

RIGOROUS METHOD OF MINIMUM ENERGY CALCULATION FOR A FULLY THERMALLY COUPLED DISTILLATION SYSTEM

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Previously, Tanskanen and Pohjola (2000) and Tanskanen and Malinen (2005) have presented a method of rigorous minimum energy calculation for non-ideal multicomponent distillation. The method is based on column simulation with a large number of equilibrium stages to mimic infinitely high columns, and it has been successfully tested with simple distillation column systems. This paper describes a research work where the rigorous minimum energy calculation method is extended to a fully thermally coupled distillation column system, also called Petlyuk column. In the solving procedure, a bounded Newton homotopy method is used to detect the optimum point of operation for the column system, which is described with the full set of MESH equations. There are no restrictions for the thermodynamic models used. The results are promising and indicate that the numerical challenges encountered when rigorously solving complex distillation problems can be tackled by utilising modern numerical solving approach.

KEYWORDS: minimum energy calculation, minimum reflux, thermally coupled distillation systems, bounded homotopy continuation methods, optimisation

INTRODUCTION

The energy demand of a distillation system is an increasingly important process performance factor due to the growing interest in saving energy. Detection of the minimum energy requirements of different distillation systems can offer an early assessment tool for comparing structural alternatives. Knowledge about the steady-state minimum energy operation conditions also helps to operate real column systems towards lower energy consumption.

Complex distillation column sequences, which have thermal couplings between the columns, can reduce the energy demand of a separation task by up to 30% compared to the traditional simple distillation column arrangements. The capital costs of complex columns may also be significantly lower than those of traditional arrangements for the same separation task. Therefore, much research has recently also focused on these integrated process systems (e.g. Caballero and Grossmann (2004), Fidkowski and Agrawal (2001), Halvorsen (2001) and Jiménez *et al.* (2003)).

Minimum reflux detection for complex column sequences is unnecessarily often based on the constant molar overflow and constant relative volatility assumptions (e.g. Fidkowski and Krolikowski (1986), Halvorsen and Skogestad (2003)). Column systems in which

the complex main column is represented as two simple columns have also been used as a simplification (e.g. Triantafyllou and Smith (1992) and Annakou and Mizsey (1996)). The dynamic model has also been used as a method to detect the minimum energy operation point (e.g. Halvorsen and Skogestad (1997), Hernández and Jiménez (1999)).

The use of short-cut methods has been defended based on the argument of the complexity of the problem and the need to rapidly assess alternative designs, perform preliminary optimisation and provide the initialisation for rigorous simulation (Fidkowski and Krolikowski (1986)). Although the aim of short-cut methods is merely to approximate the minimum energy requirement, the non-ideality of real mixtures requires a more exact and realistic approach to reach the requisite accuracy.

Calculation of the minimum reflux of simple columns has advanced to a stage where mass and energy balances together with state-of-the-art thermodynamics are exploited. Tanskanen and Pohjola (2000) and Tanskanen and Malinen (2005) have presented a rigorous minimum energy calculation method, which is based on column simulation with a large number of equilibrium stages to mimic infinitely high columns. During the solving process, manual iterations have been avoided by specifying directly the key components and applying problem-dependent homotopy to solve the respective MESH equations. The method has been successfully tested with a simple distillation column (one-feed, two-product), and it is applicable to non-ideal multicomponent distillation.

In this paper, the rigorous minimum energy calculation method is extended to a fully thermally coupled distillation column system. In the solving process, a problem-independent, bounded Newton homotopy continuation method is used to solve the process model consisting of the full set of MESH equations with a non-ideal thermodynamic model.

PROBLEM DEFINITION

Based on Christiansen et al. (1997), the fully thermally coupled distillation column system (shown in Figure 1), also called the Petlyuk column, is “*a column arrangement separating three or more components using a single reboiler and a single condenser, in which any degree of separation (purity) can be obtained by increasing the number of stages (provided the reflux is above a certain minimum value).*” This definition gives a good basis for explaining the procedure introduced herein when defining rigorously the minimum energy demand of a fully thermally coupled distillation column system.

