

# Separation Network Synthesis: Thermally Integrated Distillation Columns

## 1 Definition of Index Sets

1. FEED = state that is the feed to the system  
e.g. FEED = {ABC}
2. COMP =  $\{i \mid i \text{ is a component in the mixture}\}$   
e.g. COMP = {A, B, C}
3. TASKS =  $\{t \mid t \text{ is a separation task}\}$   
e.g. TASKS = {A/BC, AB/C, A/B, B/C}
4. STATES =  $\{s \mid s \text{ is a state/mixture in the network}\}$   
e.g. STATES = {ABC, AB, BC, A, B, C}
5.  $FS_F$  = {columns whose feed is the initial mixture}  
e.g.  $FS_F$  = {A/BC, AB/C}
6.  $TS_s$  = {tasks  $t$  that state  $s$  is able to produce}  
e.g.  $TS_{ABC}$  = {A/BC, AB/C}
7.  $ST_s$  = {tasks  $t$  that are able to produce state  $s$ }  
e.g.  $ST_A$  = {A/BC, A/B}
8.  $RECT_s$  = {tasks  $t$  that produce state  $s$  through a rectifying section}  
e.g.  $RECT_B$  = {B/C}
9.  $STRIP_s$  = {tasks  $t$  that produce state  $s$  through a stripping section}  
e.g.  $STRIP_C$  = {AB/C, B/C}
10.  $LK_t$  =  $\{i \mid i \text{ is the light key component in separation task } t\}$   
e.g.  $LK_{A/BC}$  = A
11.  $HK_t$  =  $\{i \mid i \text{ is the heavy key component in separation task } t\}$   
e.g.  $HK_{A/BC}$  = B
12.  $ISTATE$  =  $\{s \mid s \text{ is an intermediate state in the network}\}$   
e.g.  $ISTATE$  = {AB, BC}
13.  $PRE_i$  = {tasks  $t$  that produce final product  $i$  through a rectifying section}  
e.g.  $PRE_B$  = {B/C}
14.  $PST_i$  = {tasks  $t$  that produce final product  $i$  through a stripping section}  
e.g.  $PST_C$  = {AB/C, B/C}

15.  $IREC_s = \{\text{tasks } t \text{ that produce an intermediate state } s \text{ through a rectifying section}\}$   
e.g.  $IREC_{AB} = \{AB/C\}$
16.  $ISTRIP_s = \{\text{tasks } t \text{ that produce an intermediate state } s \text{ through a stripping section}\}$   
e.g.  $ISTRIP_{BC} = \{A/BC\}$
17.  $ROOTS = \{r \mid r \text{ is an Underwood root}\}$   
e.g.  $ROOTS = \{\phi_1, \phi_2\}$   
*note: a separation task with  $N$  components will have  $N-1$  active Underwood roots*
18.  $RUA_t = \{\text{active Underwood roots for task } t\}$   
e.g.  $RUA_{AB/C} = \{\phi_{1,AB/C}, \phi_{2,AB/C}\}$

## 2 Variables

$FT_t, BT_t, DT_t$ : Total molar flow rates of feed, bottoms, and distillate for task  $t$  in units [kmol/hr]

$F_{i,t}, B_{i,t}, D_{i,t}$ : Component molar flow rates of feed, bottoms, and distillate for component  $i$  and task  $t$  in units [kmol/hr]

$V_{r,t}, L_{r,t}$ : Total vapor and liquid molar flow rates for rectifying section of task  $t$  in units [kmol/hr]

$V_{s,t}, L_{s,t}$ : Total vapor and liquid molar flow rates for stripping section of task  $t$  in units [kmol/hr]

$V_{max,t}$ : Maximum vapor flow rate of the rectifying and stripping sections for task  $t$ .  
 $V_{max,t} = \max\{V_{r,t}, V_{s,t}\}$

$N_{min\ trays,t}$ : Minimum number of trays for a separation task  $t$

$Q_{reb,t}, Q_{cond,t}$ : Reboiler and condenser heat duties for task  $t$  in units [kJ/hr]

$\phi_{r,t}$ : Possible active Underwood root  $r$  in task  $t$

$Y_t$ : Boolean variable for existence of a separation task  $t$

$Z_s$ : Boolean variable for existence of a heat exchanger for an intermediate state  $s$

$W_i$ : Boolean variable for existence of a heat exchanger for a final state  $i$

$z_{i,t,r}$ : Intermediate variable used for reformulation of Underwood equations indexed by components,  $i$ , task,  $t$ , and Underwood roots,  $r$ .

