Digital Supplementary Material

Synthesis of Liquid Mixture Separation Networks Using Multi-Material Membranes

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S.1. DAE Model Reformulation

The DAE model can be reformulated into differential equations. Dividing Eq. (1) by its summations over $i \in \mathbf{I}$, we derive the term $dF_i^{\mathrm{F}}/d\dot{F}^{\mathrm{F}}$, thereby resulting in the following equation:

$$\frac{dF_i^{\mathrm{F}}}{d\dot{F}^{\mathrm{F}}} = \frac{\pi_i\left(G_i\hat{X}_i - \tau_i\hat{Y}_i\right)}{\sum_{i'\in\mathbf{I}}\pi_{i'}(G_{i'}\hat{X}_{i'} - \tau_{i'}\hat{Y}_{i'})_i} = Y_i \qquad i \in \mathbf{I} \qquad \text{(SM. 1)}$$

Subsequently, we use Eq. (SM. 1) and express the resulting equation in terms of the molar flowrate of component i on the feed-side, F_i^{F}, and the feed molar flowrate, $\dot{F}$:

$$Y_i = \left(\frac{\pi_i G_i}{\sum_{i'\in\mathbf{I}}\pi_{i'}\left(G_{i'}\hat{X}_{i'} - \tau_{i'}\hat{Y}_{i'}\right) + \pi_i\tau_i}\right)\frac{F_i^{\mathrm{F}}}{\dot{F}^{\mathrm{F}}} \qquad i \in \mathbf{I} \qquad \text{(a)}$$

$$\frac{dF_i^{\mathrm{F}}}{d\dot{F}^{\mathrm{F}}} = \left(\frac{\pi_i G_i}{\sum_{i'\in\mathbf{I}}\pi_{i'}\left(G_{i'}\hat{X}_{i'} - \tau_{i'}\hat{Y}_{i'}\right) + \pi_i\tau_i}\right)\frac{F_i^{\mathrm{F}}}{\dot{F}^{\mathrm{F}}} \qquad i \in \mathbf{I} \qquad \text{(b)}$$

$$\frac{dF_i^{\mathrm{F}}}{F_i^{\mathrm{F}}} = \left(\frac{\pi_i G_i}{B + \pi_i\tau_i}\right)\frac{d\dot{F}(1-\bar{C})}{\dot{F}(1-\bar{C})} \qquad i \in \mathbf{I} \qquad \text{(c)}$$

$$\frac{dF_i^{\mathrm{F}}}{F_i^{\mathrm{F}}} = -\left(\frac{\pi_i G_i}{B + \pi_i\tau_i}\right)\frac{d\bar{C}}{(1-\bar{C})} \qquad i \in \mathbf{I} \qquad \text{(d)}$$

Eq. (d) is then integrated across the membrane surface from $\bar{C} = 0, F_i^{\mathrm{F}} = F_i$ to $\bar{C} = C, F_i^{F} = L_i$, where C represents the final stage cut and L_i denotes the retentate molar flowrate. Additionally, the collective driving force at the entry and exit of the membrane unit is denoted as B^0 and B^{L}, respectively.

$$\int_{F_i^{\mathrm{F}}=F_i}^{F_i^{\mathrm{F}}=L_i}\frac{dF_i^{\mathrm{F}}}{F_i^{\mathrm{F}}} = -\int_{\bar{C}=0,B=B^0}^{\bar{C}=C,B=B^{\mathrm{L}}}\left(\frac{\pi_i G_i}{B + \pi_i\tau_i}\right)\frac{d\bar{C}}{(1-\bar{C})} \qquad i \in \mathbf{I} \qquad \text{(e)}$$

Under the stated assumption of uniform driving force and uniform activity coefficients across the membrane surface, Eq. (e) can be integrated to obtain the following set of nonlinear algebraic equations:

$$\ln\left(\frac{L_i}{F_i}\right) = \left(\frac{\pi_i G_i}{B + \pi_i\tau_i}\right)\ln(1-C) \qquad i \in \mathbf{I} \qquad \text{(SM. 2)}$$

$$\sum_{i\in\mathbf{I}} L_i = (1-C)\sum_{i\in\mathbf{I}} F_i \qquad \text{(SM. 3)}$$