After specifying the feed, the number of stages and the feed and side draw stages, the total degrees of freedom of the Petlyuk system is five. If three degrees of freedom are used by specifying mole fractions of the key components in the product flows (i.e. one specified mole fraction in each product flow), there are still two degrees of freedom left. This is the main difference between the simple column and complex fully thermally coupled column system. In the case of the simple column, there are only two degrees of freedom, by which the separation can be fully specified. In the case of Petlyuk, the two extra degrees of freedom left can be utilised in the detection of the minimum energy operation point for the column system.

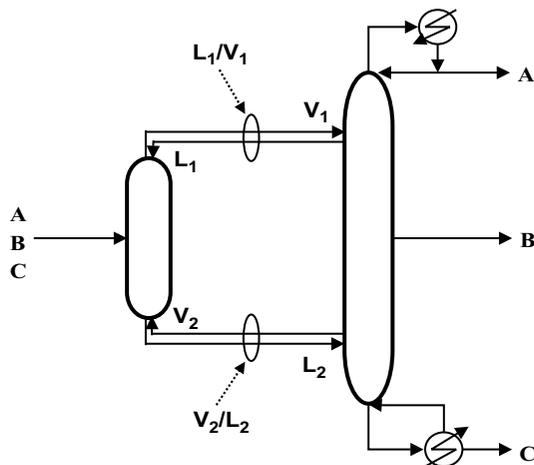


Figure 1. Petlyuk column with thermal couplings between the prefractionator and the main column

Because the aim is to detect the rigorous minimum energy demand for pre-specified separation, a sufficiently large number of stages is needed to mimic infinitely high columns. The sufficiently large number of column stages can be understood as an assurance for pinch points to form into every column section. The formation of pinch points indicates that, within the feed in question, the number of column stages is sufficient. This guarantees that the minimum energy condition of the simulated column sequence corresponds to the minimum energy condition of the specific column sequence having an infinite number of stages. Now, based on the definition mentioned above, the pre-specified separation target (product flow purities) can be achieved provided that the reflux is above a certain minimum value.

Especially when specifying systems separating azeotropic mixtures care must be put on how to specify the system in a reasonable way. The specifications should not break the possible natural restriction of the separation in the form of azeotropes and distillation boundaries. Residue curve maps have been found to be a useful tool when considering proper specifications for the product flows.

SOLVING PROCEDURE

Because of the strong interactions between the fully thermally coupled columns, the procedure is based on the equation-oriented solving method to rigorously detect the minimum energy operation point for the column system. The solution is possible without any simplifications of the process or the thermodynamic models.

The column model consisting of the full set of MESH equations has been implemented in the high-level programming language MATLAB. Thus, all phases in the equation-oriented solving approach are fully transparent, and the procedure can be developed independently of the restrictions set by the modular approach used in many commercial simulation packages. The column system specifications are made and the results saved within Microsoft Excel.

The target of the procedure is to detect the minimum energy operation conditions for the column system, not to optimise the system's structure or to minimise the capital costs. This is why the column sequence structure (i.e. the number of stages, the feed and side draw stage locations and the reboiler and condenser types) is fixed together with the feed conditions (i.e. feed composition, feed amount, pressure and temperature).

The rigorous minimum energy determination of a fully thermally coupled distillation column sequence can be carried out in three phases. All the phases are implemented and coded in such a way that no manual intervention between the phases is needed. Thus, the whole solving procedure can be carried out at the "touch of a button."

The three phases are:

1. Solve the initial profiles for the column sequence using the main column's reflux and boilup ratio values (e.g. values of one) and the proper liquid/vapour (L_1/V_1) and vapour/liquid (V_2/L_2) molar flow ratios between thermally coupled columns (e.g. 0.1). The liquid side draw flow is specified to be some small portion (e.g. ten percent) of the molar liquid flow down from the side draw stage.
2. Solve the column sequence with exact mole fraction specifications for product flows (one mole fraction specification for each product flows) by using the initial column profile obtained in the first phase. Specify the L_1/V_1 and V_2/L_2 molar flow ratios between the thermally coupled columns to fulfil the separation target. The L_1/V_1 and V_2/L_2 values of 0.2 for zeotropic and 0.5 for azeotropic mixtures have been found to be good for the simulated cases.
3. Detect the minimum energy operation point, i.e. the minimum reboiler duty, based on the optimisation of the L_1/V_1 and V_2/L_2 values. Use the solution obtained in the second phase as a starting point for minimisation.

