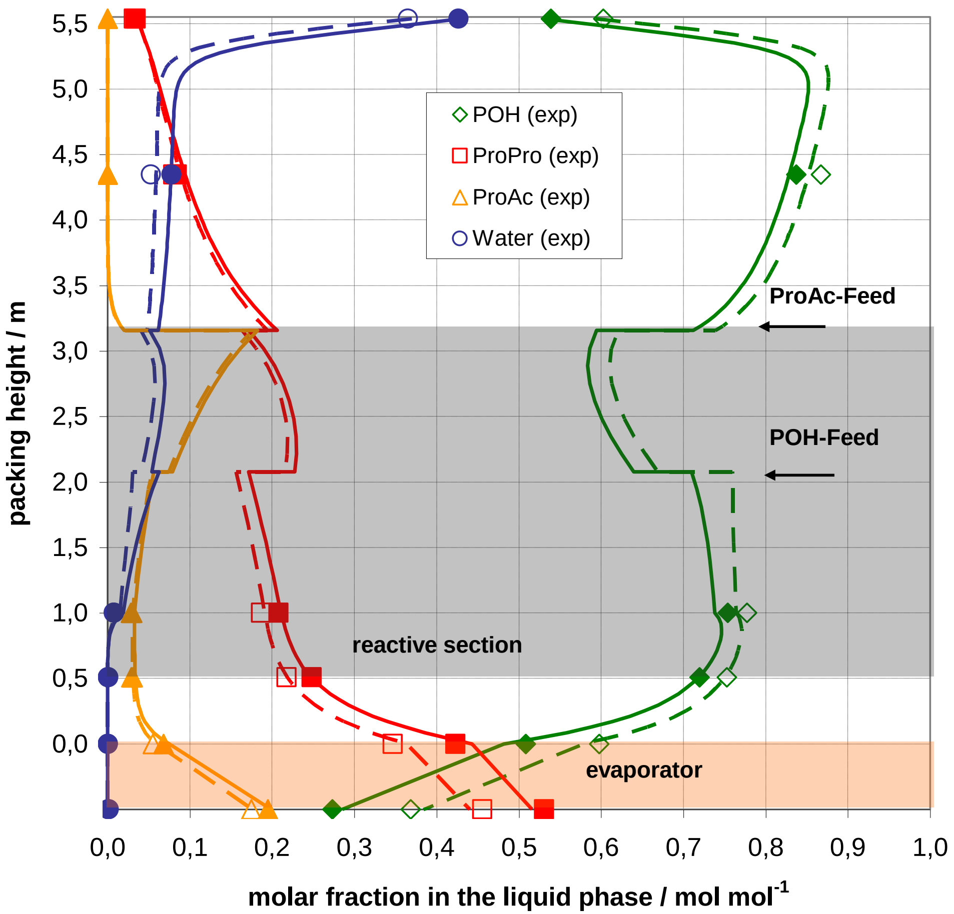


Sequential configuration

$X_{ProAc,exp}$ = 74.4 % $X_{ProAc,sim}$ = 73.6 %

$X_{POH,exp}$ = 29.8 % $X_{POH,sim}$ = 29.4 %

