Lecture 9

Synthesis of distillation sequences
Given is
- A mixture of chemical components of given composition
- A set of products to separate the mixture

Usual assumption
- The set of products are the chemical components that we wish to separate as pure chemicals
Basic options

1. **Sharp separation** vs **Partial separation**

   ![Diagram 1](image1)

2. **Simple columns** vs **Complex columns**

   ![Diagram 2](image2)

   *One feed, two products*
Types of complex columns – sharp splits

**Direct split**

- Partial condenser
- Side draws
- Pre-fractionation
- Petlyuk

**Indirect split**

- Partial reboiler
- Side draw
Πολυπλοκότητα προβλήματος

- (Thomson & King, 1972)

Available sequences:

\[ N_{\text{seq}} = \frac{2(N-1)!}{N!(N-1)!} \]

- Σημείωση: Not the largest combinatorial problem.
  (The synthesis of HEN offers more options)
- Additional challenges in modelling the sequences

- (Westerberg & Stephanopoulos, 1975)
  (Gomez & Seader, 1976)

- Constant pressures, constant reflux rations, fixed recoveries. Use of B&B methods to select the most efficient distillation sequence

- (Faith & Morari, 1979)

- Development of a synthesis approach for variable pressures
Development of MILP model for non-thermally integrated columns

- Determine light and heavy keys

- For each component L

\[
\begin{align*}
    d_L &= \xi_L f_L \\
    b_L &= (1 - \xi_L) f_L
\end{align*}
\]

$\xi_L$ = product recovery in lights and bottom
Assume high recoveries in all separations (almost 100%)

- Problem simplification:
  - The set of components splits into products recovered on either the distillate or the bottoms product
    \[ C = (C^{top}, C^{bottom}) \]
  - Note: we could generalize in the case of adjacent products using the concept of product recovery
We further assume:

- The heat load of each condenser is identical to that of the reboiler (e.g. that the separation work is negligible in comparison to the heat loads) – good and reasonable assumption in most cases.
- The load is a linear function of the column feed, $F$ (who gives such expressions?)
- The Annualized cost involves CAPEX and OPEX terms (who gives such expressions?)

\[
Q^C = K^C \times F \\
Q^R = K^R \times F \\
Cost = (\alpha y + \beta F) + (C_c Q^C + C_R Q^R)
\]

$C_c$ : κόστος της ψυχρής παροχής
$C_R$ : κόστος της θερμής παροχής
Problem representation – task representation

- (Hendry & Hughes, 1972)
- Illustration:
Sequencing synthesis problem

Problem: 3 components

\[
\begin{array}{c}
\text{A} \\
\text{B} \\
\text{C}
\end{array}
\quad \rightarrow \quad \left\{ \begin{array}{l}
\text{Separate first A from BC; then B from C} \\
\text{Separate AB from C; then A from B}
\end{array} \right.
\]

Problem: 4 components

\[
\begin{array}{c}
\text{F} \\
\text{C} \\
\text{D}
\end{array}
\quad \rightarrow \quad \left\{ \begin{array}{c}
\text{A} \\
\text{B} \\
\text{F} \\
\text{C} \\
\text{D} \\
\text{F}
\end{array} \right.
\]

Energy \( E \)

Cost = \( a + b \cdot F + c \cdot E \)

- One should consider all possible cases: A/BCD, ABC/D; also all different sequences that follow in Hughes’ tree
- Different cost models for each sequence
Sequencing synthesis problem (2)

- One possible case: A/BCD
  - Followed by either
    - B/CD and C/D or
    - BC/D and B/C

- Our previous case: AB/CD
  - Followed by either
    - A/B
    - C/D

- Another case: ABC/D
  - Followed by either
    - AB/C and A/B or
    - A/BC and B/C

- Bear in mind common columns in the decision tree
  Use a superstructure approach to combine all options!
The superstructure involves 10 different separation tasks
Mass balances (1)

Initial node

(ABCD) \[ F_1 + F_2 + F_3 = F \]
Mass balances (2)

Intermediate node

\[(BCD) \quad F_4 + F_5 - \xi_1^{BCD} F_1 = 0\]
Mass balances (3)

Final node

\[(BC) \quad F_9 - \xi_5^{BC} F_5 - \xi_6^{BC} F_6 = 0\]
Logical constraints

\[ y_1 + y_2 + y_3 = 1 \]

\[
\begin{align*}
\{ & y_4 + y_5 \leq 1 \\ & y_6 + y_7 \leq 1 \}
\end{align*}
\]

\[
\begin{align*}
\{ & y_4 - y_1 \leq 0 \\ & y_5 - y_1 \leq 0 \\ & y_6 - y_3 \leq 0 \\ & y_7 - y_3 \leq 0 \}
\end{align*}
\]

\[
\begin{align*}
\{ & y_8 - y_2 - y_4 \leq 0 \\ & y_9 - y_5 - y_6 \leq 0 \\ & y_{10} - y_2 - y_7 \leq 0 \}
\end{align*}
\]