Optimisation is based on the minimisation of the reboiler duty instead of the boilup rate or the boilup ratio. In the optimisation process, the simple strategy of steepest descent has been found to be adequate. The Jacobian matrix is approximated numerically based on the method of finite differences. Problem sparsity has been taken into account, and thus the number of function evaluations has been reduced substantially.

In the case of highly non-ideal systems, local Newton-Raphson based solving methods cannot guarantee converged result and thus, a significant amount of manual iteration work might be needed during the solving process. To avoid this weakness, the homotopy continuation methods are utilised to improve the robustness of the solving process. Although the homotopy continuation methods have the property of global convergence, problems may appear. In the case of problem-independent homotopy, the homotopy

path may run outside the domain, i.e. the intermediate solutions of the homotopy path may have variable values that are not defined. This may cause difficulties, especially when solving chemical engineering problems. For example, if negative mole fraction values are substituted into the subroutines computing enthalpy and phase equilibrium values, error stops may be generated and the homotopy path tracking process interrupted.

Perhaps the most promising suggestion to overcome the problem of unboundedness of the homotopy path is the class of bounded homotopy methods proposed by Paloschi (1995, 1997). Bounded homotopy methods have several advantages concerning the possibility to track the homotopy path in real space without running outside the domain of interest. Nonetheless, problems may appear if the variables, such as mole fractions in distillation calculation, come very close to the domain boundary. This means that the bounding zone, in which the bounded homotopy method will bound the homotopy path, must be narrow. This, however, will introduce difficulties in the form of challenging the path-tracking task.

To improve the bounded homotopy method's ability to track the homotopy path very near the problem domain boundaries, the method of variables mapping developed by the authors (Malinen and Tanskanen (2006)) is adopted into use here. To make the mapping as effective and easy as possible to carry out, the maximum (b_i^{\max}) and minimum (b_i^{\min}) values of the variables are specified. These values can be realized as the domain boundary values, i.e. the real physical restrictions or some artificial values. The mapping equations used in bounded homotopy path tracking are formulated as follows:

Variable x_i mapping from the finite space into the infinite space:

$$x_i^{\text{inf}} = \log_{10} \left(\frac{2 \cdot (x_i - b_i^{\min})}{b_i^{\max} - b_i^{\min}} \right), \quad \text{when } x_i < 0.5 \cdot (b_i^{\max} + b_i^{\min}) \quad (1)$$

$$x_i^{\text{inf}} = \log_{10} \left(\frac{0.5 \cdot (b_i^{\max} - b_i^{\min})}{b_i^{\max} - x_i} \right), \quad \text{when } x_i \geq 0.5 \cdot (b_i^{\max} + b_i^{\min}). \quad (2)$$

Respectively, variable x_i^{inf} mapping from the infinite space into the finite space:

$$x_i = b_i^{\min} + 0.5 \cdot (b_i^{\max} - b_i^{\min}) \cdot 10^{x_i^{\text{inf}}}, \quad \text{when } x_i^{\text{inf}} < 0 \quad (3)$$

$$x_i = b_i^{\max} - 0.5 \cdot \frac{(b_i^{\max} - b_i^{\min})}{10^{x_i^{\text{inf}}}}, \quad \text{when } x_i^{\text{inf}} \geq 0. \quad (4)$$

The bounded Newton homotopy method is used in every phase of the rigorous minimum energy detection procedure. Even though the solving process based on homotopy continuation takes much computing time, it provides the means to obtain the converged solution, i.e. the minimum energy operation point, in a robust way.

RESULTS

Tables 1 and 2 show the results representing the capability of the procedure to determine the minimum energy operation point for ternary zeotropic and azeotropic mixtures. The thermodynamic models and parameters used in the calculation are based on the Wilson thermodynamic package and on the database of the commercial simulation program HYSYS.

