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On the Equations of Holland in the Solution of Problems in Multicomponent Distillation

Abstract: Holland developed certain equations to be used to accelerate or induce convergence in multicomponent distillation calculations. In practice this procedure has been the most successful of any adjunct to the basic Thiele-Geddes or Lewis-Matheson procedure for solving these problems. It is of importance, therefore, to ascertain the conditions under which the Holland equations can be guaranteed to possess the required type of solution at each iteration. The types of specifications which fulfill these conditions are determined.

Introduction

Holland[1] suggested a procedure for accelerating or inducing convergence in the solution of multicomponent distillation problems. In practice this procedure has been the most successful of any adjunct to the basic Thiele–Geddes[2] or Lewis–Matheson[3] procedure for solving multicomponent distillation problems. When coupled with other computational improvements such as the tridiagonal matrix solution as carried out by Ball,[4] the "constant composition" heat balance calculation suggested by Holland[1] and the author's "adaptive parameter adjustment" scheme[5] for improving the "K_b-method" of determining stage temperatures, the fastest and most widely applicable algorithm known to the author results.

It is of importance, therefore, to ascertain the conditions under which the Holland equations can be guaranteed to possess the required type of solution at each iteration. It is often assumed that such a solution exists and is unique under any circumstances which may occur during the course of computation. Without resorting to pathological situations, however, it is a straightforward procedure to exhibit counterexamples to this assumption when the temperature of a product stream is specified or the mole fraction is specified for a component in a product stream. This was done in Ref. 5 and an example is also given in the appendix to this paper. On the other hand, as will be proven subsequently, when the total flow of each product stream is specified a unique positive finite solution to the Holland equations always exists.

For a full treatment of the way in which the subsequent Eqs. (1) arise the reader is referred to Holland.[1] More briefly, using the diagram of a complex column as given in Fig. 1, we have a device for separating a vapor-liquid mixture of various chemical compounds ("components") into different fractions represented by the streams leaving the unit. The purpose of this fractionation is to obtain in the bottom product stream ("bottoms") a preponderance of those compounds having relatively large molecular weights ("heavies"), to obtain in the overhead product stream ("distillate") a preponderance of those compounds having relatively small molecular weights ("lights") and to obtain in each remaining product stream ("side-draw") a mixture with a preponderance of compounds having molecular weights within a narrow range. Many columns have only one feed and no side-draws, and are accordingly referred to as simple columns, as distinguished from the complex column depicted in Fig. 1.

A column, or tower, is operated by supplying heat, Q_R, through the reboiler at the bottom of the tower and abstracting heat, Q_C, through the condenser at the top of the tower. Liquid, L, from each plate falls to the next lower plate while vapor, V, rises to the next higher plate, thus causing the higher compounds to become more concentrated in the upper part of the unit while the heavier components become more concentrated in the lower part of the unit; however, all components are present to some extent throughout the tower. Also sometimes used though not shown in Fig. 1 are interheaters and intercoolers which supply and abstract heat at various places along the tower. The operation of a tower is determined by many things such as the total rate of feed at each feed location, the rate at which heat is supplied to and abstracted from the tower, the total rate ("withdrawal rate")