Hybrid separation process: model validation



Sequential configuration

$X_{ProAc,exp}$ = 74.4 % $X_{ProAc,sim}$ = 73.6 %

$X_{POH,exp}$ = 29.8 % $X_{POH,sim}$ = 29.4 %

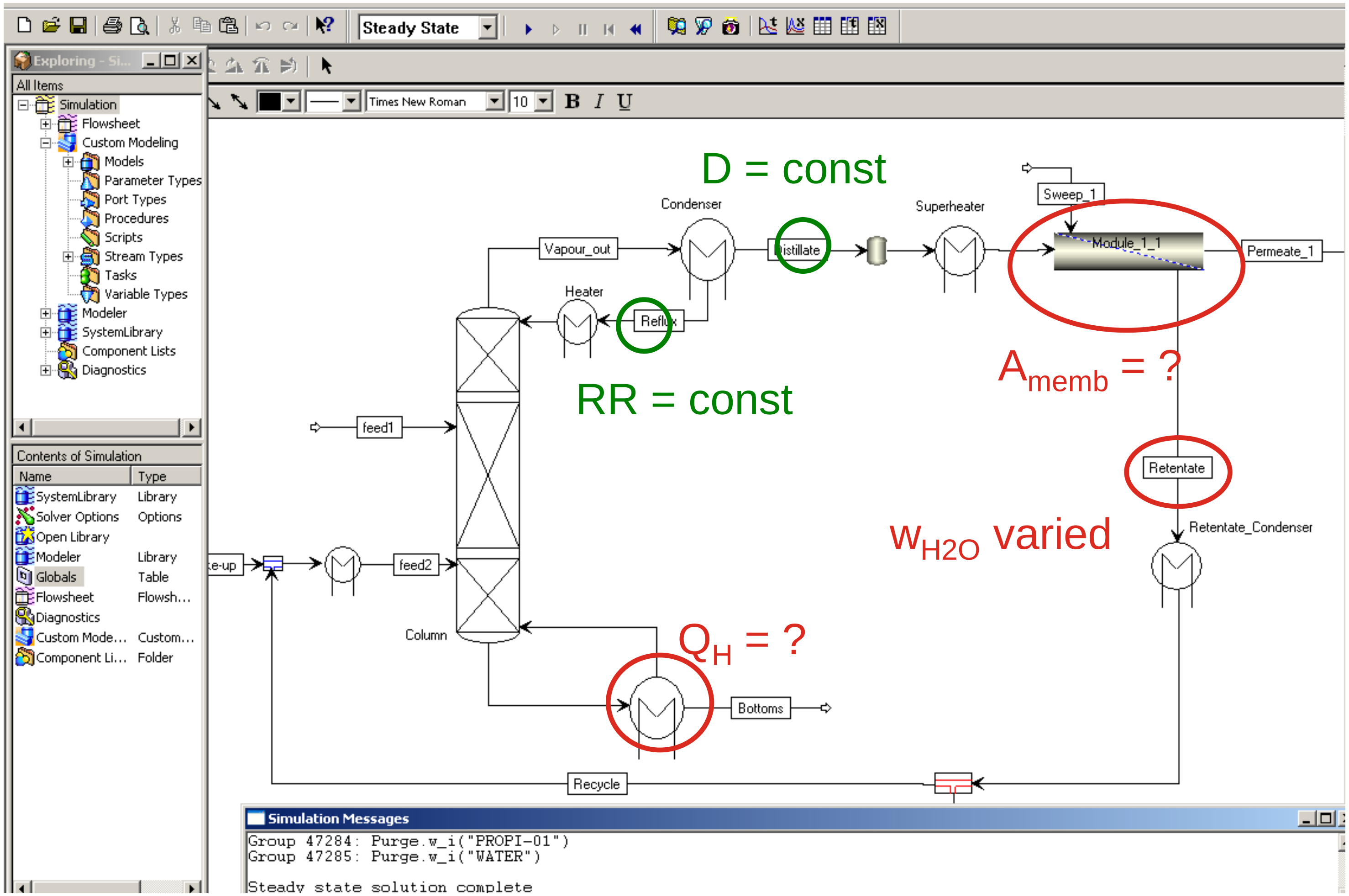
Closed recycle

$X_{ProAc,exp}$ = 73.3 % $X_{ProAc,sim}$ = 73.5 %

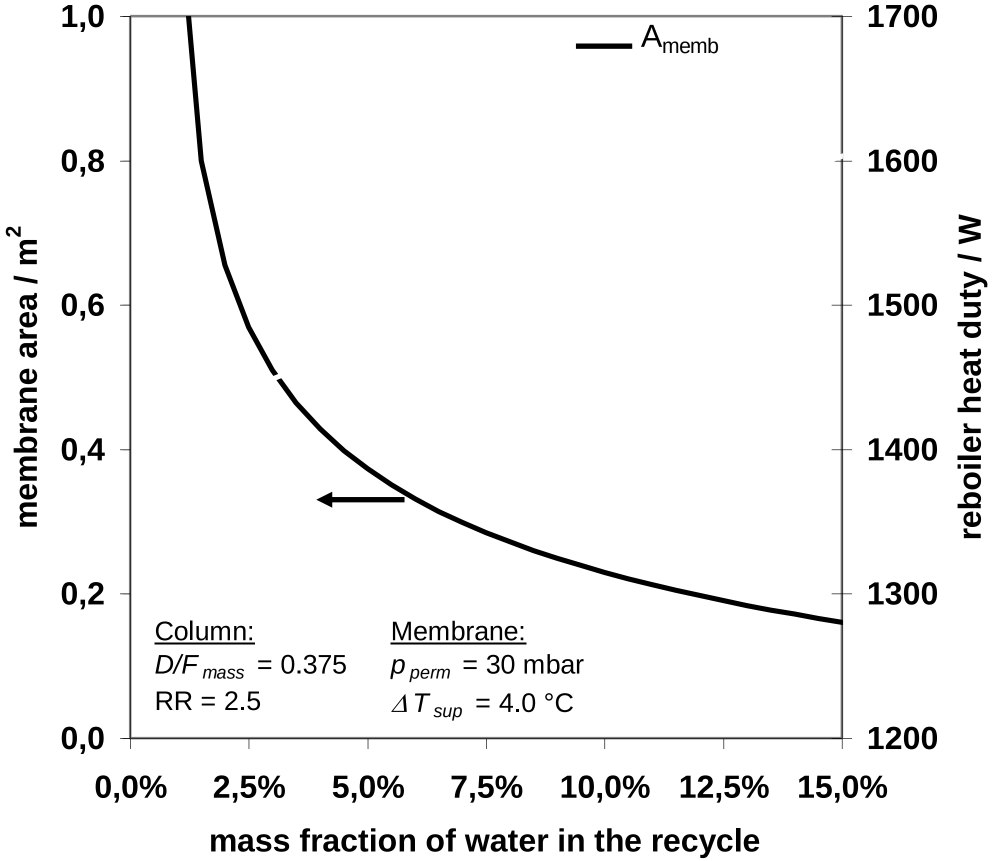
$X_{POH,exp}$ = 31.8 % $X_{POH,sim}$ = 31.9 %

