

Solving distillation problems by terrain methods

Angelo Lucia*, Feng Yang

Department of Chemical Engineering, University of Rhode Island, Kingston, 16 Greenhouse Road, Kingston, RI 02881-0805, USA

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Abstract

This paper clearly shows that the recently proposed terrain methodology of Lucia and Yang (2003) can be used to solve steady-state distillation problems in a reliable and efficient manner. Numerical results are presented that show that terrain methods are superior to Newton's method and homotopy-continuation for distillation examples with multiple solutions.

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1. Introduction

Distillation has always been a workhorse of physical separation in the process industry and probably will remain so for the foreseeable future. Thus, methods for solving mathematical models of distillation operations have been of widespread interest for many years. There are literally hundreds of papers on solving distillation problems. See, for example, Wang and Henke (1966), Goldstein and Stanfield (1970), Naphtali and Sandholm (1971), Ishio and Otto (1973), Shewchuk (1974), Kubicek, Hlavacek, and Prochaska (1976), Block and Hegner (1976) and Hess, Gallun, Bentzen, Holland, McDaniel, and Telow (1977). Also see Magnussen, Michelsen, and Fredunslund (1979), Buzzi-Ferraris and Morbidelli (1981), Wayburn and Seader (1983), Prokopakis and Seider (1983), Byrne and Baird (1985), Vickery and Taylor (1986), Kingsley (1986), Venkataraman and Lucia (1988), Widagdo, Seider, and Sebastian (1989), Kooijman and Taylor (1992), Taylor, Achuthan, and Lucia (1996). Obviously this list is by no means complete. One of the more interesting subclasses of problems in the distillation modeling area is the class that exhibits multiple two-phase solutions for a given set of column specifications. See, Shewchuk (1974), Magnussen et al. (1979), Wayburn

and Seader (1983), Prokopakis and Seider (1983), Byrne and Baird (1985), Kingsley (1986), Venkataraman and Lucia (1988), Taylor et al. (1996). These problems typically have three solutions whose parameterized bifurcation diagrams are usually S-shaped. As a result, multiple starting points are required if local equation-solving methods (e.g., Newton's method) are used and more than one solution is desired. However, even from so-called 'good' initializations (e.g., Venkataraman and Lucia, 1988) local methods can have difficulty finding at least one of the solutions. On the other hand, global methods like homotopy-continuation can require large numbers of function evaluations to find all solutions of interest or even fail to negotiate sharp turning points in the bifurcation diagram. The primary objective for this paper is to demonstrate to the reader that the recently proposed terrain methodology of Lucia and Yang (2002, 2003) can be used without modification to solve large-scale problems like distillation in a reliable and efficient manner.

2. Mathematical model

One set of equations for the j th stage of any two-phase, equilibrium-staged distillation consists of the component material balances

$$l_{j-1,i} - l_{ji} - v_{ji} + v_{j+1,i} + f_{ji} = 0, \quad i = 1, 2, \dots, n_c \quad (1)$$

* Corresponding author. Tel.: +1 401 874 2814; fax: +1 401 874 4689.
E-mail address: lucia@egr.uri.edu (A. Lucia).

equilibrium equations

$$\frac{K_{ji}l_{ji}}{\left(\sum l_{jk}\right)} - \frac{v_{ji}}{\left(\sum v_{jk}\right)} = 0, \quad i = 1, 2, \dots, n_c \quad (2)$$

energy balance equation

$$\left(\sum l_{j-1,i}\right) H_{j-1} - \left(\sum l_{ji}\right) H_j - \left(\sum v_{ji}\right) h_j + \left(\sum v_{j+1,i}\right) h_{j+1} + \left(\sum f_{ji}\right) H_j^F + Q_j = 0 \quad (3)$$

where f_{ji} , l_{ji} and v_{ji} are the feed, liquid and vapor molar flows for the i th component for the j th stage, K_{ji} is the equilibrium ratio or K -value for the i th component on the j th stage, H_j and h_j denote liquid and vapor enthalpy respectively and Q_j is the heat duty to the j th stage. Because we treat the heat duty at any stage as a variable, we also add specification equations for the heat duties to the equation set for all stages except the top and bottom stages. Two additional specifications are also required to fix the column operation. Thus there are $n_s(2n_c + 2)$ equations and unknown variables for the distillation model, where n_c denotes the number of components in the mixture and n_s is the number of stages in the column.