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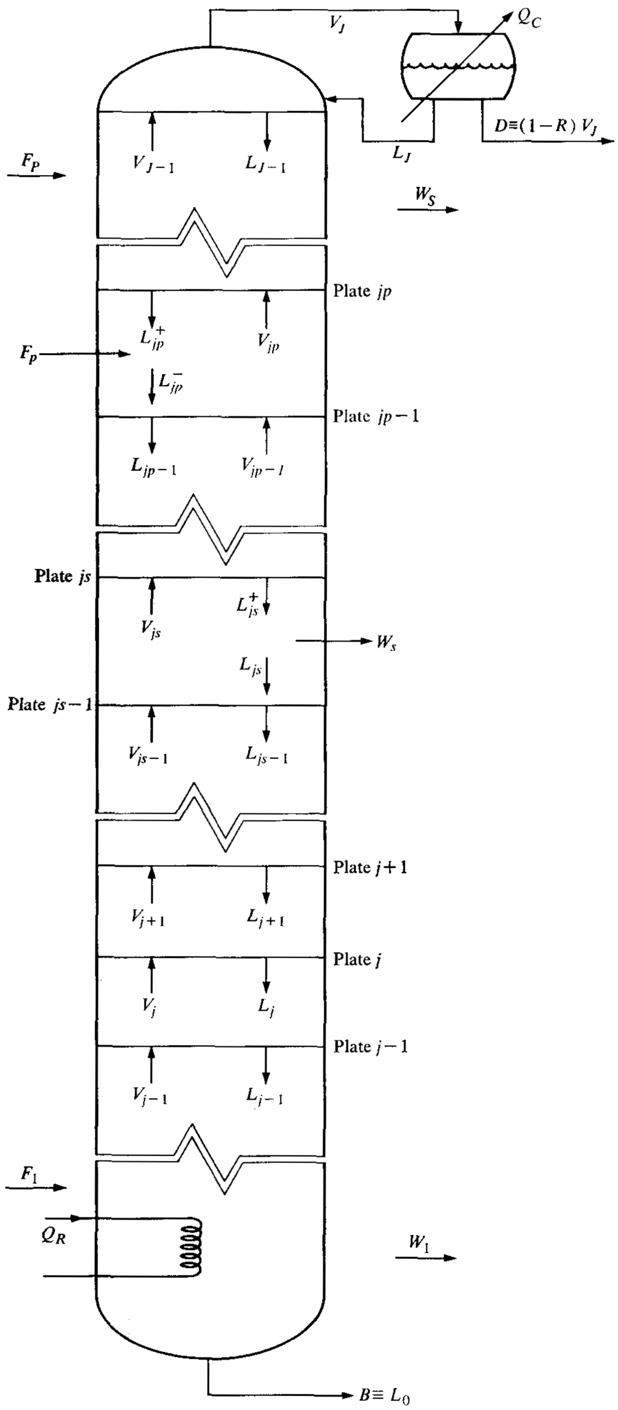


Figure 1 A complex distillation column.

at which each product stream is produced and the relative locations of the feed streams, product streams, interheaters and intercoolers. Regardless of how the column is operated, however, when conditions do not change in time (i.e. steady state) the total rate at which each component leaves the tower must be the same as the total rate at which it is fed to the column. It is this fact which is used to derive the individual summands in Eqs. (1). As these relations which are used as auxiliary formulae to accelerate or induce convergence in iterative methods for solving distillation problems have been derived several places in the literature they will not be rederived here.

Existence and uniqueness proofs

The Holland equations for a complex column (multiple feeds and side-draws) [see Holland[1], Billingsley[6]] where the total withdrawal rate of the bottom product stream and of each side-draw are specified are

$$G_0 \equiv B - \sum_i \frac{F_i^0}{1 + \left(\frac{d_i}{b_i}\right)_c \theta_0 + \sum_{s=1}^{S} \left(\frac{w_{is}}{b_i}\right)_c \theta_s} = 0$$

$$G_s \equiv W_s - \sum_i \frac{\left(\frac{w_{is}}{b_i}\right)_c \theta_s F_i^0}{1 + \left(\frac{d_i}{b_i}\right)_c \theta_0 + \sum_{s=1}^{S} \left(\frac{w_{is}}{b_i}\right)_c \theta_s} = 0, \qquad (1)$$

$$s = 1, 2, 3, \cdots, S,$$

where the variables are defined in the section on nomenclature. The unknowns in these equations are the θ_s, $s = 0, 1, \cdots, S$. All known quantities are positive and an overall material balance around the unit requires

$$\sum_i F_i^0 - B - \sum_s W_s = D > 0. \qquad (2)$$

To simplify subsequent discussion a vector, $\Theta = (\theta_0, \theta_1, \theta_2, \cdots, \theta_S)^T$, will be termed positive or non-negative if and only if $\theta_s > 0$ or $\theta_s \geq 0$, $s = 0, 1, \cdots, S$. Only non-negative vectors, Θ, will be considered henceforth. Obviously, a positive solution, $\Theta = (1, 1, \cdots, 1)^T$ exists when all the d_i, b_i and w_{is} are values actually taken from an operating unit since, in this case, a component material balance around the entire unit yields

$$F_i^0 = b_i + d_i + \sum_s w_{is}$$

or

$$B = \sum_i b_i = \sum_i F_i^0/[1 + (d_i/b_i) + \sum_s (w_{is}/b_i)]$$

$$W_s = \sum_i w_{is} = \sum_i (w_{is}/b_i) b_i.$$

It will be shown that a positive solution exists for arbitrary positive values of $(d_i/b_i)_c$ and the $(w_{is}/b_i)_c$ provided only that the specified positive values B and W_s, $s = 1, 2, \cdots, S$ satisfy Eq. (2).