➡ Excellent agreement between experiments and simulation

Hybrid separation process: process analysis

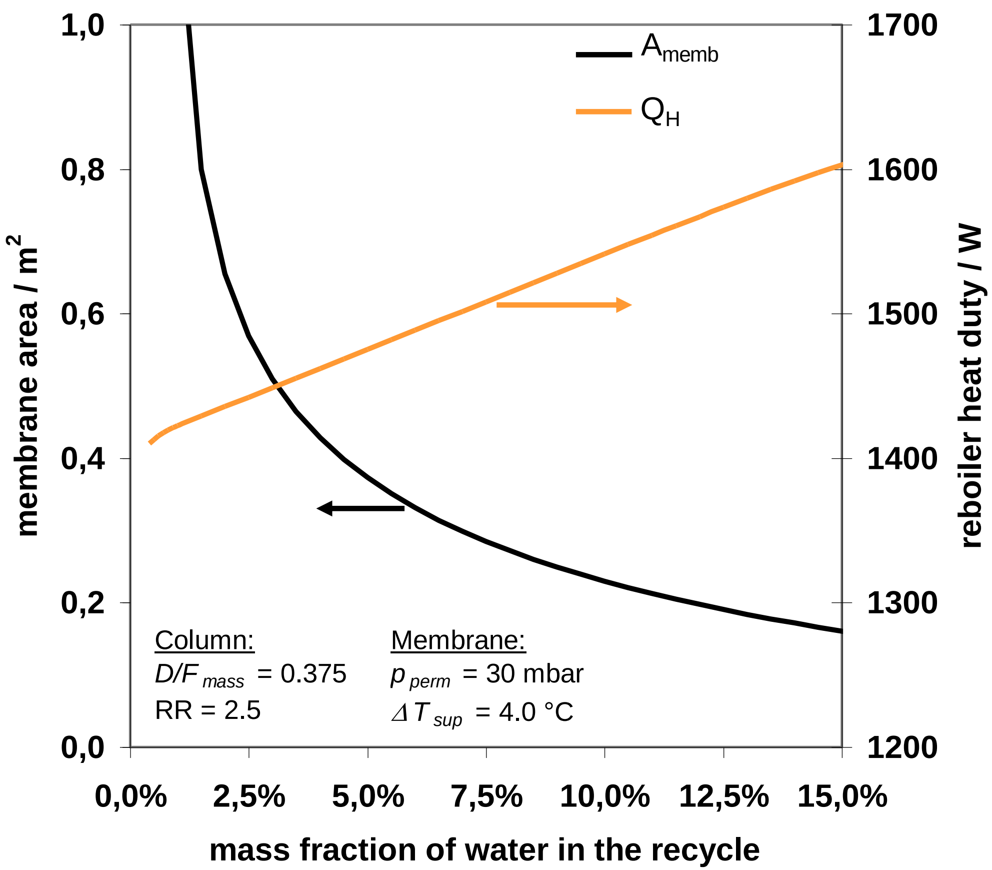


Hybrid separation process: process analysis



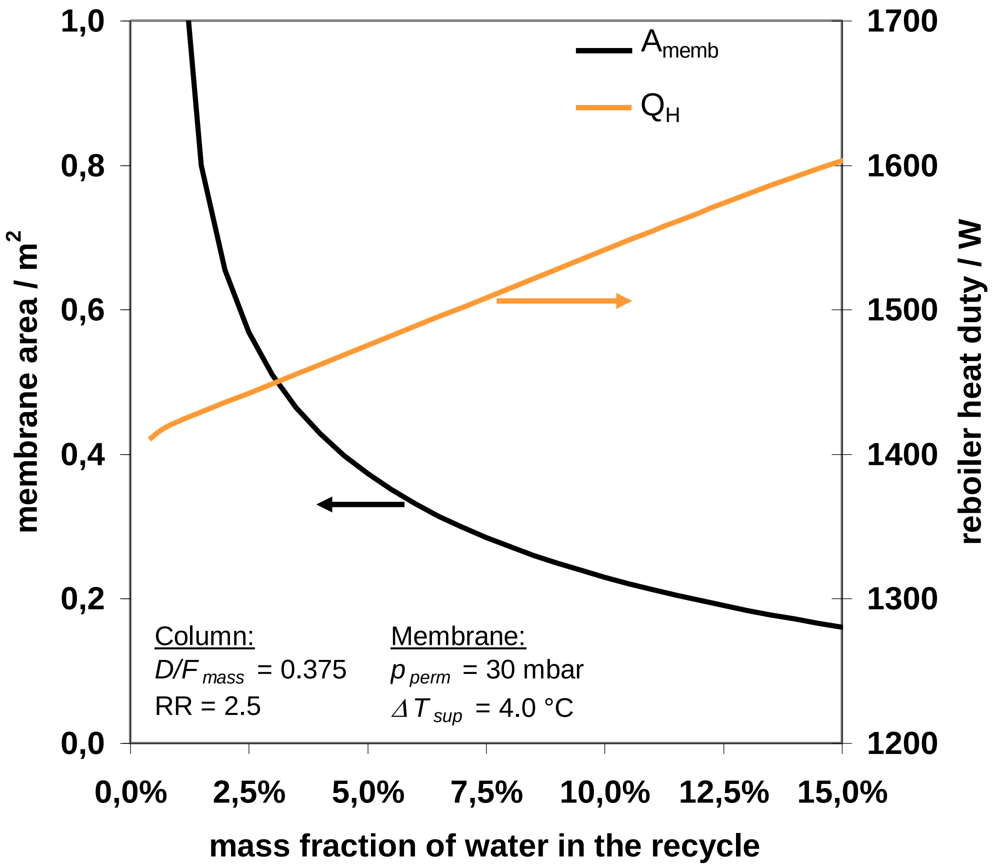
- Strong influence on A_{Memb}
- High degree of dewatering requires large A_{Memb}