3. Terrain methodology

The main ideas of terrain following are based on the fact that neighboring stationary points (i.e., solutions and saddle points) are connected along valleys. These valleys, say \mathcal{V} , are defined by

$$= \{\min g^T g \text{ such that } F^T F = L \text{ for all } L \in \mathcal{L}\} \quad (4)$$

where F is a vector function in the unknown variables Z , $g = J^T F$ is the gradient of the least squares function, $F^T F$, J is the Jacobian matrix of F , L is a given level or contour of the least squares function and \mathcal{L} is a collection of levels. Using a reliable local equation-solving method to find a first stationary point, terrain following consists of intelligently moving up and down the valleys of the least-squares landscape to find other solutions or saddle points. Downhill movement is based on equation solving while uphill movement uses predictor-corrector calculations to locate singular points. Predictor steps are uphill Newton or Newton-like steps because they tend to flow along valleys but do drift some. Corrector steps, on the other hand, are only used intermittently to force iterates back to the current valley by solving the nonlinearly constrained optimization problem defined in Eq. 4. See Lucia and Yang (2002, 2003) for details.

Example 1 (A simple distillation). To illustrate the difficulties in finding some distillation solutions, consider the two-stage atmospheric distillation of a mixture of 45.3 kmol/h of acetonitrile, 12.72 kmol/h of water and 15.56 kmol/h of acrylonitrile proposed by Lucia, Guo, Richey, and Derebail (1990). Here, as in the other distillation example described in this paper, the liquid and vapor phases were modeled by the

Table 1
Solutions for an acetonitrile–water–acrylonitrile distillation at 1 atm

Stage	l (kmol/h) ^a	v (kmol/h)	T (K)	Q (kcal/mol)
1	13.439	4.3706	345.905	-2.15388×10^6
	23.301	10.1653		
	27.760	15.4641		
2	0.1594	17.8101	347.752	3.13983×10^6
	2.5547	33.4662		
	0.0959	43.2237		
1	9.8817	4.4617	345.603	-2.39671×10^6
	40.2163	10.0090		
	14.4020	15.5293		
2	0.0683	14.3434	356.983	3.38187×10^6
	2.7109	50.2252		
	0.0307	29.9313		
1	3.0192	4.5050	346.102	-2.6728×10^6
	58.9581	9.9473		
	2.5226	15.5477		
2	0.0250	7.5243	364.752	3.6592×10^6
	2.7727	68.9054		
	0.0123	18.0704		

^a Component molar flows in the order – acetonitrile, water, acrylonitrile.

UNIQUAC and Hayden-O'Connell equations, respectively (see, Prausnitz, Anderson, Grens, Eckert, & Hsieh, 1980 for the model equations and properties data) and no scaling of equations or variables was used. This seemingly simple problem has three two-phase distillation solutions for a range of distillate flow rates as well as three-phase behavior on one or both trays. An example of the multiple two-phase solutions is given in Table 1 for a fixed distillate rate of 30 kmol/h and fixed vapor boil-up of 94.5 kmol/h. Fig. 1, on the other hand, gives the complete bifurcation diagram for the two-phase solutions where water purity in the bottom product is plotted as a function of distillate flow rate. Note that the upper turning point in Fig. 1 (i.e., the one for which the water purity is

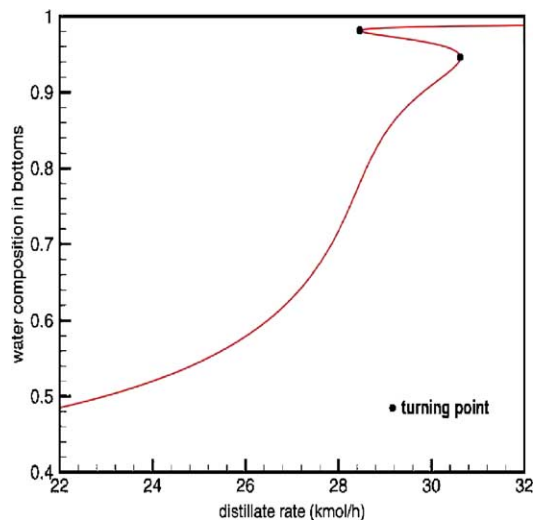


Fig. 1. Parameterized two-phase solutions to a simple distillation.

approximately 0.99) is quite sharp and potentially difficult to navigate.

