

Supplementary information to "Efficient optimization-based design of membrane-assisted distillation processes"

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1 Thermodynamical property calculation

The thermodynamical property calculations are based on computational routines that have been developed at Aachener Verfahrenstechnik - Process Systems Engineering.^{1,2} For each example the nonideality of the liquid phase is modeled by means of either the UNIQUAC, the NRTL or the Wilson model.

UNIQUAC

$$\ln \gamma_i = \ln \gamma_i^{(C)} + \ln \gamma_i^{(R)}, \quad (1)$$

$$\ln \gamma_i^{(C)} = \ln \frac{\Phi_i}{x_i} + \frac{z}{2} q_i \ln \frac{\Theta_i}{\Phi_i} + l_i - \frac{\Phi_i}{x_i} \sum_{k=1}^{n_c} x_k l_k, \quad (2)$$

$$\ln \gamma_i^{(R)} = q'_i \left(1 - \ln \sum_{k=1}^{n_c} \Theta'_k \tau_{ki} - \sum_{k=1}^{n_c} \frac{\Theta'_k \tau_{ik}}{\sum_{m=1}^{n_c} \Theta'_m \tau_{mk}} \right), \quad (3)$$

$$l_i = \frac{z}{2} (r_i - q_i) - (r_i - 1), \quad (4)$$

$$z = 10, \quad (5)$$

$$\tau_{ij} = \exp \left(- \frac{(u_{ij} + u_{T,ij} T) - (u_{jj} + u_{T,jj} T)}{RT} \right), \quad (6)$$

$$\Phi_i = \frac{r_i x_i}{\sum_{k=1}^{n_c} r_k x_k}, \quad (7)$$

$$\Theta_i = \frac{q_i x_i}{\sum_{k=1}^{n_c} q_k x_k}, \quad (8)$$

$$\Theta'_i = \frac{q'_i x_i}{\sum_{k=1}^{n_c} q'_k x_k}. \quad (9)$$

For this model the parameters q_i , q'_i , r_i , u_{ij} , $u_{T,ij}$ and an accordingly defined general gas constant R are required. The parameters $u_{T,ij}$ can describe a temperature dependency of the binary interaction parameters u_{ij} and is not included in the classical UNIQUAC model.

If not stated explicitly $u_{T,ij} = 0$. If q'_i is not stated explicitly, $q'_i = q_i$ holds.

NRTL

$$\ln \gamma_j = \frac{\sum_{k=1}^{n_c} \tau_{kj} G_{kj} x_k}{\sum_{l=1}^{n_c} G_{lj} x_l} + \sum_{k=1}^{n_c} \left[\frac{x_k G_{jk}}{\sum_{l=1}^{n_c} G_{lk} x_l} \left(\tau_{jk} - \frac{\sum_{m=1}^{n_c} \tau_{mk} G_{mk} x_m}{\sum_{l=1}^{n_c} G_{lk} x_l} \right) \right], \quad (10)$$

$$\tau_{kj} = \frac{(g_{kj} + g_{T,kj}T) - (g_{jj} + g_{T,jj}T)}{RT}, \quad (11)$$

$$G_{kj} = \exp(-\alpha_{kj} \tau_{kj}). \quad (12)$$

For this model the parameters g_{ij} , $g_{T,ij}$, α_{ij} and an accordingly defined general gas constant R are required. The parameters $g_{T,ij}$ can describe a temperature dependency of the binary interaction parameters g_{ij} and are not included in the classical NRTL model. If not stated explicitly $g_{T,ij} = 0$.

Wilson

$$\ln \gamma_i = 1 - \ln \sum_{k=1}^{n_c} x_k \Lambda_{ik} - \sum_{l=1}^{n_c} \frac{x_l \Lambda_{li}}{\sum_{k=1}^{n_c} x_k \Lambda_{lk}}, \quad (13)$$

$$\Lambda_{ij} = \frac{v_j^{0L}}{v_i^{0L}} \exp \left(-\frac{(\lambda_{ij} + \lambda_{T,ij}T) - (\lambda_{ii} + \lambda_{T,ii}T)}{RT} \right). \quad (14)$$

For this model the parameters v_i^{0L} , λ_{ij} , $\lambda_{T,ij}$ and an accordingly defined general gas constant R are required. The parameters $\lambda_{T,ij}$ can describe a temperature dependency of the binary interaction parameters λ_{ij} and is not included in the classical Wilson model. If not stated explicitly $\lambda_{T,ij} = 0$.