Finally, to compute the required membrane area, we take a summation over $i \in \mathbf{I}$ of Eq. (1), substitute B, and then integrate the resulting differential equation from $\hat{A} = 0$ and $\dot{F}^{\mathrm{F}} = \dot{F}$ to $\hat{A} = A$ and $\dot{F}^{F} = \dot{L}$.

$$\int_{\dot{F}^{\mathrm{F}}=\dot{F}}^{\dot{F}^{\mathrm{F}}=\dot{L}} d\dot{F}^{\mathrm{F}} = \int_{\hat{A}=0}^{\hat{A}=A} B\,d\hat{A} \qquad \text{(f)}$$

$$A = \frac{1}{B}\left(\dot{F} - \dot{L}\right) \quad \text{(SM. 4)}$$

S.2. Supplemental Table

Table S1. Simulation data for NLAE model validation.

Input	Value
Components	Methyl Acetate, Ethyl Acetate, Isopropanol, 1-Butanol
Permeance (in mol m^2 s^{-1})	0.041/0.108/0.030/0.183
Component feed flow rate (in mol s^{-1})	0.2/0.2/0.3/0.3
Operating temperature (in K)	323.15
Feed-side/permeate-side pressure (in atm)	1.00/0.01

S.3. Mathematical Formulation

S.3.1. Nomenclature

Sets

$i \in \mathbf{I}$	chemical components
$n \in \mathbf{N}$	membrane stages
$m \in \mathbf{M}$	membrane materials/types

Subsets

$\mathbf{I^P} \subset \mathbf{I}$	target components
$\mathbf{N}_n^{>} \subset \mathbf{N}$	membrane stages after stage n
$\mathbf{N}_n^{<} \subset \mathbf{N}$	membrane stages before stage n

Parameters

α^{UB}	upper bound on required membrane area per stage
$\theta_{n,m}$	operating temperature at stage n of membrane m
$\kappa_{i,m}^{\mathrm{G}}$	gas permeability of component i in membrane m
$\delta_{n,m}$	material thickness at stage n of membrane m
$\phi_{n,m}^{\mathrm{F}}/\phi_{n,m}^{\mathrm{P}}$	feed-side/permeate-side pressure at stage n of membrane m
$\phi_{i,n,m}^{\mathrm{SAT}}$	saturation pressure of component i at stage n of membrane m
$\varphi_i^{\mathrm{PUR}}/\varphi_i^{\mathrm{REC}}$	purity/recovery for target component i
ϑ_i	partial molar volume of component i
$\mu_{i,j}/\gamma_{i,j}$	binary parameters in activity coefficient model

$\zeta_i^{\mathrm{LB}}/\zeta_i^{\mathrm{UB}}$	lower/upper bound on activity coefficient of component i
$\xi^{\mathrm{LB}}/\xi^{\mathrm{UB}}$	lower/upper bound on the complement of stage cut
$\Psi^{\mathrm{LB}}/\Psi^{\mathrm{UB}}$	lower/upper bound on total molar flowrates
$\mathcal{R}$	universal gas constant