Hybrid separation process: process analysis



- Strong influence on A_{Memb}
- High degree of dewatering requires large A_{Memb}
- Lower Q_H at higher recycle purity required

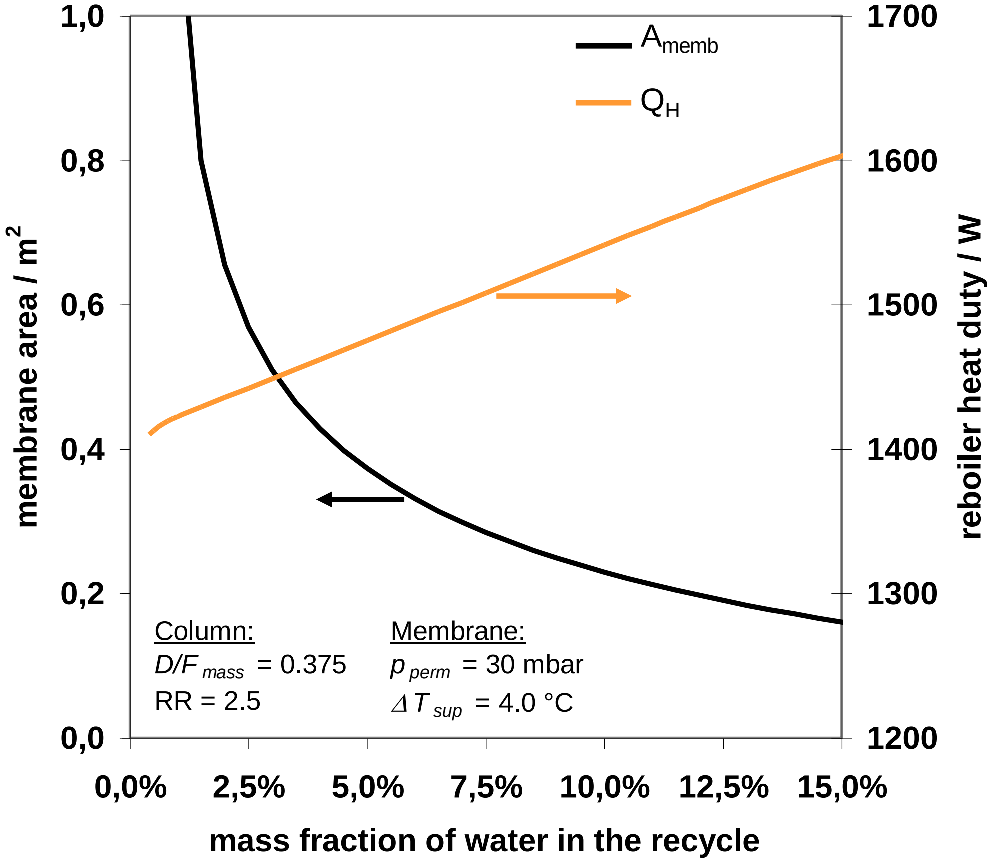
Hybrid separation process: process analysis



- Strong influence on A_{Memb}
- High degree of dewatering requires large A_{Memb}
- Lower Q_H at higher recycle purity required

