

Non-Linear Programming Distillation Model for Simultaneous Process Optimisation

Keat Ping Yeoh, Chi Wai Hui*

Department of Chemical and Biological Engineering, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong SAR, China
 kehui@ust.hk

Distillation columns are prevalent process units used to purify chemicals. However, they consume significant amounts of energy for heating and cooling. A rigorous non-linear programming (NLP) distillation model where mass, equilibrium, summation and heat (MESH) equations are applied to each stage is proposed to be used in conjunction with existing Heat Integration models for simultaneous process optimisation to reduce energy consumption. In the NLP model, a continuous variable is used for each stage to determine whether a stage is active or inactive. This variable controls whether there are any changes in stream properties as a stream passes through a stage. It also enforces the equilibrium constraint for streams exiting active stages. The continuous variable mimics a binary variable as it only takes on values of 0 or 1 in an optimal solution.

The distillation model and a Heat Integration model for a multi-stream heat exchanger (MHEX) were implemented in GAMS with a case study on the separation of air consisting of N₂, O₂ and Ar. Analysis of the model performance showed that it achieved convergence in a short time (6 min for a model with 2,161 variables and 25,260 equations) even with crude initialisation methods. Results of the case study showed that the energy requirement per unit mass of oxygen product is not minimized at 100 % oxygen recovery. The average oxygen recovery was around 96 %. Optimisation of the case study on an air separation unit (ASU) reduced energy requirement by 5.4 % for some cases.

1. Introduction

Distillation columns are energy-consuming process units used to purify products. Heat integration may be the key to reducing the energy consumption of distillation systems. More energy savings may be realized if an entire distillation system, including the heat recovery unit, is optimised simultaneously in an equation-oriented (EO) environment, as opposed to using sequential modular methods. Pinch Analysis is one method of Heat Integration which may be used to maximise the potential of heat recovery in distillation systems. In Pinch Analysis, the minimum cooling and heating utility requirements are identified given a minimum temperature approach in a heat recovery system comprised of selected process streams. However, Pinch Analysis is usually applied after a process has been designed. This sequential method of Heat Integration may result in wasted potential to reduce energy consumption. Duran and Grossmann (1986) pioneered an EO method for Pinch Analysis which can be applied to simultaneous optimisation. The method involved the use of a max operator, which proved to be challenging to commercial solvers due to its non-smooth nature. Subsequently, different methods of replacing the max operator were developed, including a smoothing approximation used in the same work by Duran and Grossmann, and the use of binary variables in a mixed integer non-linear programming (MINLP) formulation as well as an NLP model using the concept of pseudo temperatures by Hui (2014).

With the development of equation-oriented Heat Integration models, it then became possible to simultaneously optimise an entire distillation system comprised of distillation columns and a heat recovery network or MHEXs. However, simultaneous optimisation of distillation columns with Heat Integration results in large model sizes with many infeasibilities, which may lead to premature termination of the solver. The modelling of distillation columns may require discrete variables to represent the number of stages when the number of stages is not fixed, leading to further difficulties in solving the model as an MINLP formulation such as the one by Farkas et al. (2008). Other works have utilized shortcut distillation models (Yeoh et al., 2019) or developed rigorous NLP

distillation models (Kraemer et al., 2009) to circumvent the need to use an MINLP formulation. However, shortcut models may not be able to accurately capture the behaviour of distillation columns. Another rigorous NLP distillation model developed by Dowling and Biegler (2015) appears to be robust and effective in conducting flowsheet optimisation with Heat Integration and complex thermodynamic methods, showing the benefits of using a rigorous NLP model to represent the distillation process when used with Heat Integration.

In this work, an alternative formulation for an NLP-based distillation model is proposed that may be used with Heat Integration methods in simultaneous process optimisation. The novel NLP model may be more robust than previous ones as it has less variables. A case study on the optimisation of an ASU consisting of two distillation columns and a MHEX was conducted to test the robustness and effectiveness of the novel NLP distillation model in flowsheet optimisation.

2. Methodology

This section provides details on the formulation of the NLP distillation model and supporting models such as the Heat Integration model. Other information, such as the thermodynamic module and the flowsheet of the case study are also mentioned here. Assumptions and where to find parameters are also included in this section.

2.1 NLP distillation model formulation

Traditionally, distillation columns are modelled as a series of stages where flash operations occur. The inlet and outlet streams and relevant properties of a typical stage in a distillation column are shown in Figure 1. Liquid streams are saturated liquid at the bubble point, while vapour streams are saturated vapour at the dew point.