Nonnegative Continuous Variables

$A_{n,m}$	required area at stage n of membrane m
$B_{n,m}$	effective driving force at stage n of membrane m
$\hat{C}_{n,m}$	fraction of feed molar flowrate that leaves as retentate
$D_{i,n,m}$	fraction of feed molar flowrate of component i that leaves as retentate
$D_{i,n,m}^{\mathrm{LB}}/D_{i,n,m}^{\mathrm{UB}}$	lower/upper bound on the fraction of feed molar flowrate of component i that leaves as retentate
$E_{i,n,m}/H_{i,n,m}$	auxiliary variables for crossflow membrane
$F_{i,n,m}/V_{i,n,m}/L_{i,n,m}$	component i molar flowrate in feed/permeate/retentate at stage n of membrane m
$\dot{F}_{n,m}/\dot{V}_{n,m}/\dot{L}_{n,m}$	total molar flowrate in feed/permeate/retentate at stage n of membrane m
$\tilde{F}_{i,m}^{\mathrm{F}}$	disaggregated fresh feed component i molar flowrate fed to sub-network of membrane m
$G_{i,n,m}^{\mathrm{F}}$	feed-side activity coefficient of component i at stage n of membrane m
$S_{n,n',m}^{\mathrm{PRC}}/S_{n,n',m}^{\mathrm{RRC}}$	split fraction for permeate/retentate recycle between stage n and n' of membrane m
$S_{n,n',m}^{\mathrm{PBP}}/S_{n,n',m}^{\mathrm{RBP}}$	split fraction for permeate/retentate bypass between stage n and n' of membrane m
$S_{n,m}^{\mathrm{PP}}/S_{n,m}^{\mathrm{RP}}$	split fraction for permeate/retentate product from stage n of membrane m
$V_{i,n,n',m}^{\mathrm{PBP}}/L_{i,n,n',m}^{\mathrm{RBP}}$	component i molar flowrate in permeate/retentate bypass from stage n to n' of membrane m
$V_{i,n,n',m}^{\mathrm{PRC}}/L_{i,n,n',m}^{\mathrm{RRC}}$	component i molar flowrate in permeate/retentate recycle from stage n to n' of membrane m
$V_{i,n,m}^{\mathrm{PP}}/L_{i,n,m}^{\mathrm{RP}}$	component i molar flowrate in permeate/retentate product collected from stage n of membrane m
$V_{i,m}^{\mathrm{P}}/L_{i,m}^{\mathrm{P}}$	component i molar flowrate in final permeate/retentate product of membrane m

$X^{\mathrm{F}}_{i,n,m}/X^{\mathrm{P}}_{i,n,m}/X^{\mathrm{R}}_{i,n,m}$ component i molar fraction ($\in [0,1]$) in feed/permeate/retentate at stage n of membrane m

Binary Variables

Y_m 1 if the fresh feed first enters membrane m

$Z_{n,m}$ 1 if stage n of membrane type m is active

S.3.2. Model Constraints

S.3.2.1. NLAE Unit Model-based Constraints

The following constraints describe the crossflow membrane unit at stage n of membrane type m:

$$L_{i,n,m} = D_{i,n,m}F_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 5)}$$

$$E_{i,n,m} = \frac{B_{n,m}}{\pi_{i,m}} + \tau_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 6)}$$

$$H_{i,n,m}G_{i,n,m} = E_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 7)}$$

$$H_{i,n,m}\ln\left(D_{i,n,m}\right) = \ln\left(\hat{C}_{n,m}\right) \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 8)}$$

$$F_{i,n,m} = L_{i,n,m} + V_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 9)}$$

$$\dot{F}_{n,m} = \sum_{i \in \mathbf{I}} F_{i,n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 10)}$$

$$\dot{L}_{n,m} = \sum_{i \in \mathbf{I}} L_{i,n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 11)}$$

$$\dot{V}_{n,m} = \sum_{i \in \mathbf{I}} V_{i,n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 12)}$$

$$\dot{F}_{n,m}X^{\mathrm{F}}_{i,n,m} = F_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 13)}$$

$$\dot{L}_{n,m}X^{\mathrm{R}}_{i,n,m} = L_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 14)}$$

$$\dot{V}_{n,m}X^{P}_{i,n,m} = V_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 15)}$$

$$\ln\left(G_{i,n,m}\right) = \frac{\sum_{j \in \mathbf{I}} \mu_{j,i}\gamma_{j,i}X^{\mathrm{R}}_{j,n,m}}{\sum_{j \in \mathbf{I}} \gamma_{j,i}X^{\mathrm{R}}_{j,n,m}} + \sum_{j \in \mathbf{I}} \frac{\gamma_{i,j}X^{\mathrm{R}}{}_{j,n,m}}{\sum_{i' \in \mathbf{I}} \gamma_{i'j}X^{\mathrm{R}}_{i',n,m}} \left(\gamma_{i,j} - \frac{\sum_{i' \in \mathbf{I}} \mu_{i',j}\gamma_{i'j}X^{\mathrm{R}}_{i',n,m}}{\sum_{i' \in \mathbf{I}} \gamma_{i'j}X^{\mathrm{R}}_{i',n,m}}\right) \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 16)}$$

$$\dot{L}_{n,m} = \hat{C}_{n,m}\dot{F}_{n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 17)}$$

$$\dot{V}_{n,m} = B_{n,m} A_{n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 18)}$$