:δεν χρειάζονται γιατί καλύπτονται από τα ισοζύγια μάζας
Generalization (arbitrary set of components)

- Set of intermediate products: \( IP = \{ m \} \)

- Set of columns: \( COL = \{ k \} \)

\[
FS_m = \{ \text{set of distillation columns with intermediate } m \} 
\]

\[
IS_F = \{ \text{set of distillation columns receiving fresh feed} \} 
\]

\[
PS_m = \{ \text{distillation columns producing intermediate } m \} 
\]
Mathematical formulation – general case

\[
\min C = \sum_{k \in \text{COL}} \left\{ (a_k y_k + \beta_k F_k) + C_C Q^C_k + C_R Q^R_k \right\}
\]

Subject to:

\[
\sum_{K \in \text{IFS}_F} F_K = F
\]

\[
\sum_{j \in \text{FS}_m} F_j - \sum_{K \in \text{PS}_m} \xi_k^m F_k = 0, \quad m \in \text{IP}
\]

\[
\begin{cases}
Q^R_k - K_R F_k = 0 \\
Q^C_k - K_C F_k = 0 \\
F_k - U \cdot Y_k \leq 0 \quad & k \in \text{COL} \\
F_k, Q^R_k, Q^C_k \geq 0 \\
y_k \in \{0, 1\}
\end{cases}
\]

Model generic and requires only cost parameters that could be available from simulations or short-cut calculations
How about thermally integrated columns?
Thermally integrated separations

Simple distillation

Towards

Complex columns

✓ Energy costs
The transition to thermally integrated systems

Simple columns

Complex column: Partial condenser

Conversion

Integration
Possible integration scheme on the Hughes tree

Number of hybrids for $n$ components:

$$H_n = \sum_{l=1}^{n-2} \left( l \cdot \sum_{k=1}^{n-l-1} (n-l-k) \right)$$
Complex columns – distribution of intermediates

1️⃣ stage:
Production of intermediates

2️⃣ stage:
Sharp splits

3️⃣ στάδιο:
Sharp splits
Multiple feeds

Mixing products of different compositions
Complex columns – distribution of intermediates

Possible schemes

In distributing intermediates

Number of multiple columns for
a mixture of \( n \) components:

\[
MF_n = \sum_{l=1}^{n-3} \frac{(n-l)(n-l-1)(n-l-2)}{6} \quad \forall n \geq 4
\]
Innovative schemes

Overlapping hybrids

Hybrid 1 + Hybrid 2
Back to the synthesis problem

Set components:
- $h_i$: Hybrids
- $t_i$: task
- $c_i$: Transformation

Basic idea:
1) Besides each sequence: $t_1 - t_4 - t_8$
2) Build combinations: e.g. $h_1$ from $t_1$, $t_4$
3) And a second combination: e.g. $C1$, or $c_2$ or $c_3$ or $c_4$ from $h_1$

Mathematical model:
1) New logical schemes: $h - t - c$
2) New cost models for transformations
3) Energy savings for transformations
Energy discount 0.5 M€/yr

Without integration the A/B/CD yields 8 M€/yr

Illustration example: t1 – t4 – t8

Energy cost

Choice: Petlyuk

Total Cost = 5+3+1 = 9 M€/yr

Revised energy cost = 7.5+1 = 8.5 M€/yr

h1 = 7.5 M€/yr

t8 = 1 M€/yr
\[ E = \text{energy cost} \]

\[ S = \text{discount from hybrids} \]

**Sharp splits**

\[ E_{t1} = 5 \text{ M€/yr} \]

\[ E_{t2} = 3 \text{ M€/yr} \]

\[ E_{t3} = 1 \text{ M€/yr} \]

**Use of hybrids**

\[ E_{h1} = 7.5 \text{ M€/yr} \]

\[ E_{t3} = 1 \text{ M€/yr} \]

**Discount in using hybrid \( h_1 \)**

\[ S_{h1} = E_{t1} + E_{t2} - E_{h1} \]

\[ S_{h1} = 5 + 3 - 7.5 = 0.5 \]

**Total cost** =  

\[ \text{Obj} = E_{t1} + E_{t2} + E_{t3} - S_{h1} \]
\[
\begin{align*}
E_{t1} &= 4 \\
E_{t2} &= 12 \\
E_{t3} &= 8 \\
E_{t4} &= 3
\end{align*}
\]