Table 1. Minimum energy operation point for various acetone/benzene/toluene distillations

	Feed composition: liquid, mole fraction	Product flow compositions: distillate (mole frac) side draw (mole frac) bottom (mole frac)	Optimised variables: L_1/V_1 V_2/L_2	Reflux ratio Reboil ratio	Reboiler duty Condenser duty (kJ/kmole feed)
1	0.3500/0.4000/0.2500	0.9000 /0.1000/0.0000	0.257	1.5107	30,364
		0.0000/ 0.9000 /0.1000	0.577	3.8435	28,719
		0.0000/0.1000/ 0.9000			
2	0.3500/0.4000/0.2500	0.9500 /0.0500/0.0000	0.272	1.9511	33,601
		0.0000/ 0.9500 /0.0500	0.615	4.1446	31,836
		0.0000/0.0500/ 0.9500			
3	0.3500/0.4000/0.2500	0.9900 /0.0100/0.0000	0.295	2.3442	36,365
		0.0000/ 0.9900 /0.0100	0.655	4.3989	34,495
		0.0000/0.0100/ 0.9900			
4	0.1500/0.7000/0.1500	0.9500 /0.0500/0.0000	0.189	5.7466	32,362
		0.0000/ 0.9500 /0.0500	0.469	8.0815	31,192
		0.0000/0.0500/ 0.9500			
5	0.1500/0.7000/0.1500	0.9900 /0.0100/0.0000	0.307	7.9776	40,963
		0.0000/ 0.9900 /0.0100	0.482	8.5273	39,688
		0.0000/0.0100/ 0.9900			
6	0.4000/0.2000/0.4000	0.9500 /0.0500/0.0000	0.376	1.4817	32,218
		0.0500/ 0.9500 /0.0000	0.542	2.2904	29,960
		0.0000/0.0500/ 0.9500			
7	0.1500/0.4000/0.4500	0.9500 /0.0500/0.0000	0.373	5.5525	32,340
		0.0000/ 0.9500 /0.0500	0.448	2.1359	30,295
		0.0000/0.0500/ 0.9500			
8	0.2000/0.6000/0.2000	0.9500 /0.0500/0.0000	0.193	4.3155	34,224
		0.0000/ 0.9500 /0.0500	0.623	5.7441	32,767
		0.0000/0.0500/ 0.9500			

60 stages in the prefractionator, 138 stages in the main column, total condenser and partial reboiler.
Pressure: 101.3 kPa; boiling liquid feed.

Table 2. Minimum energy operation point for various acetone/chloroform/benzene distillations

	Feed composition: liquid, mole fraction	Product flow compositions:		Optimised variables: L_1/V_1 V_2/L_2	Reflux ratio Reboil ratio	Reboiler duty Condenser duty (kJ/kmole feed)
		distillate (mole frac)	side draw (mole frac) bottom (mole frac)			
9 a	0.1500/0.7000/0.1500	0.0500/ 0.9500 /0.0000		0.736	21.3105	298,393
		0.3300 /0.6699/0.0001		0.776	61.4217	298,336
		0.0002/0.0498/ 0.9500				
10 a	0.1500/0.7000/0.1500	0.0100/ 0.9900 /0.0000		0.736	25.7367	318,188
		0.3300 /0.6699/0.0001		0.776	68.1255	318,140
		0.0000/0.0100/ 0.9900				
11 a	0.1000/0.8000/0.1000	0.0100/ 0.9900 /0.0000		0.668	11.2196	219,965
		0.3300 /0.6699/0.0001		0.821	70.6416	219,952
		0.0000/0.0100/ 0.9900				
12 a	0.1000/0.7000/0.2000	0.0100/ 0.9900 /0.0000		0.703	25.5558	397,063
		0.3300 /0.6699/0.0001		0.769	63.7506	396,989
		0.0000/0.0100/ 0.9900				
13 b	0.2000/0.4000/0.4000	0.9500 /0.0500/0.0000		0.620	17.3271	52,315
		0.1857/ 0.6550 /0.1593		0.652	5.2467	52,133
		0.0000/0.0500/ 0.9500				
14 b	0.3500/0.4000/0.2500	0.9000 /0.0998/0.0002		0.526	6.0883	49,835
		0.2221/ 0.6000 /0.1779		0.703	10.2025	49,788
		0.0008/0.0992/ 0.9000				

a: 200 stages in the prefractionator, 138 stages in the main column, total condenser and partial reboiler.

b: 200 stages in the prefractionator, 238 stages in the main column, total condenser and partial reboiler.

Pressure: 101.3 kPa; boiling liquid feed.