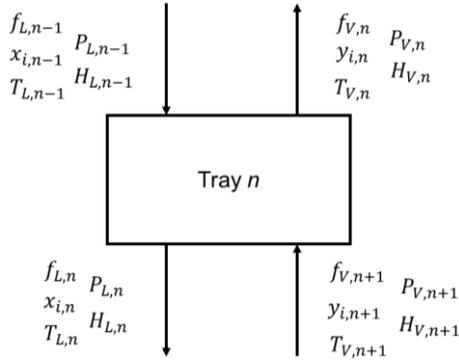


Figure 1: Representation of streams and relevant stream properties of a typical stage in a distillation column

In Figure 1, f represents the molar flowrates, x and y represent the liquid and vapour composition of component i , T and P represent the temperature and pressure of the stream, while H represents the enthalpy of the stream. L and V show whether the streams are liquid or vapour and n represents the stage where the stream exits from. An active stage is a stage where the outlet stream properties differ from those of the inlet. The liquid and vapour outlet streams of an active stage are in equilibrium with each other. Inactive stages are stages where the outlet stream properties are the same as those of the inlet. The constraint where the liquid and vapour outlet streams are in equilibrium with each other does not apply to inactive stages and needs to be relaxed. Since the number of active stages is an optimisation variable and is not known a priori, a surplus of stages will first be defined. A continuous variable is used to activate or deactivate the stages.

MESH equations such as the ones shown below are applicable to both active and inactive stages.

$$f_{L,n-1} + f_{V,n+1} = f_{L,n} + f_{V,n} \quad \forall L \in \text{Liq}; V \in \text{Vap}; n \in N \quad (1)$$

$$f_{L,n-1}x_{i,n-1} + f_{V,n+1}y_{i,n+1} = f_{L,n}x_{i,n} + f_{V,n}y_{i,n} \quad \forall L \in \text{Liq}; V \in \text{Vap}; n \in N; i \in I \quad (2)$$

$$H_{L,n-1} + H_{V,n+1} = H_{L,n} + H_{V,n} \quad \forall L \in \text{Liq}; V \in \text{Vap}; n \in N \quad (3)$$

The difference between an active and inactive stage lies in whether the flowrate, composition or temperature change. The following set of complementarity constraints are used to control whether a stage is active.

$$(1 - Z_n)(f_{L,n} - f_{L,n-1}) = \varepsilon_n \quad \forall L \in \text{Liq}; n \in N \quad (4)$$

$$(1 - Z_n)(x_{i,n} - x_{i,n-1}) = \varepsilon_n \quad \forall n \in N; i \in I \quad (5)$$

$$(1 - Z_n)(T_{L,n} - T_{L,n-1}) = \varepsilon_n \quad \forall L \in Liq; n \in N \quad (6)$$

Z_n is a continuous variable with a value between 0 and 1. It is used to activate or deactivate a stage as necessary. ε is a slack variable with a lower limit of 0. The summation of ε_n is added to the objective function to be minimized and it'll take on a value of 0 in optimal solutions. From Eq(4) to Eq(6), whenever any one of the stream properties change, Z_n must take on a value of 1 in order to force the left-hand side of the equations to be 0 to satisfy the equality constraint. In this way, stages where $Z_n = 1$ are active stages where the stream properties change. Conversely, whenever the stream properties do not change, Z_n could take on any value, as the left-hand side of the equation will always be equal to 0. However, as the following constraints also apply, it is not physically possible for Z_n to take on a value of 1 for an inactive stage.

This formulation where zero-flowrates do not occur circumvents a problem highlighted in Grossmann et al. (2005), where MESH equations are reduced when inlet and outlet flows are non-existent, leading to convergence failure. Addition of the slack variable was also done to avoid this problem, as 0 x 0 values are possible within the complementarity constraints in Eq(4) to Eq(6). Many of these characteristics to avoid singular equations are found in (Dowling and Biegler, 2015) but it can be observed that the novel model has less variables, as bypass streams do not need to be defined for each stage.