### 2.1 Variable Bounding

Bounding of certain decision variables is critical in obtaining the correct solutions. Specifically, the Underwood roots,  $\phi_{r,t}$ , should be bounded by the relative volatilities of species,  $\alpha_i$ . For example, a separation task for a three component mixture  $ABC$ , will have 2 Underwood roots,  $\phi_1, \phi_2$ . These roots must be bounded by:  $\alpha_A > \phi_1 > \alpha_B > \phi_2 > \alpha_C$ . For given species relative volatilities of  $\alpha_A = 10$ ,  $\alpha_B = 5$ ,  $\alpha_C = 1$ , we enforce the variable bounds  $5.01 \leq \phi_1 \leq 9.99$ ,  $1.01 \leq \phi_2 \leq 4.99$

### 3 Parameters

$F_0, F_{i,0}$ : Total and component inlet molar flows to system in units of [kmol/hr]

$z_i$ : Molar fraction of component  $i$  in feed

$\alpha_i$ : Relative volatility of component  $i$  with respect to the least volatile component in the mixture

$x_i$ : specified recovery of component  $i$

$\lambda_i$ : vaporization enthalpy for species  $i$  in units [kJ/mol]

$C_{cw}, C_h$ : utility cost coefficients for cooling and heating

$N$ : estimated life time of equipment in years

$i$ : interest rate on capital

## 4 Global Constraints

$$\begin{aligned}
\min \quad TAC &= \frac{i(1+i)^N}{(1+i)^N - 1} (\text{Capital Costs}) + \text{Operarting Costs} \\
s.t. \quad &\sum_{t \in FS_F} FT_t = F_0 \\
&\sum_{t \in FS_F} F_{i,t} = F_{0,i} \quad \forall i \in COMP \\
&\sum_{t \in TS_s} FT_t = \sum_{t \in RECT_s} DT_t + \sum_{t \in STRIP_s} BT_t \quad \forall s \in STATES \\
&\sum_{t \in TS_s} F_{i,t} = \sum_{t \in RECT_s} D_{i,t} + \sum_{t \in STRIP_s} B_{i,t} \quad \forall i \in COMP, \quad \forall s \in STATES
\end{aligned}$$

## 5 Column Disjuncts

$$\left[ \begin{array}{l}
Y_t \\
FT_t = DT_t + BT_t \\
DT_t + L_{r,t} = V_{r,t} \\
BT_t + V_{s,t} = L_{s,t} \\
FT_t = \sum_{i \in COMP} F_{i,t} = 0 \\
DT_t = \sum_{i \in COMP} D_{i,t} = 0 \\
BT_t = \sum_{i \in COMP} B_{i,t} = 0 \\
D_{i,t} \geq x_i F_{i,t} \quad i \in LK_t \\
B_{i,t} \geq x_i F_{i,t} \quad i \in HK_t \\
\sum_{i \in COMP} \frac{\alpha_i F_{i,t}}{\alpha_i - \phi_{r,t}} - (V_{r,t} - V_{s,t}) = 0 \quad \forall r \in RU A_t \\
\sum_{i \in COMP} \frac{\alpha_i D_{i,t}}{\alpha_i - \phi_{r,t}} \leq V_{r,t} \quad \forall r \in RU A_t \\
\sum_{i \in COMP} \frac{\alpha_i B_{i,t}}{\alpha_i - \phi_{r,t}} \leq -V_{s,t} \quad \forall r \in RU A_t \\
N_{trays,t} = 2 \cdot N_{min trays,t} \\
Height_t = f(\text{Diameter}, N_{trays}) \\
Area_t = g(\rho_L, \rho_V, V_{r,t}, V_{s,t}, \sigma) \\
Cost_t = h(\text{Diameter}, N_{trays}, P)
\end{array} \right] \quad \vee \quad \left[ \begin{array}{l}
\neg Y_t \\
FT_t = BT_t = DT_t = 0 \\
F_{i,t} = B_{i,t} = D_{i,t} = 0 \quad \forall i \in COMP \\
V_{r,t} = V_{s,t} = 0 \\
L_{r,t} = L_{s,t} = 0 \\
Area_t = Height_t = N_{trays,t} = 0 \\
Cost_t = 0
\end{array} \right]$$