S.3.2.2. Superstructure-based Constraints

The material balance around the final product mixers is given by:

$$L^{\mathrm{P}}_{i,m} = \sum\nolimits_{n \in \mathbf{N}} L^{\mathrm{RP}}_{i,n,m} \qquad i \in \mathbf{I}, m \in \mathbf{M} \qquad \text{(SM. 19)}$$

$$V^{\mathrm{P}}_{i,m} = \sum\nolimits_{n \in \mathbf{N}} V^{\mathrm{PP}}_{i,n,m} \qquad i \in \mathbf{I}, m \in \mathbf{M} \qquad \text{(SM. 20)}$$

The material balance constraint around the splitter for retentate and permeate leaving stage n are:

$$L_{i,n,m} = L^{\mathrm{RP}}_{i,n,m} + \sum\nolimits_{n' \in \mathbf{N}^{>}_{n}} L^{\mathrm{RBP}}_{i,n,n',m} + \sum\nolimits_{n' \in \mathbf{N}^{<}_{n}} L^{\mathrm{RRC}}_{i,n,n',m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 21)}$$

$$V_{i,n,m} = V^{\mathrm{PP}}_{i,n,m} + \sum\nolimits_{n' \in \mathbf{N}^{>}_{n}} V^{\mathrm{PBP}}_{i,n,n',m} + \sum\nolimits_{n' \in \mathbf{N}^{<}_{n}} V^{\mathrm{PRC}}_{i,n,n',m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 22)}$$

Split fraction constraints to ensure the component concentration in all retentate/permeate outgoing streams are the same:

$$S^{\mathrm{RP}}_{n,m} + \sum\nolimits_{n' \in \mathbf{N}^{>}_{n}} S^{\mathrm{RBP}}_{n,n',m} + \sum\nolimits_{n' \in \mathbf{N}^{<}_{n}} S^{\mathrm{RRC}}_{n,n',m} = Z_{n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 23)}$$

$$S^{\mathrm{PP}}_{n,m} + \sum\nolimits_{n' \in \mathbf{N}^{>}_{n}} S^{\mathrm{PBP}}_{n,n',m} + \sum\nolimits_{n' \in \mathbf{N}^{<}_{n}} S^{\mathrm{PRC}}_{n,n',m} = Z_{n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 24)}$$

The material balance around the feed mixer is as follows:

$$\begin{aligned} F_{i,n,m} = \; & \tilde{F}_{i,m} + \sum\nolimits_{m \in \mathbf{M}} L^{\mathrm{P}}_{i,m}(1 - Y_m) + \sum\nolimits_{n' \in \mathbf{N}^{>}_{n}} \left(V^{\mathrm{PRC}}_{i,n',n,m} + L^{\mathrm{RRC}}_{i,n,'n,m}\right) \\ & + \sum\nolimits_{n' \in \mathbf{N}^{<}_{n}} \left(V^{\mathrm{PBP}}_{i,n',n,m} + L^{\mathrm{RBP}}_{i,n',n,m}\right) \end{aligned} \qquad \begin{array}{l} i \in \mathbf{I}, \\ n = 1, \\ m \in \mathbf{M} \end{array} \qquad \text{(SM. 25)}$$

$$F_{i,n,m} = \sum\nolimits_{n' \in \mathbf{N}^{>}_{n}} \left(V^{\mathrm{PRC}}_{i,n',n,m} + L^{\mathrm{RRC}}_{i,n,'n,m}\right) + \sum\nolimits_{n' \in \mathbf{N}^{<}_{n}} \left(V^{\mathrm{PBP}}_{i,n',n,m} + L^{\mathrm{RBP}}_{i,n',n,m}\right) \qquad \begin{array}{l} i \in \mathbf{I}, \\ n \in \mathbf{N} \backslash \{\mathbf{1}\}, \\ m \in \mathbf{M} \end{array} \qquad \text{(SM. 26)}$$