Eq(4) to Eq(6) are not required for the vapour streams, as the vapour stream properties will be determined by the MESH equations. The change in H, enthalpy, is not included because it is calculated from other properties. Change in pressure is not considered either as the outlet stream pressures will be controlled by other equations. As mentioned before, the liquid and vapour outlets are in equilibrium for an active stage but not for an inactive one. The following set of equations are used to enforce the equilibrium constraint whenever a stage is active.

$$y_{i,n} \geq K_{eq,i,n}x_{i,n} - (1 - Z_n)M \quad \forall n \in N; i \in I \quad (7)$$

$$y_{i,n} \leq K_{eq,i,n}x_{i,n} + (1 - Z_n)M \quad \forall n \in N; i \in I \quad (8)$$

$$K_{eq,i,n} = \frac{P_{sat,L,i,n}}{P_{V,n}} \quad \forall L \in Liq; V \in Vap; n \in N; i \in I \quad (9)$$

Where $K_{eq,i,n}$ is the equilibrium constant. In this work, it is calculated from the saturated pressure of the liquid and the vapour pressure following Raoult's Law in Eq(9). M is a sufficiently large scalar.

Whenever $Z_n = 1$, indicating an active stage, Eq(7) and Eq(8) are effectively equal to Eq(10).

$$y_{i,n} = K_{eq,i,n}x_{i,n} \quad \forall n \in N; i \in I \quad (10)$$

As mentioned before, it is not possible for Z_n to take on a value of 1 for an inactive stage. This is because fulfilling Eq(10) without any changes in stream properties would require the inlet liquid and vapour streams to already be in equilibrium prior to entering the stage. Such a point is known as a Pinch point, and according to Westerberg and Wahnschafft (1996), an infinite number of stages is needed to go through the Pinch Point. Since the liquid and vapour outlets of an inactive stage cannot be in equilibrium, $(1 - Z_n)$ cannot take on the value of 0, and $(1 - Z_n)$ multiplied by M is used to relax the equilibrium constraint for inactive stages.

Other than being in equilibrium with each other, another constraint where the temperature of the liquid and vapour outlet are equal applies to active stages. The constraint is implemented via Eq(11) and Eq(12).

$$T_{L,n} \geq T_{V,n} - (1 - Z_n)M \quad \forall L \in Liq; V \in Vap; n \in N \quad (11)$$

$$T_{L,n} \leq T_{V,n} + (1 - Z_n)M \quad \forall L \in Liq; V \in Vap; n \in N \quad (12)$$

Similar to the implementation of the equilibrium constraint, Eq(11) and Eq(12) ensure that the outlet temperatures are equal when Z_n is 1, and the constraint is relaxed otherwise.

The novel NLP distillation model also includes pressure drop across active stages. Pressure drop is implemented via Eq(13) and Eq(14). The pressure of liquid and vapour streams leaving an active stage are also equal. This is implemented by Eq(15) and Eq(16) for $Z_n = 1$, and relaxed when Z_n is not 1.

$$P_{L,n} = P_{L,n-1} - Z_n\Delta P \quad \forall L \in Liq; n \in N \quad (13)$$

$$P_{V,n} = P_{V,n+1} - Z_n\Delta P \quad \forall V \in Vap; n \in N \quad (14)$$

$$P_{L,n} \geq P_{V,n} - (1 - Z_n)M \quad \forall L \in Liq; V \in Vap; n \in N \quad (15)$$

$$P_{L,n} \leq P_{V,n} + (1 - Z_n)M \quad \forall L \in Liq; V \in Vap; n \in N \quad (16)$$

ΔP refers to a fixed value of pressure drop. While Z_n can take on any value except 1 for an inactive stage, the pressure drop equations would force it to take on a value of 0 whenever the objective of the optimisation is to reduce energy consumption. In an energy consumption minimization problem, the optimal value would be one without unnecessary pressure drop for an inactive stage that does not carry out a flash operation. The NLP model is able to mimic an MINLP model since Z_n only takes on binary values in an optimal solution.

2.2 Heat Integration model

The MHEX model used in the case study is modified from the NLP Heat Integration model proposed by Hui (2014). This model was selected instead of the Multi-M model found in the same work in order to solve the optimisation problem as an NLP problem instead of MINLP. The original NLP model formulation encompasses Eq(17) to Eq(29) in (Hui, 2014). As the original model was used for utility targeting in HEN, Eq(28) and Eq(29) from Hui (2014) are modified to the following equations.

$$QSOA_k(x) \geq QSIA_k(x) \quad (17)$$

$$\Omega(x) = 0 \quad (18)$$

The equations are modified in this way because there are no hot or cold utilities. Eq(18) enforces energy balance for the streams entering and exiting the MHEX.