4. Newton's method

Consider the case where the two specifications for the column are a distillate flow rate of 30 kmol/h and vapor boil-up of 94.5 kmol/h. If we use the ideal solution starting point strategy described in Venkataraman and Lucia (1988), direct prediction Newton's method finds the intermediate purity water (or second) solution shown in Table 1 in 13 iterations (or function evaluations). Six iterations are required to generate the ideal solution starting point and seven iterations are needed to find the intermediate solution. Convergence to any solution was assumed when $\|F\| \leq 10^{-8}$, where F is the collection of model equations. If, on the other hand, we use the nonideal starting point strategy given in Venkataraman and Lucia, Newton's method finds the low purity water (or first) solution given in Table 1 again in 13 iterations. Somewhat to our surprise it is the third solution, the high purity water solution, that is the most 'difficult' two-phase solution to find in this example! To find the high purity water solution a very careful initialization is required and this is consistent with the remarks made by many others (e.g., Magnussen et al., 1979; Prokopakis and Seider, 1983).

5. Continuation

We also used Newton's method with homotopy-continuation and stepsize adjustment to follow the bifurcation diagram in Fig. 1 to find the solutions to this problem for the exact same set of specifications. However, this was computationally expensive. Recall that it takes 13 function evaluations to find the low purity solution using the nonideal starting point in Venkataraman and Lucia (1988). From this low purity solution, an additional 756 function evaluations were required to find the remaining two solutions using an initial stepsize in the parameter of $\Delta D = 0.05$ kmol/h. However, many smaller steps in the distillate flow rate parameter were needed to negotiate the turning points at $D = 28.468$ kmol/h and $D = 30.623$ kmol/h and locate the intermediate and high purity solutions. Continuation from the intermediate solution gives essentially the same results.

6. Terrain-following

We used terrain following with analytical first and second derivatives to solve the exact same problem and had no difficulties whatsoever finding all three solutions (and the two saddle points between them) in 96 function evaluations. In particular, we used the nonideal starting point described in Venkataraman and Lucia (1988) to first find the low purity solution in 13 iterations. This low purity solution has a bottoms water composition of 0.90914. Using an initial perturbation

in the direction of the eigenvector associated with the smallest eigenvalue of $J^T J$ (i.e., $\lambda = 8.114 \times 10^{-13}$), it took our terrain algorithm 21 function evaluations to follow the valley uphill to a saddle point with a bottoms water composition of 0.94787. From here, perturbation in the eigen-direction associated with the negative eigenvalue ($\lambda = -8.5700 \times 10^{-7}$) at the saddle in a direction away from the low purity solution led downhill to the intermediate purity solution with a bottoms water composition of 0.96477 in 18 iterations. Uphill movement from this second solution was again initiated in the eigen-direction associated with the smallest positive eigenvalue of $J^T J$ (i.e., $\lambda = 9.2652 \times 10^{-13}$) in a direction away from the (first) saddle point. This eventually led to a second saddle point with a water composition of the bottom stream of 0.98053 in 30 function evaluations. Finally, downhill eigen-perturbation from the second saddle in the direction associated with the negative eigenvalue ($\lambda = -3.8448 \times 10^{-7}$) and away from the intermediate purity solution resulted in the downhill computation of the high purity solution with a bottoms water composition of 0.98672 in 15 iterations.

Note that our terrain-following methodology easily tracks the valley that connects the three solutions and two saddle points in this example, which turns out to be essentially a straight line embedded on a curved surface. We know the valley is straight because the principal eigenvector defining the valley changes very little over the course of the calculations. In fact, the principal eigenvector turns out to be dominated by the condenser and reboiler duties. Because the valley is essentially straight, uphill Newton steps track the valley sufficiently well and no corrector iterations (or intermittent solutions to Eq. (4)) are required to return iterates to the valley. Finally we use inverse power iteration to find only some of the smallest positive or largest negative eigenvalues of the Hessian matrix of $F^T F$ at any stationary point (and the associated eigenvectors) but do not actually form matrices like $J^T J$ to avoid numerical difficulties caused by truncation.

Example 2 (*The ethanol–benzene–water column*). The standard configuration for this column can be found in several literature articles (Magnussen et al., 1979; Prokopakis and Seider, 1983) and consists of 42 equilibrium stages including the reboiler and partial condenser, two feed streams, and standard specifications of reflux ratio and bottoms flow rate. The objective of this column is to produce high purity (or denatured) ethanol in the bottom of the column by adding benzene entrainer to form a minimum boiling ternary azeotrope, which is taken overhead. If the stages are numbered from top to bottom, then the primary feed stream, which is essentially an azeotropic mixture of 87 kmol/h of ethanol and 13 kmol/h of water, is fed to stage 4 of the column. A second feed stream, which originates from a decanter, consists of 98 kmol/h of ethanol, 214 kmol/h of benzene and 61 kmol/h of water and is fed to the top tray of the column. In this particular example it is assumed that all stages except the reboiler and condenser operate adiabatically. Thus, there are $n_s(2n_c + 2) = 336$ equations and variables in our component flow rate