$\forall t \in TASKS$

## 6 Underwood Equation Reformulation

Reformulation of the Underwood equations can be done by introducing an intermediate variable to put the constraints in a quadratic form. The Underwood equations, which are used to calculate the vapor flow rates for a given separation task, can be stated as:

$$\begin{aligned} \sum_{i \in COMP} \frac{\alpha_i F_{i,t}}{\alpha_i - \phi_{r,t}} - (V_{r,t} - V_{s,t}) &= 0 \\ \sum_{i \in COMP} \frac{\alpha_i D_{i,t}}{\alpha_i - \phi_{r,t}} &\leq V_{r,t} \\ \sum_{i \in COMP} \frac{\alpha_i B_{i,t}}{\alpha_i - \phi_{r,t}} &\leq -V_{s,t} \end{aligned}$$

Introducing the variable  $z_{i,t,r}$  as:

$$\begin{aligned} z_{i,t,r} &= \frac{1}{\alpha_i - \phi_{r,t}} \\ z_{i,t,r}(\alpha_i - \phi_{r,t}) &= 1 \end{aligned}$$

Now the four constraints can be written as

$$\begin{aligned} z_{i,t,r}(\alpha_i - \phi_{r,t}) &= 1 \quad \forall i \in COMP \quad \forall r \in RU A_t \\ \sum_{i \in COMP} z_{i,t,r} \alpha_i F_{i,t} - (V_{r,t} - V_{s,t}) &= 0 \quad \forall r \in RU A_t \\ \sum_{i \in COMP} z_{i,t,r} \alpha_i D_{i,t} &\leq V_{r,t} \quad \forall r \in RU A_t \\ \sum_{i \in COMP} z_{i,t,r} \alpha_i B_{i,t} &\leq -V_{s,t} \quad \forall r \in RU A_t \end{aligned}$$

Where  $\alpha_i$  is a parameter while  $z_{i,r,t}$  and the flow rates are decision variables. Thus the constraints are quadratic and can be used to calculate the vapor flow rates in a section for a separation task.

## 7 Intermediate Product Heat Exchanger Disjuncts

$$\left[ \begin{array}{l} Q_{cond,t} DT_t = V_{r,t} \sum_{i \in COMP} \lambda_i D_{i,t} \quad \forall t \in IRECT_s \\ Q_{reb,t} BT_t = V_{s,t} \sum_{i \in COMP} \lambda_i B_{i,t} \quad \forall t \in ISTRIP_s \\ Cost_s = f(Area_s, U, \Delta T_{lm}) \end{array} \right] \vee \left[ \begin{array}{l} \sum_{t \in TS_s} (V_{r,t} - V_{s,t}) - \sum_{t \in RECT_s}^{-Z_s} V_{r,t} + \sum_{t \in STRIP_s} V_{s,t} = 0 \\ \sum_{t \in TS_s} (L_{r,t} - L_{s,t}) - \sum_{t \in RECT_s} L_{r,t} + \sum_{t \in STRIP_s} L_{s,t} = 0 \\ Q_{cond,t} = 0 \quad \forall t \in IRECT_s \\ Q_{rec,t} = 0 \quad \forall t \in ISTRIP_s \\ Cost_s = 0 \end{array} \right]$$

$\forall s \in ISTATE$

## 8 Final Product Heat Exchanger Disjuncts

$$\left[ \begin{array}{l} Q_{cond,t}DT = V_{r,t} \sum_{j \in COMP}^{W_i} \lambda_j D_{j,t} \quad \forall t \in PRE_i \\ Q_{reb,t}BT = V_{s,t} \sum_{j \in COMP} \lambda_j B_{j,t} \quad \forall t \in PST_i \\ Cost_i = f(Area_s, U, \Delta T_{lm}) \end{array} \right] \vee \left[ \begin{array}{l} \neg W_i \\ Q_{cond,t} = 0 \quad \forall t \in PRE_i \\ Q_{reb,t} = 0 \quad \forall t \in PST_i \\ Cost_i = 0 \end{array} \right]$$