The constraints describing the retentate/permeate bypass, recycle, and product stream are as follows:

$$L^{\mathrm{RRC}}_{i,n,n',m} = S^{\mathrm{RRC}}_{n,n',m}\, L_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, n' \in \mathbf{N}^{<}_{n}, m \in \mathbf{M} \qquad \text{(SM. 27)}$$

$$L^{\mathrm{RBP}}_{i,n,n',m} = S^{\mathrm{RBP}}_{n,n',m}\, L_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, n' \in \mathbf{N}^{>}_{n}, m \in \mathbf{M} \qquad \text{(SM. 28)}$$

$$V^{\mathrm{PRC}}_{i,n,n',m} = S^{\mathrm{PRC}}_{n,n',m}\, V_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, n' \in \mathbf{N}^{<}_{n}, m \in \mathbf{M} \qquad \text{(SM. 29)}$$

$$V^{\mathrm{PBP}}_{i,n,n',m} = S^{\mathrm{PBP}}_{n,n',m}\, V_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, n' \in \mathbf{N}^{>}_{n}, m \in \mathbf{M} \qquad \text{(SM. 30)}$$

$$L_{i,n,m}^{\mathrm{RP}} = S_{n,m}^{\mathrm{RP}}\, L_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 31)}$$

$$V_{i,n,m}^{\mathrm{PP}} = S_{n,m}^{\mathrm{PP}}\, V_{i,n,m} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 32)}$$

To avoid symmetrical solutions, we use the following constraint to deactivate the succeeding stages if the current stage is inactive:

$$Z_{n+1,m} \leq Z_{n,m} \qquad n \in \mathbf{N}\backslash\{|\mathbf{N}|\}, m \in \mathbf{M} \qquad \text{(SM. 33)}$$

Since the fresh feed can enter any one of the sub-networks at the first stage, we include the following constraints:

$$\sum_{m \in \mathbf{M}} Y_m = 1 \qquad \text{(SM. 34)}$$

Additionally, we disaggregate the component i molar flowrate F_i^{F} into $\tilde{F}_{i,m}$:

$$F_i^{\mathrm{F}} = \sum_{m \in \mathbf{M}} \tilde{F}_{i,m} \qquad i \in \mathbf{I} \qquad \text{(SM. 35)}$$

$$\sum_{i \in \mathbf{I}} \tilde{F}_{i,m} \leq \Psi^{\mathrm{UB}} Y_m \qquad m \in \mathbf{M} \qquad \text{(SM. 36)}$$

Big-M constraints are introduced so that the molar flowrate and the membrane area can only be positive when the corresponding stage is active:

$$\Psi^{\mathrm{LB}} Z_{n,m} \leq \sum\nolimits_{i \in \mathbf{I}} F_{i,n,m} \leq \Psi^{\mathrm{UB}} Z_{n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 37)}$$

$$\Psi^{\mathrm{LB}} Z_{n,m} \leq \sum\nolimits_{i \in \mathbf{I}} L_{i,n,m} \leq \Psi^{\mathrm{UB}} Z_{n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 38)}$$

$$\Psi^{\mathrm{LB}} Z_{n,m} \leq \sum\nolimits_{i \in \mathbf{I}} V_{i,n,m} \leq \Psi^{\mathrm{UB}} Z_{n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 39)}$$

$$A_{n,m} \leq \alpha^{\mathrm{UB}} Z_{n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 40)}$$

Finally, the objective function, Ω, is defined as follows:

$$\Omega = \sum_{m \in \mathbf{M}} \sum_{n \in \mathbf{N}} A_{n,m} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 41)}$$

S.3.2.3. Variable Bounds & Tightening Constraints

The following variable bounds are incorporated in the model:

$$\xi^{\mathrm{LB}} \leq \hat{C}_{n,m} \leq \xi^{\mathrm{UB}} \qquad n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 42)}$$

$$\zeta_i^{\mathrm{LB}} \leq G_{i,n,m} \leq \zeta_i^{\mathrm{UB}} \qquad i \in \mathbf{I}, n \in \mathbf{N}, m \in \mathbf{M} \qquad \text{(SM. 43)}$$

$$\left(\min_{i\in\mathbf{I}}\pi_{i,m}\right)Z_{n,m}\le B_{n,m}\le\left(\max_{i\in\mathbf{I}}\pi_{i,m}\right)Z_{n,m}\qquad n\in\mathbf{N},m\in\mathbf{M}\qquad\text{(SM. 44)}$$