⇒ Optimisation problem

Hybrid separation process: process analysis



- Strong influence on A_{Memb}
- High degree of dewatering requires large A_{Memb}
- Lower Q_H at higher recycle purity required

Optimisation problem

Modified stochastic optimisation algorithm based on a differential evolution* approach (MDE)

* Babu, Angira (2008): Comp. Chem. Eng., 30, p. 989–1002
Angira, Babu (2006): Chem. Eng. Sci., 61, p. 4707–4721
Frerick et al. (2008): Chem. Ing. Tech., 1, p.1-10

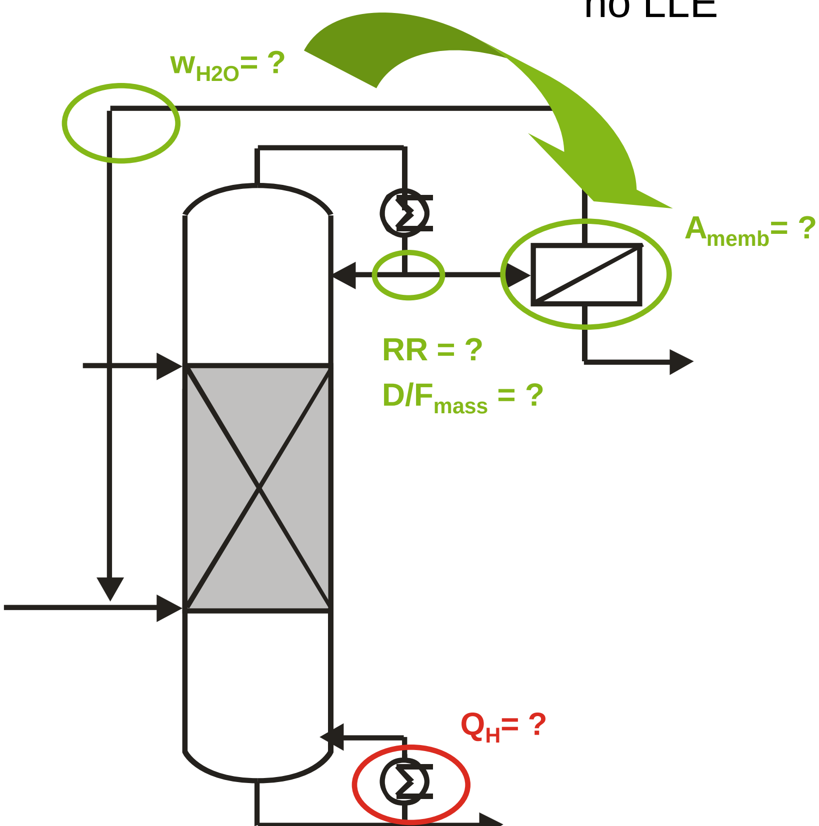
Hybrid separation process: rigorous optimisation using MDE

Objective function: min $Q_H = f(RR, D/F_{mass}, w_{H2O,recycle})$

Solution space: $0.8 \leq RR \leq 5$; $0.25 \leq D/F_{mass} \leq 0.55$; $0.02 \leq w_{H2O,recycle} \leq 0.15$