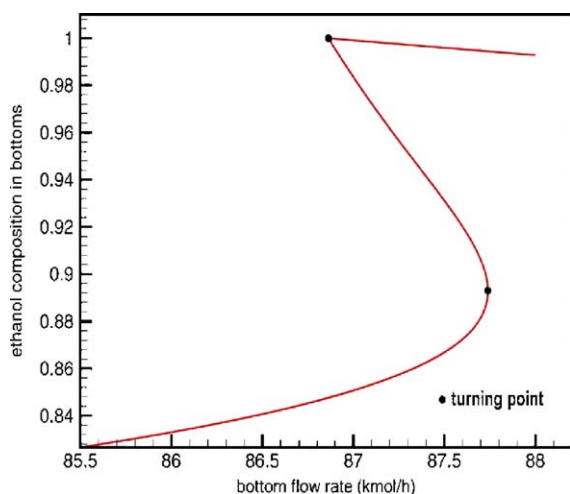


Fig. 2. Parameterized two-phase solutions to an azeotropic distillation.

formulation of this distillation model. Here we have used the UNIQUAC model with parameters given by Prokopakis and Seider (1983) to model the liquid phase activity coefficients and the Hayden-O'Connell model for the vapor phase.

Fig. 2 shows the solutions to this column as a function of bottoms flow rate. Note that there are three solutions for any bottoms flow rate between 86.8634 and 87.7385 kmol/h for a fixed reflux ratio of 1.1.

7. Newton's method

For a reflux ratio of 1.1 and a bottoms flow rate of 87 kmol/h, the nonideal starting point strategy in Venkataraman and Lucia together with Newton's method can find the high purity ethanol solution in nine iterations. Newton's method also easily finds the low purity ethanol solution for the exact same specifications from an ideal starting point in 12 iterations – 5 iterations to generate the ideal solution starting point as described in Venkataraman and Lucia and 7 iterations to actually find the low purity ethanol solution. For this distillation it is the intermediate purity solution that is difficult to find. We quote Venkataraman and Lucia (p. 64, 1988). "A third liquid composition profile intermediate to the above two profiles was also obtained with a very careful initialization procedure . . . There exists an extremely narrow region of composition about the final solution from which convergence to this intermediate profile can be obtained." This very small domain of attraction for the intermediate solution requires a fair amount of work to find!

8. Continuation

We also used Newton's method with homotopy-continuation to find solutions to this distillation for the same exact set of specifications. Here the bottoms flow rate, B , was

used as the continuation parameter and an initial stepsize of $\Delta B = 0.01$ kmol/h was used. However, once again many function evaluations were required in the neighborhood of the turning points. From the high purity ethanol solution, which was located in nine function evaluations from the non-ideal starting point of Venkataraman and Lucia, continuation required an additional 1006 function evaluation to find the intermediate and low purity ethanol solutions.

9. Terrain-following

Using the nonideal starting strategy described in Venkataraman and Lucia (1988), Newton's method easily converges to the high purity ethanol solution in nine iterations to an accuracy of $\|F\| \leq 10^{-6}$. The corresponding ethanol composition of the bottoms stream is 0.999085. From here, partial eigen-decomposition using the inverse power method and uphill Newton movement initiated in the eigen-direction associated with the smallest eigenvalue ($\lambda = 5.9402 \times 10^{-13}$) finds a saddle point in four iterations. The ethanol composition of the bottoms stream at this saddle is 0.992836 where the principal negative eigenvalue of the Hessian matrix of $F^T F$ is -9.85938×10^{-7} . Downhill movement from this saddle in a direction away from the high purity solution finds the intermediate ethanol purity solution with an ethanol composition of 0.983440 in another 12 Newton iterations. Using the inverse power method, the smallest eigenvalue of $J^T J$ at the intermediate solution is $\lambda = 3.9080 \times 10^{-13}$. Uphill movement from this intermediate purity solution in the eigen-direction associated with this smallest eigenvalue and away from the (first) saddle locates a second saddle point in eight iterations. The ethanol composition of the bottoms stream at this second saddle point is 0.897348. The principal negative eigenvalue at this second saddle is -6.61202×10^{-7} . Downhill movement from this second saddle finds the low purity solution with an ethanol composition of 0.850672 in 23 more Newton iterations. In all, only 56 function evaluations were needed to easily find all three solutions and two saddle points.

10. Conclusions

The terrain-following methodology of Lucia and Yang (2002, 2003) was successfully applied to two distillation examples. Numerical results for these and other examples have been extremely encouraging and clearly show that the terrain methodology is a reliable and efficient technique for solving systems of nonlinear algebraic equations, particularly those with multiple solutions.

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