Cost = \(4 + 12 + 8 + 3 = 27\)

Discount = 0

Total cost = \(27 - 0 = 27\)

\[
\begin{align*}
H_{1, petlyuk} &= 14 \\
E_{t1} &= 4 \\
E_{t2} &= 12 \\
E_{t3} &= 8 \\
E_{t4} &= 3
\end{align*}
\]

Cost = \(4 + 12 + 8 + 3 = 27\)

Discount = \(4 + 12 - 14 = 2\)

Total cost = \(27 - 2 = 25\)

\[
\begin{align*}
H_{2, petlyuk} &= 15 \\
E_{t1} &= 4 \\
E_{t2} &= 12 \\
E_{t3} &= 8 \\
E_{t4} &= 3
\end{align*}
\]

Cost = \(4 + 12 + 8 + 3 = 27\)

Discount = \(12 + 8 - 15 = 5\)

Total cost = \(27 - 5 = 22\)

\[
\begin{align*}
H_{3, petlyuk} &= 10.5 \\
E_{t1} &= 4 \\
E_{t2} &= 12 \\
E_{t3} &= 8 \\
E_{t4} &= 3
\end{align*}
\]

Cost = \(4 + 12 + 8 + 3 = 27\)

Discount = \(8 + 3 - 10.5 = 0.5\)

Total cost = \(27 - 0.5 = 26.5\)
Total cost = Cost of sequence – Savings from hybrid/transformation
Total cost = 

Cost of sequence = \( E_t_1 + E_t_2 + E_t_3 \) – Savings = \( S_{h_1,c_3} \)

Generalization:

\[
\text{min} \quad Obj = \sum_{t \in T} E_t - \sum_{h \in H} \sum_{c \in C} S_{h,c}
\]
Generalization of the representation concept

Cost $E$ (parameter) may refer to:
- Energy cost
- Steam load
- Heat duties (condenser, reboiler)
- Total cost
- Combinations of the above

Discount, $S_{h,c}$ (parameter), in using transformation $c$
from hybrid $h$ comprised by $[t]$ transformations

$$\sum_{t \in T_h} E_t - E_{h,c} = S_{h,c} \quad \forall h \in \text{Hybrids}, \ c \in \text{Transformations}$$

Cost of possible sequence
Cost of transformation
Discount in using transformation $c$
From hybrid $h$
Shortcut calculations to assess parameters

Basic design parameters affecting the cost

- **Number of stages (NN)**: column size → cost
- **Reflux ratio (RR)**: vapor → operating cost

Cost calculation

- Simple columns
- Side draws
- Pre-fractionator
- Petlyuk
- Side reboilers

Fixed parameters
- Feed composition
- Product specs
- Operating pressure

Background data
- Thermodynamics (Antoine coefficients)
- Equilibria (UNIQUAC, NRTL)

Cost (Ε) column/hybrid
- Energy
- Construction
- Operation

Cost models for different cases

- Purchased Cost, $S = \frac{M \times S}{280}(101.3A^{0.85}F_t)$
- Purchased Cost, $S = \frac{M \times S}{280}(101.9D^{1.06}H^{0.82}F_t)$
- $Q_c = \Delta H_i \cdot V = U_c A_c \Delta T_n = w_e C_p(t_{max} - 90)$
Illustration example

\[ Q_{\text{condenser}} = \Delta H \frac{V}{\bar{V}} \cdot V \]
\[ Q_{\text{reboiler}} = \Delta H \frac{V}{\bar{V}} \]

Height (H) & Diameter (D_r) = f(\text{NN, V})

\[ H = \frac{2N}{F_0} + H_0 \]
\[ A_r = \frac{V/\rho_m}{1.5(3600)/\sqrt{M_D/\rho_m}} = \frac{V_1}{\sqrt{M_A/\rho_m}} \]
\[ D_r = 0.0164\sqrt{V} \left( \frac{M_A}{\rho_m} \right)^{1/8} \]

(Douglas, 1988)

Cost for heating, cooling

Investment cost

\[ S_{\text{υβρίδιο}} = E_{\text{στήλης}} - E_{\text{υβρίδιο}} \]

\[ E_{\text{στήλης/υβρίδιο}} = \]

- ενέργεια
- επένδυση,
- ενέργεια + ετησ. επένδυση

Εναλλακτικά
\[ E = K_0 + K_1 \cdot N_r + K_2 \cdot V \]
Tasks, hybrids and transformations