For each example, the vapor pressure is calculated by means of the extended Antoine equation.

Antoine equation

$$\ln(p_i^0) = c_{1,i} + \frac{c_{2,i}}{T + c_{3,i}} + c_{4,i}T + c_{5,i}\ln(T) + c_{6,i}T^{c_{7,i}} \quad \text{for } c_{8,i} \leq T \leq c_{9,i}. \quad (15)$$

For this model the parameters $c_{1,i}, \dots, c_{9,i}$ are required. The parameters $c_{8,i}$ and $c_{9,i}$ define the range of validity of the model.

The vapor phase is either assumed ideal ($\varphi_i = 1$), or the fugacity coefficient of the mixture is determined by means of the Redlich-Kwong equation of state (EOS).

Redlich-Kwong EOS

$$\ln(\varphi_i) = \frac{b_i}{b}(Z - 1) - \ln \left[Z \left(1 - \frac{b}{V} \right) \right] + \frac{1}{bRT^{3/2}} \left[\frac{ab_i}{b} - 2\sqrt{aa_i} \right] \ln \left(1 + \frac{b}{V} \right), \quad (16)$$

$$p = \frac{RT}{V - b} - \frac{a}{T^{1/2}V(V + b)}, \quad (17)$$

$$Z = \frac{V}{V - b} - \frac{a}{RT^{3/2}(V + b)}, \quad (18)$$

$$a = \sum_{i=1}^{n_c} \sum_{j=1}^{n_c} y_i y_j a_{ij}, \quad (19)$$

$$a_{ij} = \sqrt{a_i a_j} (1 - k_{ij}), \quad (20)$$

$$b = \sum_{i=1}^{n_c} y_i b_i, \quad (21)$$

$$a_i = 0.42728 \frac{R^2 T_{c,i}^{5/2}}{p_{c,i}}, \quad (22)$$

$$b_i = 0.08664 \frac{RT_{c,i}}{p_{c,i}}. \quad (23)$$

For this model the parameters are given by the critical pressure $p_{c,i}$ and the critical temperature $T_{c,i}$ of the individual components and a mixing parameter k_{ij} , which, if not stated otherwise, is assumed to be zero.

Calculation of molar enthalpies

The vapor enthalpies

$$h^V(y, p, T) = h^{ig}(y, T) + \Delta h^V(y, p, T), \quad (24)$$

$$h^{ig}(y, T) = \sum_{i=1}^{n_c} y_i h_i^{0,ig}(T), \quad (25)$$

$$h_i^{0,ig}(T) = h_i^{0,ig}(T_0) + \int_{T=T_0}^T c_{p,i}^{ig}(T) dT, \quad (26)$$

and the liquid enthalpies

$$h^L(x, p, T) = h^{ig}(x, T) + \Delta h^L(x, p, T), \quad (27)$$

$$\Delta h^L(x, T) = \sum_{i=1}^{n_c} x_i \Delta h_i^{0L}(T) + h^{ex}(x, T), \quad (28)$$

$$h^{ex}(x, T) = -RT^2 \sum_{i=1}^{n_c} x_i \frac{\partial \ln \gamma_i}{\partial T}, \quad (29)$$

$$\Delta h_i^{0L}(p, T) = \Delta h_i^{0V}(p^0, T) - \Delta_{vap} H_i^*(T) + \Delta_{h,i}^{0L}(p, T) \quad (30)$$

are calculated on the basis of models for ideal gas heat capacities c_p^{ig} and the heat of vaporization $\Delta_{vap} H_i^*(T)$. The liquid excess enthalpy h^{ex} calculation is based on the activity coefficient model, while the calculation of the enthalpy departure of the vapor phase ($\Delta h^V, \Delta h_i^{0V}$) is based on the fugacity coefficient model, or assumed to be equal to zero, in case of an ideal gas. The pressure correction term $\Delta_{h,i}^{0L}(T, p)$ for the liquid enthalpy is assumed to be zero for all case studies. The reference state for all calculations is given by $T_0 = 298.15$ K and $p_0 = 101325$ Pa.