2.3 Thermodynamic module

An ideal thermodynamic module was employed to calculate stream properties such as enthalpy and vapour-liquid equilibrium. The ideal thermodynamic module utilizes equations and parameters from the IDEAL method available in Aspen Plus V11. For example, Eq(19) is used to calculate the ideal gas specific heat capacity.

$$C_{P,i} = Co_{1,i} + Co_{2,i} \left(\frac{Co_{3,i}/T}{\sinh(Co_{3,i}/T)} \right)^2 + Co_{4,i} \left(\frac{Co_{5,i}/T}{\cosh(Co_{5,i}/T)} \right)^2 \quad \forall i \in I \quad (19)$$

Where Co_1 to Co_4 are component-specific parameters. In this work, $C_{P,i}$ is assumed to be constant and equal to $Co_{1,i}$ due to the complexity of the sinh and cosh operators in Eq(19). This simplification does not greatly affect the calculation of $C_{P,i}$, as the contributions of the second and third terms of Eq(19) are negligible compared to the first term within this study.

2.4 Details of the case study and models of the auxiliary equipment

Figure 2 shows the flowsheet used in the case study. The ASU shown here uses the double-column configuration, found to be the best in (Dowling and Biegler, 2015). Optimisation of the flowsheet is conducted at different oxygen product purities with the objective of minimizing energy consumption per kg of oxygen product.

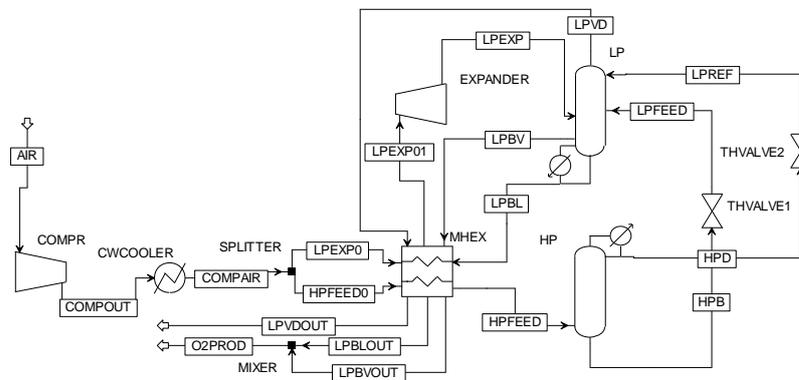


Figure 2: Flowsheet of the ASU used in the case study

Table 1 shows the specifications that were fixed in the optimisation. All feeds to the columns enter at a pressure ΔP higher than the feed stage. Results of the case study were verified in Aspen Plus.

Table 1: Specifications of the process

Equipment	Specification	Equipment/Stream	Specification
Compressor (COMPR)	Isentropic GPSA, Efficiency = 100 %	Distillation Column (HP)	No reboiler, total condenser (Cond) Feed enters at bottom-most stage Stage Pressure Drop, $\Delta P = 0.00689$ bar Condenser duty = LP Reb Duty ($\Delta T_{\min} = 1.5$ °C)
Expander	Isentropic GPSA, Efficiency = 80 %	Distillation Column (LP)	No condenser, total reboiler (Reb) LPREF stream enters at top-most stage
Cooler (CWCOOLER)	Outlet T = 30 °C Pressure Drop = 0		LPFEED and LPEXP enter on same stage Top stage pressure = 1.01325 bar Stage Pressure Drop, $\Delta P = 0.00689$ bar
MHEX	$\Delta T_{\min} = 1.5$ °C	Stream AIR	Flow: 1 kmol/h, Pressure: 1.01325 bar, T = 25 °C Mol frac: 0.78 N ₂ , 0.21 O ₂ , 0.01 Ar
Valves	Adiabatic	Stream LPEXP01	Saturated vapor at dew point

Compressor work and work produced by the expander are calculated using the following formula:

$$W = \frac{f_V \cdot R \cdot T_{in} \cdot \gamma}{\gamma - 1} \cdot \left(\left(\frac{P_{V,out}}{P_{V,in}} \right)^{\frac{\gamma-1}{\gamma}} - 1 \right) \quad \forall V \in Vap \quad (20)$$

Where γ is the ratio of specific heat capacities, $\frac{c_p}{c_v}$, R is the ideal gas constant and η is the efficiency. W is multiplied by η for the expander and divided by η for the compressor. Expander work will be negative.

The flowsheet optimisation is shown below, with the objective function to minimize energy consumption per unit mass of O₂ product. MW is the molecular weight while f(x) and g(x) are the equality and inequality constraints.

$$\min Obj = \frac{W_{COMPR} + W_{EXPANDER}}{\sum_i f_{O2PROD} y_{i,O2PROD} MW_i} + \sum_n \varepsilon_n \quad \text{s.t.} \quad f(x) = 0, g(x) \leq 0 \quad (21)$$

3. Results and discussion

The performance of the novel NLP distillation model will be discussed with regard to its speed and optimisation results. Process parameters and objective values from the case study will also be shown and analysed.