$$\forall i \in COMP$$

## 9 Logical Relations

1. A given state  $s$  can give rise to at most one task (i.e. cannot split a stream into two different columns)

$$\sum_{t \in TS_s} Y_t \leq 1 \quad \forall t \in TASKS$$

2. A given state can be produced by at most two tasks: one must be from a rectifying section of a task and one must be from a stripping section of a task

$$\sum_{s \in RECT_s} Y_t \leq 1 \quad \forall s \in STATES$$

$$\sum_{s \in STRIP_s} Y_t \leq 1 \quad \forall s \in STATES$$

3. All products must be produced by at least one task

$$\sum_{t \in (PRE_i \cup PST_i)} Y_t \geq 1 \quad \forall i \in COMP$$

4. Pure product  $i$  can only be produced by at most one rectifying section and one stripping section

$$\sum_{t \in PRE_i} Y_t \leq 1 \quad \forall i \in COMP$$

$$\sum_{t \in PST_i} Y_t \leq 1 \quad \forall i \in COMP$$

5. If a given final product is produced by exactly one task, the heat exchanger associated with the state must be selected

$$\left( \sum_{t \in (PRE_i \cup PST_i)} Y_t = 1 \right) \implies W_i \quad \forall i \in COMP$$

6. If a given final state is produced by 2 tasks, then there is no heat exchanger associated with the state

$$\left(\bigvee_{t \in PRE_i} Y_t\right) \wedge \left(\bigvee_{t \in PST_i} Y_t\right) \implies \neg W_i \quad \forall i \in COMP$$

7. Cannot have a heat exchanger for an intermediate state if the state is not produced by a task

$$1 - Z_s + \sum_{t \in ST_s} Y_t \geq 1 \quad \forall s \in ISTATE$$

8. Connectivity relation 1 (e.g. the existence of task AB/C implies A/B)

$$Y_t \implies \bigvee_{k \in TS_s} Y_k \quad \forall t \in ST_s$$

9. Connectivity relation 2 (e.g. the existence of task A/B implies AB/C)

$$Y_t \implies \bigvee_{k \in TS_s} Y_k \quad \forall t \in TS_s$$

## 10 Column Sizing

### 10.1 Column Area

Calculation procedure from Chapter 9.2 of: Stichlmair, J. G., Klein, H., & Rehfeldt, S. (2021). Distillation Principles and Practice. Newark American Institute Of Chemical Engineers Ann Arbor, Michigan Proquest.

Determination of the column area based on internal molar vapor flow rates, assumed physical properties of sieve tray column, and fixed pressure value for the column.

#### Input Variables

$\dot{V}$  internal vapor flow rate of column in [kmol/sec]. If rectifying and stripping section vapor flow rates are different, use the greater of the two

$\hat{M}$  molecular weight of input mixture [kg/kmol]

$\rho_g$  density of gas [kg/m<sup>3</sup>]

$\rho_L$  density of liquid [kg/m<sup>3</sup>]

$\sigma$  surface tension [N/m]

#### Parameters

*Assumed to be the same for all columns in the network*

$\phi = 0.1$  relative free area of tray

$d_h = 8mm$  diameter of sieve holes in plate

$l_w/D_c = 0.7$  relative weir length

$g = 9.81m/s^2$  gravitational constant

### Calculation Procedure:

(1) Maximum gas load:  $F_{max}$

$$F_{max} = 2.5 \cdot (\phi^2 \cdot (\rho_L - \rho_g) \cdot g)^{1/4}$$

(2) Minimum gas load:  $F_{min}$

Have to consider two criteria for minimum gas load in a column: non-uniform gas flow ( $F_{min,1}$ ) and criterion for weeping ( $F_{min,2}$ ). Take the maximum of the two values.

$$F_{min,1} = \phi \cdot \sqrt{2 \cdot \frac{\sigma}{d_h}}$$

$$F_{min,2} = \phi \cdot \sqrt{0.37 \cdot d_h \cdot g \cdot \frac{(\rho_L - \rho_G)^{1.25}}{\rho_g^{0.25}}}$$

(3) Column Cross-Sectional Area:  $A_c$

Take the geometric mean of the minimum and the maximum gas load to determine  $F$  for the column.