$$\frac{\left(\min_{i\in\mathbf{I}}\pi_{i,m}\right)}{\pi_{i,m}}Z_{n,m}+\min_{i\in\mathbf{I}}\tau_{i,n.m}\le E_{i,n,m}\le\frac{\left(\max_{i\in\mathbf{I}}\pi_{i,m}\right)}{\pi_{i,m}}Z_{n,m}+\max_{i\in\mathbf{I}}\tau_{i,n,m}\qquad\begin{matrix}i\in\mathbf{I},\\ n\in\mathbf{N},\\ m\in\mathbf{M}\end{matrix}\qquad\text{(SM. 45)}$$

$$\frac{1}{\zeta_i^{\mathrm{UB}}}\left(\frac{\left(\min_{i\in\mathbf{I}}\pi_{i,m}\right)}{\pi_{i,m}}Z_{n,m}+\min_{i\in\mathbf{I}}\tau_{i,n,m}\right)\le H_{i,n,m}\le\frac{1}{\zeta_i^{\mathrm{LB}}}\left(\frac{\left(\max_{i\in\mathbf{I}}\pi_{i,m}\right)}{\pi_{i,m}}Z_{n,m}+\max_{i\in\mathbf{I}}\tau_{i,n,m}\right)\qquad\text{(SM. 46)}$$

$$\xi^{\mathrm{LB}\left(\frac{\zeta_i^{\mathrm{LB}}}{\left(\frac{\left(\max_{i\in\mathbf{I}}\pi_{i,m}\right)}{\pi_{i,m}}Z_{n,m}+\max_{i\in\mathbf{I}}\tau_{i,n,m}\right)}\right)}\le D_{i,n,m}\le\xi^{\mathrm{UB}\left(\frac{\zeta_i^{\mathrm{UB}}}{\left(\frac{\left(\min_{i\in\mathbf{I}}\pi_{i,m}\right)}{\pi_{i,m}}Z_{n,m}+\min_{i\in\mathbf{I}}\tau_{i,n,m}\right)}\right)}\qquad\text{(SM. 47)}$$

$$\ln\left(\nu_{i,n,m}^{\mathrm{LB}}\right)H_{i,n,m}\le\ln\left(\hat{C}_{n,m}\right)\le\ln\left(\nu_{i,n,m}^{\mathrm{UB}}\right)H_{i,n,m}\qquad i\in\mathbf{I},n\in\mathbf{N},m\in\mathbf{M}\qquad\text{(SM. 48)}$$

$$\nu_{i,n,m}^{\mathrm{LB}}F_{i,n,m}\le L_{i,n,m}\le\nu_{i,n,m}^{\mathrm{UB}}F_{i,n,m}\qquad i\in\mathbf{I},n\in\mathbf{N},m\in\mathbf{M}\qquad\text{(SM. 49)}$$

$$\xi^{\mathrm{LB}}\dot{F}_{n,m}\le\dot{L}_{n,m}\le\xi^{\mathrm{UB}}\dot{F}_{n,m}\qquad n\in\mathbf{N},m\in\mathbf{M}\qquad\text{(SM. 50)}$$

$$\left(\frac{\Psi^{\mathrm{LB}}}{\max_{i\in\mathbf{I}}\pi_{i,m}}\right)Z_{n,m}\le A_{n,m}\le\left(\frac{\Psi^{\mathrm{UB}}}{\min_{i\in\mathbf{I}}\pi_{i,m}}\right)Z_{n,m}\qquad n\in\mathbf{N},m\in\mathbf{M}\qquad\text{(SM. 51)}$$

Finally, the following tightening constraints are derived:

$$\ln\left(\xi^{\mathrm{LB}}\right)+\left(\frac{\ln\left(\xi^{\mathrm{LB}}\right)}{\xi^{\mathrm{UB}}-\xi^{\mathrm{LB}}}\right)\left(\xi^{\mathrm{LB}}-\hat{C}_{n,m}\right)\le\ln\left(\hat{C}_{n,m}\right)\le\hat{C}_{n,m}-1\qquad n\in\mathbf{N},m\in\mathbf{M}\qquad\text{(SM. 52)}$$