Vapor heat capacity

The ideal gas heat capacity is either calculated by means of a polynomial calculation

$$c_{p,i}^{ig}(T) = c_{1,i} + c_{2,i}T + c_{3,i}T^2 + c_{4,i}T^3 + c_{5,i}T^4 + c_{6,i}T^5 \quad \text{for } c_{7,i} \leq T \leq c_{8,i}, \quad (31)$$

$$c_{p,i}^{ig}(T) = c_{9,i} + c_{10,i}T^{c_{11,i}} \quad \text{for } T < c_{7,i}, \quad (32)$$

$$c_{p,i}^{ig}(T) = c_p^{ig}(T = c_{8,i}) + (T - c_{8,i}) \left(\frac{dc_p(T = c_{8,i})}{dT} \right) \quad \text{for } T > c_{8,i}, \quad (33)$$

for which the parameters $c_{1,i}, \dots, c_{11,i}$ are required, or by means of a hyperbolic correlation (DIPPR eq. 107)

$$c_{p,i}^{ig}(T) = c_{1,i} + c_{2,i} \left(\frac{c_{3,i}/T}{\sinh(c_{3,i}/T)} \right)^2 + c_{4,i} \left(\frac{c_{5,i}/T}{\cosh(c_{5,i}/T)} \right)^2 \quad \text{for } c_{6,i} < T < c_{7,i}, \quad (34)$$

for which the parameters $c_{1,i}, \dots, c_{7,i}$ are required.

Heat of vaporization

The heat of vaporization is either calculated by means of the Watson correlation

$$\Delta_{vap}H_i^*(T) = \Delta_{vap}H_{1,i}^* \left(\frac{1 - \frac{T}{T_{c,i}}}{1 - \frac{T_{1,i}}{T_{c,i}}} \right)^{a_i + b_i \left(1 - \frac{T}{T_{c,i}} \right)} \quad \text{for } T \geq T_{min,i}, \quad (35)$$

for which the parameters $\Delta_{vap}H_{1,i}^*$, $T_{1,i}$, a_i , b_i , $T_{min,i}$, as well as the critical temperature $T_{c,i}$ of component i are required, or a DIPPR correlation

$$\Delta_{vap}H_i^*(T) = c_{1,i}(1 - T_{r,i})^{(c_{2,i} + c_{3,i}T_{r,i} + c_{4,i}T_{r,i}^2 + c_{5,i}T_{r,i}^3)} \quad \text{for } c_{6,i} \leq T \leq c_{7,i}, \quad (36)$$

$$T_{r,i} = \frac{T}{T_{c,i}}. \quad (37)$$

For this model the parameters $c_{1,i}, \dots, c_{7,i}$ are required.

1.1 Binary system of ethanol and water

The liquid phase behavior is described by the Wilson or the UNIQUAC model. The gas constant for both models is $R = 1.98721$. The parameters are similar to those used by Bausa and Marquardt³ and are listed in Table 1 and 2

Table 1: Parameter for Wilson model (eqs. (13)-(14))

	ethanol	water
λ_{ij}		
ethanol	0.	91.126
water	765.813	0.
$\lambda_{T,ij}$		
ethanol	0.	0.4506
water	0.5411	0.
V_j	58.69	18.07

Table 2: Parameter for UNIQUAC model (eqs. (1)-(9))

	ethanol	water
u_{ij}		
ethanol	0.	-31.629
water	-96.473	0.
$u_{T,ij}$		
ethanol	0.	0.4759
water	0.6843	0.
q_j	1.9720	1.4
r_j	2.1055	0.92

The vapor phase is assumed ideal and the parameters for the extended Antoine equation are listed in Table 3.

Table 3: Parameter for Antoine equation (eq. (15))

	ethanol	water
$c_{1,i}$	8.10753e1	6.51544e1
$c_{2,i}$	-7.82780e3	-6.84291e3
$c_{3,i}$	0.	0.
$c_{4,i}$	-1.85859e-3	2.78351e-3
$c_{5,i}$	-7.96131	-6.13638
$c_{6,i}$	2.36730e-17	3.31168e-18
$c_{7,i}$	6.	6.
$c_{8,i}$	3.02559e2	3.12967e2
$c_{9,i}$	5.162e2	6.473e2

The vapor heat capacities are calculated by means of the polynomial model and the parameters are listed in Table 4.