Depending on the simulation case, the column system structure (i.e. the number of stages in the columns, and the location of the feed and side draw stages) is modified to guarantee pinch points to form into every section of the column system. The feed is assumed to be a bubble point ternary mixture at a pressure of 101.3 kPa. Three mole fraction specifications for the product flows are represented as bolded in the tables. Reboiler and condenser duties and the final values of the optimisation variables together with the main column reflux and boilup ratios are shown as the representative results of the energy usage minimisation procedure. The example cases have been solved with a 3 GHz personal computer.

In Figures 2a and 2b, the liquid mole fraction profiles are depicted at the minimum energy operation point for the three simulated cases. It can be seen that the profiles have

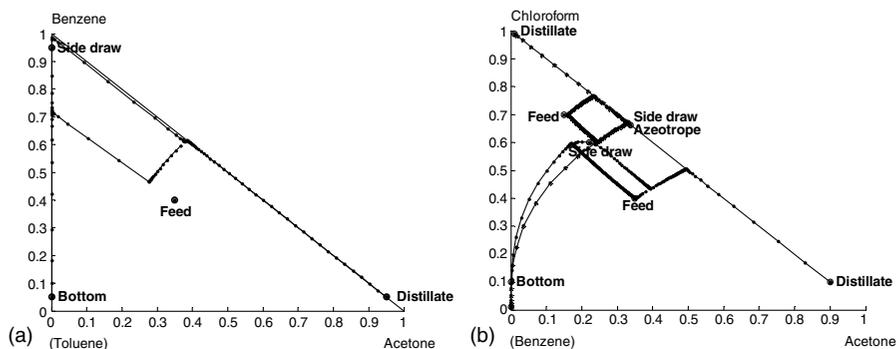


Figure 2. Liquid mole fraction profiles at the minimum energy operation point (a) in the case 2 of Table 1, and (b) in the cases 10 and 14 of Table 2

pinch points in every column section. This indicates that the simulated column systems have a sufficiently large number of stages to approximate the energy demand of an infinitely high column system. Figure 2b shows also that when separating azeotropic mixtures with minimum energy condition, composition profile of the Petlyck column can exceed the distillation boundary from the concave side of the boundary. From the convex side of the boundary the exceeding is not possible.

Results show that thermodynamic character of the mixture has remarkable effect on the easiness of the separation. The L_1/V_1 and V_2/L_2 ratios between thermally coupled columns, and also the reflux and boilup ratios are highly dependent on the feed composition and the target of separation. The higher purity requirements for product flows have been set, the higher reflux flows and flows between the columns are required. Especially when separating azeotropic mixtures, remarkable high L_1/V_1 , V_2/L_2 and reflux values may be needed to approach the separation target. Also the total number of stages of the column system may become very high when detecting the minimum energy operation point for azeotropic mixture.

The authors want to underline that, even though computing times are quite excessive, savings in total solving time can still be considerable compared with the manual iteration method. In addition, most of the time is spent on the troublesome minimisation phase. This inefficiency is partly due to the poor starting point selection. Because in general it is difficult to specify the favourable L_1/V_1 and V_2/L_2 values beforehand, the values 0.2 for zeotropic and 0.5 for azeotropic distillations were selected here.

The components' low mole fraction values due to the high number of column stages used in the simulated cases also introduced challenges and slowed down the homotopy path tracking process. Even though the variables mapping used here to track the homotopy path brought significant advantages into path tracking, the homotopy path tracking near the domain boundary and inside the extremely narrow bounding zone has still not

been completely solved. It may be that the numerical accuracy will finally pose the definite minimum value for the bounding zone width that cannot be certainly bypassed.

CONCLUSIONS

The rigorous minimum energy calculation procedure introduced in this paper was found to be robust, and the minimum energy operation point for zeotropic and azeotropic mixtures could be approached without manual intervention during the detection process. By taking local Newton-Raphson based methods into use in routine phases, such as initial profile and optimisation calculations, the computing time could be notably shortened.

Even though the detection algorithm was not optimised, the results were very encouraging, indicating that the numerical challenges encountered when solving complex distillation column problems with exact mole fraction specifications can be tackled by using advanced non-linear equation set solving methods. A solution can also be achieved without a need to simplify either the process model or the thermodynamic model.

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