$$\begin{aligned}\ln\left(\nu_{i,n,m}^{\mathrm{LB}}\right)+\left(\frac{\ln\left(\nu_{i,n,m}^{\mathrm{LB}}\right)}{\nu_{i,n,m}^{\mathrm{UB}}-\nu_{i,n,m}^{\mathrm{LB}}}\right)\left(\nu_{i,n,m}^{\mathrm{LB}}-D_{i,n,m}\right)&\le\ln\left(D_{i,n,m}\right)\\ &\le D_{i,n,m}-1\end{aligned}\qquad\begin{matrix}i\in\mathbf{I},n\in\mathbf{N},\\ m\in\mathbf{M}\end{matrix}\qquad\text{(SM. 53)}$$

$$Z_{n,m}\le Y_m+\sum_{n'\in\mathbf{N}_n^{<}}\left(S_{n,n',m}^{\mathrm{RBP}}+S_{n,n',m}^{\mathrm{PBP}}\right)+\sum_{n'\in\mathbf{N}_n^{>}}\left(S_{n,n',m}^{\mathrm{RRC}}+S_{n,n',m}^{\mathrm{PRC}}\right)\qquad\begin{matrix}n=1,\\ m\in\mathbf{M}\end{matrix}\qquad\text{(SM. 54)}$$

$$Z_{n,m}\le\sum_{n'\in\mathbf{N}_n^{<}}\left(S_{n,n',m}^{\mathrm{RBP}}+S_{n,n',m}^{\mathrm{PBP}}\right)+\sum_{n'\in\mathbf{N}_n^{>}}\left(S_{n,n',m}^{\mathrm{RRC}}+S_{n,n',m}^{\mathrm{PRC}}\right)\qquad\begin{matrix}n\in\mathbf{N}\backslash\{\mathbf{1}\},\\ m\in\mathbf{M}\end{matrix}\qquad\text{(SM. 55)}$$

$$\epsilon Z_{n,m}\le S_{n,n',m}^{\mathrm{RRC/PRC}}\le Z_{n,m}\qquad n\in\mathbf{N},n'\in\mathbf{N}_n^{<},m\in\mathbf{M}\qquad\text{(SM. 56)}$$

$$\epsilon Z_{n,m}\le S_{n,n',m}^{\mathrm{RBP/PBP}}\le Z_{n,m}\qquad n\in\mathbf{N},n'\in\mathbf{N}_n^{>},m\in\mathbf{M}\qquad\text{(SM. 57)}$$

$$\epsilon Z_{n,m}\le S_{n,m}^{\mathrm{RP/PP}}\le Z_{n,m}\qquad n\in\mathbf{N},m\in\mathbf{M}\qquad\text{(SM. 58)}$$

S.4. Supplemental Figure

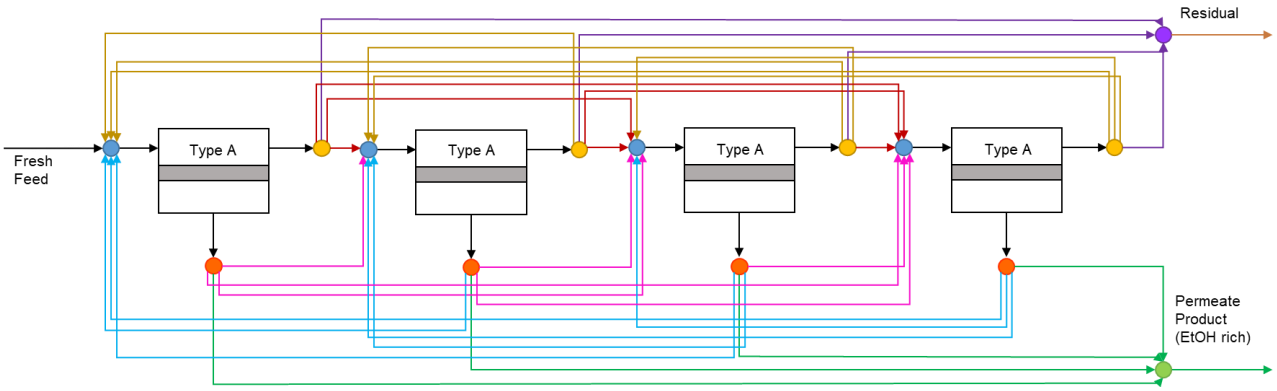

Figure S1: Schematic of a single sub-network superstructure, with four available stages, employing membrane type A.