Table 4: Parameter for vapor heat capacity model (eqs. (31)-(33))

	ethanol	water
$c_{1,i}$	9.01418e3	3.37381e4
$c_{2,i}$	2.14071e2	-7.01756
$c_{3,i}$	-8.39035e-2	2.72961e-2
$c_{4,i}$	1.37327e-6	-1.66465e-5
$c_{5,i}$	0.0	4.29761e-9
$c_{6,i}$	0.0	-4.16961e-13
$c_{7,i}$	3e2	2e2
$c_{8,i}$	1.31840e3	3e3
$c_{9,i}$	3.32560e4	3.32560e4
$c_{10,i}$	5.6902	1.89780e-20
$c_{11,i}$	1.5164	9.2846

The heat of vaporization is calculated by means of the Watson model and the parameters are listed in Table 5.

Table 5: Parameter for heat of vaporization model (eq. (35))

	ethanol	water
$\Delta_{vap}H_{1,i}^*$	3.87698e+07	4.06831e+07
$T_{1,i}$	3.51500e+02	3.73200e+02
a_i	3.87894e-01	3.10646e-01
b_i	0.0	0.0
$T_{min,i}$	1.59100e+02	2.73200e+02

The critical temperatures and molar masses are listed in Table 6.

Table 6: Critical temperatures and molar masses.

	ethanol	water
$T_c [K]$	5.16200e+02	6.47300e+02
$M [g/mol]$	46.07	18.02

1.2 Ternary system of butene, MTBE and methanol

The liquid phase behavior is described by the UNIQUAC model, with gas constant $R = -1$. The parameters are similar to those used by Bausa and Marquardt³ and are listed in Table 7.

Table 7: Parameter for UNIQUAC model (eqs. (1)-(9))

	butene	MTBE	methanol
u_{ij}			
butene	0.0	1.15450e+03	-1.16618e+03
MTBE	-7.45158e+02	0.0	-4.31664e+02
methanol	-3.98292e+01	7.61634e+01	0.0
$u_{T,ij}$			
butene	0.0	-3.91240e+00	1.32656e+00
MTBE	2.60468e+00	0.0	0.0
methanol	1.03212e-01	0.0	0.0
q_j	2.56400	3.63200	1.43200
r_j	2.92090	4.06790	1.43111

The vapor phase is assumed ideal and the parameters for the extended Antoine equation are listed in Table 8.

Table 8: Parameter for Antoine equation (eq. (15))

	butene	MTBE	methanol
$c_{1,i}$	6.84900e+01	5.58750e+01	8.17680e+01
$c_{2,i}$	-4.35020e+03	-5.13160e+03	-6.87600e+03
$c_{3,i}$	0.	0.	0.
$c_{4,i}$	0.	0.	0.
$c_{5,i}$	-7.41240e+00	-4.96040e+00	-8.70780e+00
$c_{6,i}$	1.05030e-05	1.91230e-17	7.19260e-06
$c_{7,i}$	2.	6.	2.
$c_{8,i}$	8.78000e+01	1.64550e+02	1.75470e+02
$c_{9,i}$	4.19950e+02	4.97100e+02	5.12640e+02

Due to missing parameters for MTBE, the vapor heat capacities are calculated by means of the polynomial correlation for butene and methanol, and by means of the DIPPR model for MTBE. The parameters are listed in Table 9.

Table 9: Parameter for vapor heat capacity models (eqs. (31)-(33), and (34))

POLY	butene	methanol	DIPPR	MTBE
$c_{1,i}$	-2.99356e+03	2.11517e+04	$c_{1,i}$	9.93280e+04
$c_{2,i}$	3.53198e+02	7.09244e+01	$c_{2,i}$	3.06670e+05
$c_{3,i}$	-1.99040e-01	2.58702e-02	$c_{3,i}$	1.74260e+03
$c_{4,i}$	4.46313e-05	-2.85163e-05	$c_{4,i}$	2.07640e+05
$c_{5,i}$	0.0	0.0	$c_{5,i}$	7.95590e+02
$c_{6,i}$	0.0	0.0	$c_{6,i}$	200
$c_{7,i}$	300.0	300.0	$c_{7,i}$	1500
$c_{8,i}$	1.48660e+03	1.26180e+03		
$c_{9,i}$	3.32560e+04	3.32560e+04		
$c_{10,i}$	1.02000e+01	3.78130e-02		
$c_{11,i}$	1.5	2.20140		

For similar reasons the heat of vaporization is calculated by means of the Watson model for butene and methanol, and by means of the DIPPR model for MTBE. The parameters are listed in Table 10.