$$u_G = \frac{F}{\sqrt{\rho_G}}$$

$$\dot{V} = \dot{G} \cdot \frac{\hat{M}}{\rho_G}$$

$$A_{ac} = \frac{\dot{V}}{u_G}$$

column cross section is given by:

$$A_c = A_{ac} + 2 \cdot A_d$$

area of the downcomer calculated by:

$$\frac{A_{ac}}{A_c} = 1 - \frac{2}{\pi} \cdot [\arcsin(\frac{l_w}{D_c} - \sqrt{(\frac{l_w}{D_c})^2 - (\frac{l_w}{D_c})^4}]$$

above equation allows calculation of the cross-sectional area  $A_c [m^2]$



## 10.2 Tray Number and Height

The minimum number of trays needed for a separation task is determined using the Fenske equation based on the relative volatilities of the light key and heavy key components for that task. The actual number of trays used is twice the minimum tray number ( $N_{trays} = 2 \cdot N_{trays,min}$ ). Empirical correlation for height per trays from Ch 4 of Ulrich, G. D., & Vasudevan, P. T. (2004). Chemical engineering process design and economics : a practical guide. Durham Process Publishing Cop.

### Input Variables

$x_i$  desired recovery fraction

$\alpha_i$  relative volatility for species  $i$

$D$  column diameter [m]

(1) Fenske Equation

$$N_{trays,min} = \frac{\log(\frac{x_L^2}{(1-x_L)^2})}{\log(\frac{\sum_{i \in LK} \alpha_i}{\sum_{i \in HK} \alpha_i})}$$

$$N_{trays} = 2 \cdot N_{trays,min}$$

(2) Column Height

$$\text{Height per tray: } H_t = 0.5D^{0.3}$$

Above relation was fit with a quadratic expression to formulate problem as an MIQCP.

$$\text{Height per tray: } H_t = 0.411 + 0.826D - 0.002D^2$$

$$\text{Column Height: } Height = H_t \cdot N_{trays}$$

## 11 Calculation of Capital Cost

Calculation procedure from Chapter 7 of: Turton, R., & Al, E. (2009). Analysis, Synthesis, and Design of Chemical Processes. Prentice Hall.

### 11.1 Tower Capital Cost

#### Input Variables

$D$  column diameter [m]

$H$  column height [m]

$P$  gauge pressure of the column [barg]

#### Parameters

*Assumed to be the same for all columns in the network*

$CI_{2004} = 444.2$  Cost index from the Chemical Engineering Plant Cost Index (CEPCI) for the year 2004 (year reference values were collected)

$CI_{2020} = 596.2$  CEPCI value for year 2020

$K_1, K_2$  empirical cost correlation parameters for tray and packed tower

$K_1 = 5307$

$K_2 = 603.8$

$B_1, B_2$  bare module factors vertical process vessels

$B_1 = 2.25$

$B_2 = 1.72$

$F_M = 1$  material factor for a carbon steel vertical vessel

$CA = 0.00315$  corrosion allowance value

$t_{min} = 0.0063$  minimum vessel thickness

#### Calculation Procedure:

(1) Cost correlation:  $C_p^0$

$$C_p^0 = K_1 + K_2 A$$

$A$  is the size factor of the equipment. For vertical process vessels (towers) use the vessel volume.

(2) Pressure factor:  $F_{P,vessel}$

$$F_{P,vessel} = \frac{\frac{(P+1)D}{(2)(944)(0.9)-1.2(P+1)} + CA}{t_{min}}$$

(3) Bare Module Cost:  $C_{BM}$

$$C_{BM} = C_p^0 F_{BM} = C_p^0 (B_1 + B_2 F_M F_P)$$

(4) Inflation Calculation:  $C_{p,2020}$

$$C_{tower,2020} = \frac{CI_{2020}}{CI_{2004}} \cdot C_{BM}$$

## 11.2 Tray Capital Cost

### Input Variables

$N_{trays}$  number of trays in column

### Parameters

*Assumed to be the same for all columns in the network*

$CI_{2004} = 444.2$  Cost index from the Chemical Engineering Plant Cost Index (CEPCI) for the year 2004 (year reference values were collected)

$CI_{2020} = 596.2$  CEPCI value for year 2020

$K_1, K_2, K_3$  empirical cost correlation parameters for sieve trays

$K_1 = 571.1$

$K_2 = 406.8$

$K_3 = 38$

$F_M = 1$  material factor for carbon steel sieve and valve trays

$f_q = 1$  quantity factor for trays for empirical relation

#### Calculation Procedure:

(1) Cost correlation:  $C_p^0$

$$C_p^0 = K_1 + K_2 A + K_3 A^2$$

$A$  is the size factor of the equipment. Use the area of the column for tray cost calculation.