Constrain: $x_{ProPro,bottom} \geq 0.75$

no LLE



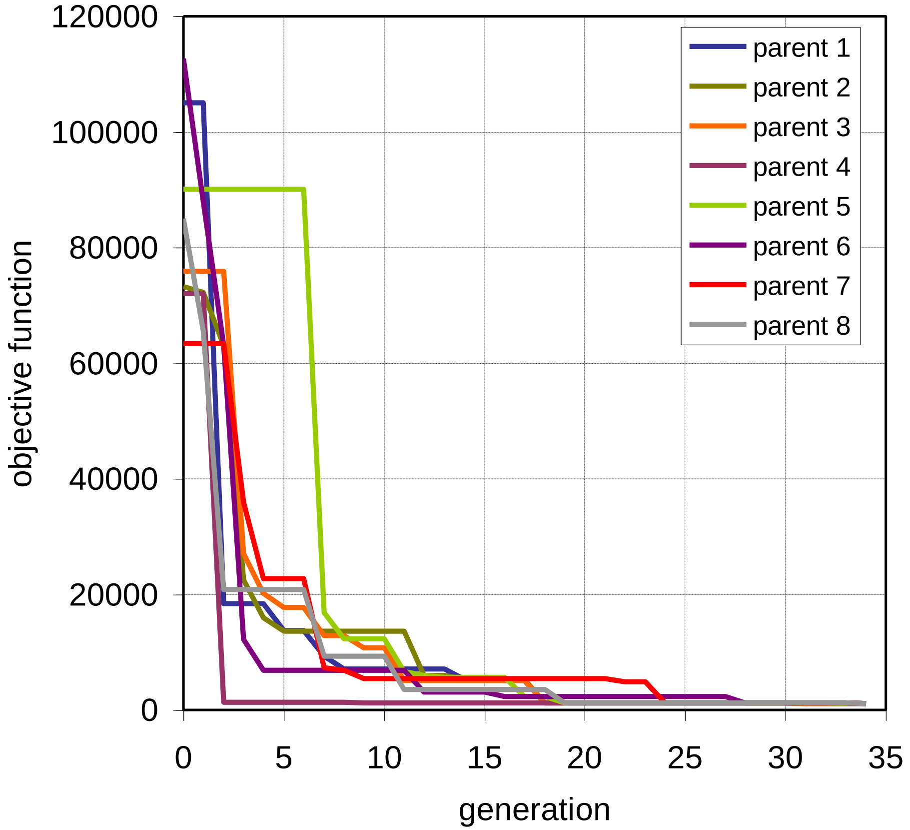
Hybrid separation process: rigorous optimisation using MDE

Objective function: min $Q_H = f(RR, D/F_{mass}, w_{H2O,recycle})$

Solution space: $0.8 \leq RR \leq 5$; $0.25 \leq D/F_{mass} \leq 0.55$; $0.02 \leq w_{H2O,recycle} \leq 0.15$

Constrain: $x_{ProPro,bottom} \geq 0.75$

no LLE



- 65 generations with 8 parents
- CPU-time ~ 12 hour
- $Q_{H,min}$ = 1059 W
- RR = 0.98; D/F_{mass} = 0.52; $w_{H2O,recycle}$ = 0.022
- A_{memb} = 3.04 m²

feasibility and applicability of the now available optimisation package based on detailed process models proven

Conclusion

- Choice of an appropriate **chemical test system**
- Development of a **generic methodology** for the design of hybrid processes
- Provision of **reliable experimental data** for the applied unit operations
- Experimental investigation of the **fully coupled unit operations**

→ **Reliable experimental database for both unit operations and hybrid process**

- **Model validation** for the stand-alone unit operations and the hybrid process
- **Analysis** of the influence of decisive operational parameters on the performance of the hybrid separation process
- **Optimisation** of the hybrid process **using** an **evolutionary algorithm**

Special thanks to:

Stefan Kluckhenn; Ildefonso Campos-Velarde; Christian Schroeder; Marcel Kotora; Martin Cruz-Dias; Carlos Duarte; Katrin Kissing; Siggi Weiss; Piotr Mitkowski; Paulo Perez; Gencay Cengiz; Rocio Castillo-Tornay; Dominik Plaßmann; Markus Hengsbach; Bharat Karanth; Mayur Dalwani; Mayuratheepan Puthirasigamany & the Laboratory of Fluid Separations