Table 10: Parameter for heat of vaporization models (eqs. (35) and (36)-(37))

Watson	butene	methanol	DIPPR	MTBE
$\Delta_{vap}H_{1,i}^*$	2.19305e+07	3.52780e+07	$c_{1,i}$	4.20240e+07
$T_{1,i}$	2.66900e+02	3.37800e+02	$c_{2,i}$	3.78260e-01
a_i	3.54701e-01	3.71655e-01	$c_{3,i}$	0.0
b_i	0.0	0.0	$c_{4,i}$	0.0
$T_{min,i}$	8.78000e+01	1.75500e+02	$c_{5,i}$	0.0
			$c_{6,i}$	1.64550e+02
			$c_{7,i}$	4.97100e+02

The critical temperatures and molar masses are listed in Table 11.

Table 11: Critical temperatures and molar masses.

	butene	MTBE	methanol
$T_c [K]$	4.19950e+02	4.97100e+02	5.12640e+02
$M [g/mol]$	5.61075e+01	8.81497e+01	3.20422e+01

1.3 Ternary system of acetone, isopropanol and water

The liquid phase behavior is described by the NRTL model with parameters similar to those used by Koch et al.⁴ listed in Table 12. The gas constant is $R = 1$.

Table 12: Parameter for NRTL model (eqs. (10)-(12))

	acetone	isopropanol	water
g_{ij}			
acetone	0.0	8.78177e+02	1.20373e+03
isopropanol	-6.54801e+02	0.0	4.32900e+02
water	-2.09967e+03	-1.42928e+03	0.0
$g_{T,ij}$			
acetone	0.0	-2.59220	-3.07680
isopropanol	2.69050	0.0	-1.32090
water	7.93850	6.62100	0.0
α_{ij}			
acetone	3.00000e-01	3.00000e-01	3.00000e-01
isopropanol	3.00000e-01	3.00000e-01	3.00000e-01
water	3.00000e-01	3.00000e-01	3.00000e-01

The non-ideality of the vapor phase is modeled by means of the Redlich-Kwong EOS. The

critical temperatures, and pressures are listed together with the molar masses in Table 13.

Table 13: Critical temperatures, pressures and molar masses.

	acetone	isopropanol	water
T_c [K]	5.08200e+02	5.08300e+02	6.47130e+02
p_c [Pa]	4.70100e+06	4.76400e+06	2.20550e+07
M [g/mol]	5.80800e+01	6.00959e+01	1.80153e+01

The parameters for the extended Antoine equation are listed in Table 14.

Table 14: Parameter for Antoine Equation (eq. (15))

	acetone	isopropanol	water
c_{1i}	6.90060e+01	7.64300e+01	7.36490e+01
c_{2i}	-5.59960e+03	-7.60700e+03	-7.25820e+03
c_{3i}	0.0	0.0	0.0
c_{4i}	0.0	0.0	0.0
c_{5i}	-7.0985000	-7.4086000	-7.3037000
c_{6i}	6.22370e-06	4.39860e-18	4.16530e-06
c_{7i}	2.0000000	6.0000000	2.0000000
c_{8i}	1.78450e+02	1.85280e+02	2.73160e+02
c_{9i}	5.08200e+02	5.08300e+02	6.47130e+02

The vapor heat capacities are calculated by means of the DIPPR model and the parameters are listed in Table 15.

Table 15: Parameter for vapor heat capacity model (eq. (34))

	acetone	isopropanol	water
$c_{1,i}$	5.70400e+04	5.72300e+04	3.33630e+04
$c_{2,i}$	1.63200e+05	1.91000e+05	2.67900e+04
$c_{3,i}$	1.60700e+03	1.42100e+03	2.61050e+03
$c_{4,i}$	9.68000e+04	1.21550e+05	8.89600e+03
$c_{5,i}$	7.31500e+02	6.26000e+02	1.16900e+03
$c_{6,i}$	2.00000e+02	1.50000e+02	1.00000e+02
$c_{7,i}$	1.50000e+03	1.50000e+03	2.27315e+03

The heat of vaporization is calculated by means of the DIPPR model and the parameters are listed in Table 16.