(2) Bare Module Cost:  $C_{BM}$

$$C_{BM} = C_p^0(N_{trays})(F_{BM})(f_q)$$

(3) Inflation Calculation:  $C_{p,2020}$

$$C_{trays,2020} = \frac{CI_{2020}}{CI_{2004}} \cdot C_{BM}$$

### 11.3 Total Columns Capital Cost

$$Cost_{column} = Cost_{tower} + Cost_{trays}$$

## 12 Heat Exchanger Sizing and Costing

Calculation procedure from Chapter 4 of: Ulrich, G. D., & Vasudevan, P. T. (2004). Chemical engineering process design and economics. Durham Process Publishing Corp.

Determination of heat exchanger area based on required heat flow rate, overall heat transfer coefficient, and delta log mean temperature difference.

Values for overall heat transfer coefficient ( $U$ ) from Ulrich table 4-15a.

### Input Variables

$\dot{Q}_{reb/cond}$  Heat flow rate for exchanger (condenser or reboiler) in [kJ/sec]

$\Delta T_{LM} = \frac{\Delta T_h - \Delta T_c}{\ln(\frac{\Delta T_h}{\Delta T_c})}$  log mean temperature difference for the fluids in the exchanger

### Parameters

$U_{reboiler} = 700 [J/m^2 sec K]$  overall heat transfer coefficient for hot side fluid of condensing steam and cold side fluid of low viscosity liquid hydrocarbons

$U_{condenser} = 150 [J/m^2 sec K]$  overall heat transfer coefficient for hot side fluid of hydrocarbon gases and cold side fluid of liquid water

$F_m = 1$  material factor for carbon steel construction heat exchanger

$F_p = 1$  pressure factor for exchanger operating below a pressure of 10 barg

$C_{reboiler}^0 = \$20,000$  base purchase price for kettle reboiler with  $100 m^2$  area with CEPCI = 400

$C_{condenser}^0 = \$15,000$  base purchase price for floating head shell and tube heat exchanger with  $100 m^2$  exchange area with CEPCI = 400

$n = 0.6$  scaling exponent for equipment

### Calculation Procedure:

(1) Area of exchanger

$$\begin{aligned} \text{Unit conversion to [J/sec]: } \dot{Q}_{reb/cond} &= \dot{Q}_{reb/cond} \cdot 1000 \\ Area &= \frac{\dot{Q}}{U \Delta T_{LM}} \quad [m^2] \end{aligned}$$

(2) Cost of exchanger

Determine K values for reboiler and condenser based on given data (Ulrich 5.36) for a carbon steel exchanger with  $100 m^2$  of exchange area

$$K_{reboiler} = \frac{C_{reboiler}^0}{(100 m^2)^n}$$

$$K_{condenser} = \frac{C_{condenser}^0}{(100 m^2)^n}$$

Cost for each exchanger(reboiler / condenser) is then determined by:

$$Cost_{reboiler} = \frac{CI_{2020}}{CI_{2004}} (K_{reboiler} Area^n)$$

$$Cost_{condenser} = \frac{CI_{2020}}{CI_{2004}} (K_{condenser} Area^n)$$

## 13 System Capital Costs

$$CAPEX = \sum_{t \in TASKS} Cost\ Column_t + \sum_{i \in COMP} Cost\ Final\ Heat\ Exchangers_i + \sum_{s \in ISTATE} Cost\ Intermediate\ Heat\ Exchangers_s$$

## 14 Operating Costs

### Input Variables

$Q_{reb,t}$  reboiler heat duty

$Q_{cond,t}$  condenser heat duty

### Parameters

$C_{cw}$  Cost coefficient for cooling utility (cooling water) [\$/kJ]

$C_H$  Cost coefficient for heating utility (steam) [\$/kJ]

$$OPEX = \sum_{t \in TASKS} C_{cw} Q_{cond,t} + C_h Q_{reb,t}$$