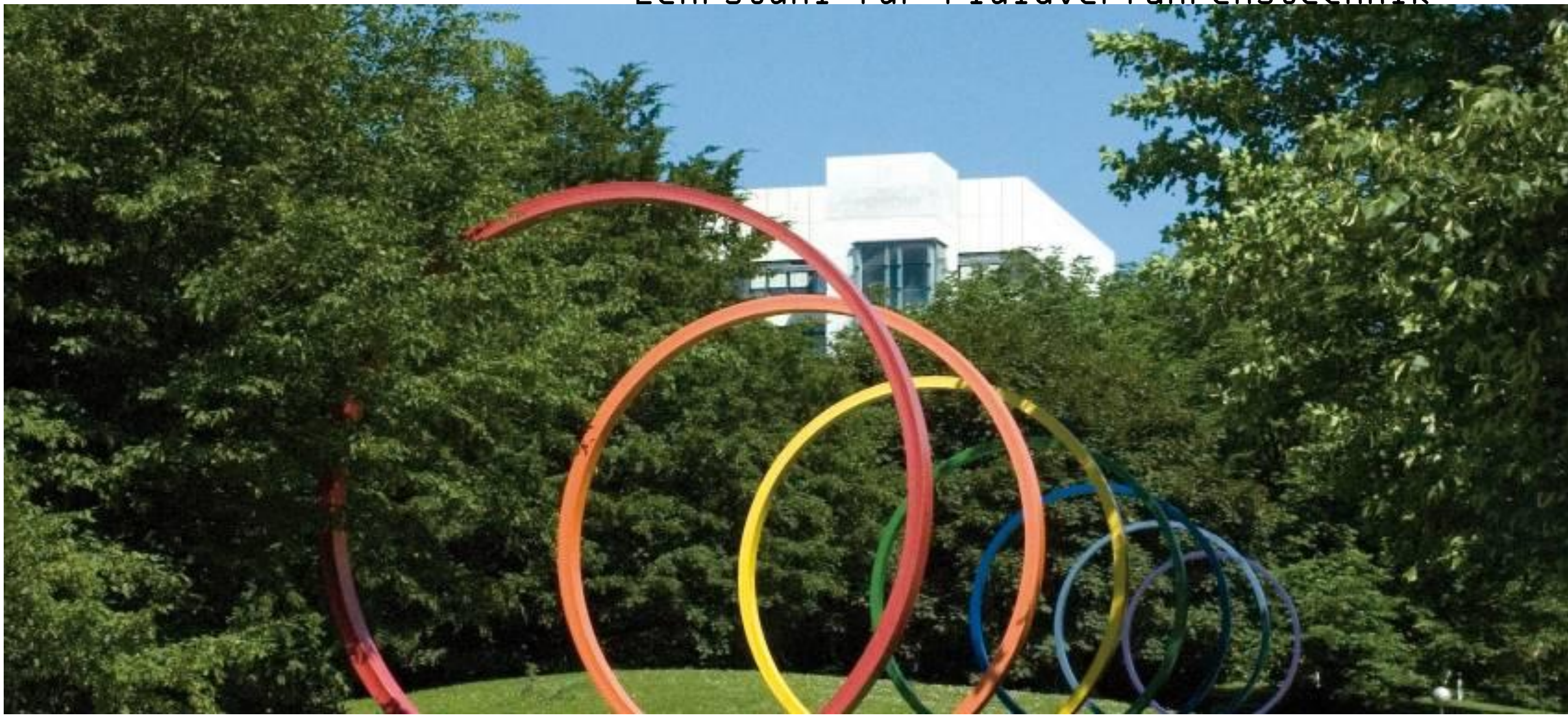
EU-Project INSERT
"Integrating Separation and Reaction Technologies"
(Contract No. NMP2-CT-2003-505862)

Marie Curie Training Site „Reactive Separations"
(Contract Nr. HPMT-CT-2001-00408)

Fonds der Chemischen Industrie

technische
universität
dortmund

Fakultät Bio- und
Chemieingenieurwesen
Lehrstuhl für Fluidverfahrenstechnik



Experimental Investigation, Analysis and Optimisation of Hybrid Separation Processes

EFCE Excellence Award in Process Intensification 2009

Carsten Buchaly