Table 16: Parameter for heat of vaporization model (eqs. (36)-(37))

	acetone	isopropanol	methanol
$c_{1,i}$	4.21500e+07	6.30800e+07	5.20530e+07
$c_{2,i}$	3.39700e-01	3.92100e-01	3.19900e-01
$c_{3,i}$	0.0	0.0	-2.12000e-01
$c_{4,i}$	0.0	0.0	2.57950e-01
$c_{5,i}$	0.0	0.0	0.0
$c_{6,i}$	1.78450e+02	1.85280e+02	2.73160e+02
$c_{7,i}$	5.08200e+02	5.08300e+02	6.47130e+02

2 Economic model

The optimal process design is determined subject to a cost function which represents the objective function of the optimization problem. The overall cost are composed of the one-time investment cost C_{inv} and the continuously spent operational cost C_{op} . In order to calculate the total annualized cost (TAC), the investment cost is annualized by means of a capital charge factor

$$f_c = \frac{ir \cdot (1 + ir)^{lt}}{(1 + ir)^{lt} - 1}, \quad (38)$$

which is based on the assumption of a certain plant lifetime (lt) and a specific interest rate (ir). The total annualized cost of each separation process

$$TAC = f_c \cdot (C_{inv,D} + C_{inv,M}) + C_{op,D} + C_{op,M}, \quad (39)$$

comprise of the investment and operational cost for the distillation column(s) ($C_{inv,D}$ and $C_{op,D}$) and the investment and operational cost for the membrane network(s) ($C_{inv,M}$ and $C_{op,M}$). The single cost items are further described in the following subsections.

2.1 Distillation columns

The operational cost for a distillation column are based on the utilities for heating and cooling, which are utilized during the annual operational time t_a .

$$C_{op,D} = t_a \cdot (m_{cool} \cdot C_{cool} + m_{steam} \cdot C_{steam}) \quad (40)$$

The required amount of cooling utility m_{cool} is calculated based on the required energy for condensation Q_C , the specific heat capacity c_P and the temperature elevation ΔT_{cool} of the cooling utility

$$m_{cool} = \frac{-Q_C}{c_P \cdot \Delta T_{cool}}. \quad (41)$$

The required steam for heating is calculated based on the heat duty in the reboiler Q_B and the enthalpy of vaporization Δh_{vap}

$$m_{steam} = \frac{Q_B}{\Delta h_{vap}}. \quad (42)$$

The choice of utilities determines their specific cost (C_{steam} and C_{cool}) and has an impact on the specific heat capacity and heat of vaporization. The costs for the different utilities are listed in Table 17 and 18. The values are based on previous publications.^{3,5-7}

Table 17: Cost parameters for steam.

p [bar]	3	10	20
T_{steam} [°C]	133.54	179.88	212.37
Δh_{vap} [$\frac{kJ}{kg}$]	2163.2	2013.6	1888.6
C_{steam} [$\frac{€}{ton}$]	12	15	18

Table 18: Cost parameters for cooling utility.

T [$^{\circ}\text{C}$]	15	-5	-15
Δt_{cool} [K]	20	10	10
c_p [$\frac{\text{kJ}}{\text{kgK}}$]	4.183	3.839	3.839
C_{cool} [$\frac{\text{€}}{\text{ton}}$]	0.05	2.0	3.5

The investment costs of the distillation columns

$$C_{inv,D} = C_{inv,pv} + C_{inv,tr} + \sum_{reb,con} C_{inv,hx} \quad (43)$$

are estimated based on a tray column assumption and composed of estimates for the pressure vessel (pv), the trays (tr) and the heat exchangers (hx). The cost estimates for each cost item are based on the correlations by Guthrie⁸ and Biegler et al.⁹ The investment costs

$$C_{inv} = BC \cdot (MPF + MF - 1) \cdot UF, \quad (44)$$

$$UF = MS/MS_{1969}, \quad (45)$$

are calculated as a product of a base cost BC , operation- and material-specific correction factors MPF and MF and an update factor UF , which is based on the Marshall-Swift index MS . For all calculations the Marshall-Swift index $MS = 1473.30$ from the third quarter of 2010 is used and related to $MS_{1969} = 281$. The conversion from Dollar to Euro is performed with an exchange rate of $0.71577 \frac{\text{€}}{\text{\$}}$. The values for MPF and MF for the different cost items are given in the work of Guthrie.⁸ The equipment is assumed to be stainless steel, with kettle reboiler and floating head condensers.

