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)

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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)

Experiments
- Pilot-scale
- Operational parameters
- Model validation

Theory
- Experimental design
- Process analysis

Unit operation 2
(membrane separation)

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 ($\chi$): 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 $A_{\text{Mem}} = 0.5\text{m}^2$

Vapour permeation: pilot-scale plant

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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

Hybrid separation process: experimental investigations

RD and VP separated

sequential configuration

„closed recycle“
Hybrid separation process: experimental investigations

- Successful experiment with coupled unit operations

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- Increase of ester concentration in the bottom
- Change of distillate composition

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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, $\Delta p$, $k_g a$, $k_l a$)
- Steady state and dynamic process simulation

Vapour permeation based on „Solution-Diffusion-Model“
- Polarisation effects (c,T)
- Hydrodynamics (co-, counter-current; $\Delta p$)
- Membrane materials

Implemented in Aspen Custom Modeler™ (ACM)
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Reactive distillation: model validation

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

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Correlations for column internals:
- Sulzer BX -> Bravo et al.**; Rocha et al.**
- Katapak-SP -> Brunazzi***

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Model validation successful

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Hybrid separation process: model validation

![Graph showing molar fraction in the liquid phase vs. packing height]

- POH (exp)
- ProPro (exp)
- ProAc (exp)
- Water (exp)

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Hybrid separation process: model validation

Sequential configuration

\[
\begin{align*}
X_{\text{ProAc,exp}} & = 74.4 \% \\
X_{\text{ProAc,sim}} & = 73.6 \% \\
X_{\text{POH,exp}} & = 29.8 \% \\
X_{\text{POH,sim}} & = 29.4 \%
\end{align*}
\]
Hybrid separation process: model validation

Sequential configuration

\[ X_{\text{ProAc,exp}} = 74.4\% \quad X_{\text{ProAc,sim}} = 73.6\% \]
\[ X_{\text{POH,exp}} = 29.8\% \quad X_{\text{POH,sim}} = 29.4\% \]

Closed recycle

\[ X_{\text{ProAc,exp}} = 73.3\% \quad X_{\text{ProAc,sim}} = 73.5\% \]
\[ X_{\text{POH,exp}} = 31.8\% \quad X_{\text{POH,sim}} = 31.9\% \]

Excellent agreement between experiments and simulation
Hybrid separation process: process analysis

\[ D = \text{const} \]

\[ \text{RR} = \text{const} \]

\[ w_{\text{H}_2\text{O}} \text{ varied} \]

\[ Q_H = ? \]

\[ A_{\text{memb}} = ? \]
Hybrid separation process: process analysis

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

Column:
- $D/F_{\text{mass}} = 0.375$
- RR = 2.5

Membrane:
- $\rho_{\text{perm}} = 30 \text{ mbar}$
- $\Delta T_{\text{sup}} = 4.0 \, ^{\circ}\text{C}$
Hybrid separation process: process analysis

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- Lower $Q_H$ at higher recycle purity required

### Graph

- **$A_{\text{memb}}$**
- **$Q_H$**

### Parameters
- **Column:**
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  - RR = 2.5
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$A_{\text{memb}}$ vs. $Q_H$

Optimisation problem

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- RR = 2.5

Membrane:
- $\rho_{\text{perm}} = 30 \text{ mbar}$
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mass fraction of water in the recycle

reboiler heat duty / W

membrane area / m$^2$
Hybrid separation process: process analysis

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

Optimisation problem

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

Hybrid separation process: rigorous optimisation using MDE

Objective function: \[ \text{min } Q_H = f(\text{RR, D/F}_{\text{mass}}, w_{\text{H2O,recycle}}) \]

Solution space: \[ 0.8 \leq \text{RR} \leq 5; \quad 0.25 \leq \text{D/F}_{\text{mass}} \leq 0.55; \quad 0.02 \leq w_{\text{H2O,recycle}} \leq 0.15 \]

Constrain: \[ x_{\text{ProPro,bottom}} \geq 0.75 \]
no LLE

Hybrid separation process: rigorous optimisation using MDE

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Constrain:
\[ x_{\text{ProPro, bottom}} \geq 0.75 \]

Constraints:
- no LLE
- \[ \text{feasibility and applicability of the now available optimisation package based on detailed process models proven} \]

- 65 generations with 8 parents
- CPU-time ~ 12 hour
- \( Q_{H,\text{min}} = 1059 \text{ W} \)
- \( RR = 0.98; \frac{D}{F}_{\text{mass}} = 0.52; w_{H2O,\text{recycle}} = 0.022 \)
- \( A_{\text{memb}} = 3.04 \text{ m}^2 \)
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
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