3.1 Performance of the novel NLP distillation model

The case study was implemented in GAMS 24.7 and solved as an NLP model using CONOPT. As predicted, values of Z_n were either 0 or 1 when the final optimisation was performed with the pressure drop constraints. Table 2 shows a comparison of model attributes and solving time for different numbers of available stages.

Table 2: Model attributes and solving time of the NLP model

O2 purity (mol %)	Available stages	Active stages	Variables	Equations	Time elapsed (min)
94	35	22	2,161	25,260	6
97	100	34	5,466	154,507	20

The NLP model generates solutions quickly as seen in Table 2, which makes it suited for multi-start initialisation to obtain multiple local optimal solutions or to provide good initial values and bounds to a global solver.

Initialisation of stream properties was done by fixing parameters such as the reflux ratio and feed temperature of the HP column, before solving the model by minimizing the sum of the slack variable. These fixed values were subsequently allowed to vary. Constraints such as condenser and reboiler coupling were added prior to optimising the system with the objective function in Eq(21) without pressure drop before the final optimisation was performed with pressure drop considered. Results from the system without pressure drop tends to activate excess stages. When pressure drop is considered, the NLP model reduced the number of stages, showing that it can optimise the number of stages to an extent. However, the solutions presented here were achieved by optimising the system again with addition or removal of stages to compare the values of the objective function. Better solutions were found but the values only differed by around 1 %. The model may benefit from using aggregate distillation models with pressure drop considerations for initialisation, as pressure drop has an impact on the solution. Even with the crude initialisation procedure used here where the final values differed greatly from the initial values, the NLP model converged most of the time.

3.2 Results of the Case Study

Table 3 shows the optimised process parameters and objective values of the case studies at different O₂ product purities. The molar flowrates of LPBL, LPBV and LPVD add up to the molar flowrate of the AIR stream.

Table 3: Process parameters and results of the case study

O ₂ PROD	Obj	O ₂ Recovery	Stages			HPFEED		HPD	Cond	LPEXP	LPBL	LPBV	
mol %	kJ kg ⁻¹	%	HP	LP	St. No.	f	T	P	f	P	P	f	f
			No.	No.	LPFeed	kmol h ⁻¹	°C	bar	kmol h ⁻¹	bar	bar	kmol h ⁻¹	
94	602	95.3	7	15	8	0.883	-178	4.13	0.370	4.08	1.07	0.213	0
95	619	96.7	8	16	7	0.786	-176	4.40	0.408	4.34	1.06	0	0.214
96	633	96.9	10	18	8	0.787	-176	4.46	0.414	4.39	1.07	0	0.212
97	663	97.0	12	22	13	0.836	-176	4.61	0.437	4.52	1.10	0	0.210
98	713	93.8	12	26	16	0.943	-175	4.66	0.515	4.58	1.12	0	0.201

In certain cases, the optimisation procedure reduced the energy consumption by 5.4 % from an initially feasible solution (e.g. 754) to the final value (e.g. 713), showing that the model can identify potential energy savings. A transition in a process parameter is seen in Table 3, where liquid product is preferred at 94 mol % purity, but vapour is preferred above 95 mol %. Higher reboiler and condenser duty may be needed to achieve higher O₂ and N₂ purities, leading to more vapour in the LP column. At lower purities, drawing liquid may be preferred as the temperature of liquid is lower than vapour of the same O₂ purity. A lower HP condenser pressure is then needed to meet the ΔT_{\min} , since the HP distillate temperature needed to heat the reboiler is lower. The minimum energy requirement does not occur at maximum number of stages or 100 % O₂ recovery, possibly because more stages and higher recovery lead to higher pressure drop throughout the columns and higher compressor power. The O₂ recovery decreases for 98 % purity, which may be due to limitations of the ASU configuration presented, as the ΔT in the MHEX is at the minimum for all streams for this purity.

4. Conclusions

A novel NLP distillation formulation was presented and implemented with a case study on an ASU. The NLP formulation was found to be fast with good convergence qualities when combined with Heat Integration models, even with crude initialisation methods. However, it tended to get stuck at local optimums and may benefit from better initialisation methods. Results of the case study found that 5.4 % energy savings are attainable compared to a feasible initial point. It was also intriguing that the minimum energy requirement and maximum product recovery do not coincide, with an average optimal product recovery of around 96 % for the proposed system.

Acknowledgements

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