The base costs are estimated with the following correlations

$$BC_{pv} = C_{0,pv} \cdot \left(\frac{H_{col}}{H_0} \right)^{\alpha_{pv}} \cdot \left(\frac{D_{col}}{D_0} \right)^{\beta_{pv}}, \quad (46)$$

$$BC_{tr} = C_{0,tr} \cdot \left(\frac{H_{col}}{H_0} \right)^{\alpha_{tr}} \cdot \left(\frac{D_{col}}{D_0} \right)^{\beta_{tr}}, \quad (47)$$

$$BC_{hx} = C_{0,hx} \cdot \left(\frac{A_{hx}}{A_0} \right)^{\alpha_{hx}}. \quad (48)$$

The base values for column height H_0 and diameter D_0 and the area of the heat exchangers A_0 , as well as the values for the exponents α and β are given in the work of Guthrie⁸ and Biegler et al.⁹ The height of the column

$$H_{col} = N_{col} \cdot H_{tray} + H_{cl} \quad (49)$$

is calculated based on the number of equilibrium trays N_{col} , a height value for a single tray H_{tray} and a clearance for liquid distributors, demister etc. H_{cl} . For all calculations a height of $H_{tray} = 0.5$ m and an additional spacing of $H_{cl} = 4$ m are assumed. The column diameter D_{col} is calculated similar to the work of Brusi¹⁰ from the gas load of the distillation column. The diameter for a specific tray n is calculated by

$$D_{col,n} = \sqrt{\frac{4 \cdot V_n}{\pi \cdot F}} \sqrt{\frac{R \cdot T_n \cdot \sum_{i=1}^{n_c} y_{n,i} \cdot M M_i}{p}}, \quad (50)$$

with the F-factor F assumed to be $2Pa^{0.5}$. While the column diameter should be determined as $\max_n D_{col,n}$, a single tray is selected and fixed in the optimization for simplicity, instead of introducing the highly nonlinear equation for every tray.

The heat exchanger areas A_{hx} are determined by means of

$$A_{hx} = \frac{Q_B}{\Delta T_{reb} \cdot k_{reb}}, \quad (51)$$

for a reboiler, and by means of

$$A_{hx} = \frac{-Q_C}{\Delta T_{con} \cdot k_{con}}, \quad (52)$$

for a condenser. ΔT_{reb} and ΔT_{con} are the (logarithmic) mean temperature differences and k_{reb} and k_{con} are the heat transfer coefficients for the reboiler and condenser. For all calculations $k_{reb} = 1 \frac{kW}{m^2K}$ and $k_{con} = 2 \frac{kW}{m^2K}$ are assumed.

2.2 Membrane network

The operational cost of the PV network are composed of the cost of the utilities required for interstage heating and permeate cooling, as well as the cost for membrane replacement.

$$C_{op,M} = t_a \cdot \sum_{ST} (m_{cool,ST} \cdot C_{cool,Perv} + m_{steam,ST} \cdot C_{steam,P}) + C_{replace} \quad (53)$$

The cost calculations for the utilities is similar to those of the distillation column (eqs. (41), (42) and (48)).

The costs for necessary membrane replacement

$$C_{replace} = C_{mem} \cdot \frac{C_{inv,mem}}{t_{mem}}, \quad (54)$$

are based on the membrane investment cost for $C_{inv,mem}$ and the average lifetime of the membranes t_{mem} .

The investment costs for the membrane network

$$C_{inv,M} = MF_P \cdot (C_{inv,mem} + C_{inv,mod}) + \sum_{st} C_{inv,st,hx}, \quad (55)$$

are based on the investment costs for the membrane material $C_{inv,mem}$, the membrane modules $C_{inv,mod}$ and the heat exchangers for preheating and permeate condensation. The heat

exchanger costs are calculated similar to the distillation column. The investment cost for membrane material and modules

$$C_{inv,mem} = C_{mem} \cdot \sum_{st} (w_{st} \cdot l_{st}), \quad (56)$$

$$C_{inv,mod} = C_{mod} \cdot \sum_{st} (w_{st} \cdot l_{st})^{0.7}, \quad (57)$$

are based on the required membrane area (given by the width w_{st} and length l_{st}). The cost parameters for the membrane network are listed in Table 19.

Table 19: Cost parameters for the membrane network

C_{mem} [€/m ²]	250
t_{mem} [a]	2 / 4
C_{mod} [€/m ²]	200
MF_{mem}	3,83

While a membrane lifetime of 2 years is considered for the first two case studies, a membrane lifetime of 4 years is considered for the last case study.

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