**Hybrids**
- $h_1 : t_{1} - t_{4}$
- $h_2 : t_{1} - t_{5}$
- $h_3 : t_{2} - t_{10}$
- $h_4 : t_{4} - t_{8}$
- $h_5 : t_{6} - t_{9}$
- $h_6 : t_{2} - t_{8}$
- $h_7 : t_{3} - t_{6}$
- $h_8 : t_{3} - t_{7}$
- $h_9 : t_{5} - t_{9}$
- $h_{10} : t_{7} - t_{10}$

**Transformations**
- $c_1 : $ Side condenser
- $c_2 : $ Side draw
- $c_3 : $ Pre-fractionator
- $c_4 : $ Petlyuk
- $c_5 : $ Side reboiler
- $c_6 : $ Side draw

**Decision variables**
$$
\gamma_{h_1,c_1} \quad \gamma_{h_1,c_2} \quad \gamma_{h_1,c_3} \ldots \quad \gamma_{h_3,c_4} \ldots \\
\gamma_{h_9,c_5} \quad \gamma_{h_6,c_6} \ldots \gamma_{h_{10},c_5} \quad \gamma_{h_{10},c_6}
$$

**Discount calculations**

**Minimize costs**
Mathematical modelling

Basic sequencing problem

Column selection: \textbf{Decision variable}

Example: \[ f_{t4} \leq Y_{t4} \cdot U_{t4} \]

\( U_t \) : Large number

Special cases

\[ Y_{t4} = 0 \implies f_{t4} \leq 0 \implies f_{t4} = 0 \]

Special cases

\[ Y_{t4} = 1 \implies f_{t4} \leq U_{t4} \implies f_{t4} \neq 0 \]

Otherwise \( Y_{t4} \)

Burdens the objective function

Generalization:

\textbf{Equation 1} \[ f_t \leq Y_t \cdot U_t \quad \forall t \in T \]
Mathematical models

Basic sequencing

Feed balance:

\[ f_{t1} + f_{t2} + f_{t3} = F_{feed}^{\text{feed}} \]

Intermediates:

\[ F_{\text{tot}}^{\text{tot}} = \sum_{t \in T_{\text{feed}}} f_t \]
Mathematical modelling

Basic sequencing

$J_{g,t}$: Part of feed $t$ that produces $g$

Example 1
for group BCD: $f_{t1} \cdot J_{g2,t1} = f_{t4} + f_{t5}$

Example 2
for group CD: $f_{t2} \cdot J_{g4,t2} + f_{t4} \cdot J_{g4,t4} = f_{t8}$

Generalization – all combinations:

**Equation 3**

$$\sum_{t \in T_{m}^{output}} f_{t} \cdot J_{g,t} = \sum_{k \in T_{m}^{input}} f_{k} \quad \forall g \in G$$

Columns that produce $g$ Columns that receive $g$
Mathematical modelling

Example:
The sequence $t_4 - t_8$ corresponds to $h_4$.

1 hybrid takes over
2 sharp splits with
1 transformation

Equation 4

$$2 \cdot Y_{h,c} \leq \sum_{k \in T_h} Y_k \quad \forall h \in H \ \& \ c \in C$$

Equation 5

$$\sum_{c \in C} Y_{h,c} \leq 1 \quad \forall h \in H$$
Mathematical modelling

Example:

\( t4 \) appears both in hybrids \( h1 \) and \( h4 \)

Equation 5

Each task up to 1 hybrid

\[
\sum_{h\in H_t} Y_{h,c} \leq 1 \quad \forall t \in T \quad \& \quad c \in C
\]

Objective function

\[
\min OBJ = \sum_{t\in T} E_t \cdot Y_t - \sum_{c\in C} \sum_{h\in H} S_{h,c} \cdot Y_{h,c}
\]

Tasks

Cost

Discount

Hybrids
Synthesis model – thermally integrated columns

\[
\begin{align*}
\min \quad OBJ &= \sum_{t \in T} E_t \cdot Y_t - \sum_{c \in C} \sum_{h \in H} S_{h,c} \cdot Y_{h,c} \\
\quad f_t &\leq Y_t \cdot U_t \quad \forall t \in T \\
F^\text{tot} &= \sum_{t \in T^\text{feed}} f_t \\
\sum_{t \in T} f_t \cdot J_{g,t} &= \sum_{k \in T^\text{input}_m} f_k \quad \forall g \in G \\
2 \cdot Y_{h,c} &\leq \sum_{k \in T_h} Y_k \quad \forall h \in H \, \& \, c \in C \\
\sum_{h \in H_t} Y_{h,c} &\leq 1 \quad \forall t \in T \, \& \, c \in C \\
\sum_{c \in C} Y_{h,c} &\leq 1 \quad \forall h \in H
\end{align*}
\]

Total cost

Logical constraints

Mass balance - feed

Mass balances – intermediates

2 columns yield one hybrid

1 hybrid per task

1 transformation per hybrid