Certain partial derivatives required later are collected at this point for convenience. To simplify notation let

$$\beta_i^{-1} \equiv 1 + (d_i/b_i)_c \theta_0 + \sum_{s=1}^{S} (w_{is}/b_i)_c \theta_s > 0. \qquad (3)$$

Then,

$$\partial\beta_i/\partial\theta_0 = -(d_i/b_i)_c\beta_i^2,$$

$$\partial\beta_i/\partial\theta_p = -(w_{ip}/b_i)_c\beta_i^2,$$

$$\partial^2\beta_i/\partial\theta_0^2 = 2(d_i/b_i)_c^2\beta_i^3,$$

$$\partial^2\beta_i/\partial\theta_p^2 = 2(w_{ip}/b_i)_c^2\beta_i^3,$$

$$\partial^2\beta_i/\partial\theta_0\,\partial\theta_p = 2(d_i/b_i)_c(w_{ip}/b_i)_c\beta_i^3,$$

$$\partial^2\beta_i/\partial\theta_p\,\partial\theta_q = 2(w_{ip}/b_i)_c(w_{iq}/b_i)_c\beta_i^3,$$

$$\partial G_0/\partial\theta_0 = \sum_i (d_i/b_i)_c F_i^0\beta_i^2 > 0,$$

$$\partial G_0/\partial\theta_s = \sum_i (w_{is}/b_i)_c F_i^0\beta_i^2 > 0,$$

$$\partial G_s/\partial\theta_0 = \theta_s \sum_i (w_{is}/b_i)_c(d_i/b_i)_c F_i^0\beta_i^2 > 0,$$

$$\partial G_s/\partial\theta_p = \theta_s \sum_i (w_{is}/b_i)_c(w_{ip}/b_i)_c F_i^0\beta_i^2 = (\theta_s/\theta_p)\,\partial G_p/\partial\theta_s > 0,$$

$$\partial G_s/\partial\theta_s = \theta_s \sum_i (w_{is}/b_i)_c^2 F_i^0\beta_i^2 - \sum_i (w_{is}/b_i)_c F_i^0\beta_i = -\sum_i \left[1 + \left(\frac{d_i}{b_i}\right)_c \theta_0 + \sum_{p\neq s}\left(\frac{w_{ip}}{b_i}\right)_c \theta_p\right] \cdot \left(\frac{w_{is}}{b_i}\right)_c F_i^0\beta_i^2 = -(\theta_0/\theta_s)(\partial G_s/\partial\theta_0) - \sum_{p;\, s\neq p\geqq 0} \partial G_p/\partial\theta_s < 0,$$

$$\partial^2 G_0/\partial\theta_s^2 = -2\sum_i (w_{is}/b_i)_c^2 F_i^0\beta^3 < 0,$$

$$\partial^2 G_s/\partial\theta_s^2 = 2\sum_i \left[1 + \left(\frac{d_i}{b_i}\right)_c \theta_0 + \sum_{p\neq s}\left(\frac{w_{ip}}{b_i}\right)_c \theta_p\right] \cdot \left(\frac{w_{is}}{b_i}\right)_c^2 F_i^0\beta_i^3 > 0 \qquad (4)$$

Further, by summing Eqs. (1) one obtains using Eq. (3),

$$G_{S+1} \equiv B + \sum_{s=1}^{S} W_s - \sum_i [\beta_i^{-1} - (d_i/b_i)_c\theta_0]F_i^0\beta_i = \sum_{s=0}^{S} G_s = 0,$$

which may be rearranged using Eq. (2) to

$$D - \sum_i (d_i/b_i)_c\theta_0 F_i^0\beta_i \equiv G_{S+1} = 0, \qquad (5)$$

where any solution of Eqs. (1) will necessarily be a solution of the set

$$G_s(\Theta) = 0, \quad s = 1, 2, 3, \cdots, S+1, \qquad (6)$$

and vice versa.

The main result of this paper may now be stated as a theorem.

Theorem 1. If the total flow rate of each product stream from a staged process is specified in accordance with Eq. (2), then a unique positive finite solution, $\Theta^* \equiv (\theta_0^*, \theta_1^*, \theta_2^*, \cdots, \theta_S^*)^T$, *to Eqs. (1) exists, provided only that the remaining (known) variables therein are positive and finite.* In a sense this is the strongest possible useful theorem since the quantities $(d_i/b_i)_c$ and $(w_{is}/b_i)_c$ must be positive and finite when derived from positive values of the b_i, d_i and w_{is}. The proof of Theorem 1 will be by induction.

Proof: Assume first that a solution exists for Eqs. (1) when there are only $S - 1$ side-draws so that $s = 1, 2, \cdots, S-1$. Then that same solution is also a solution for Eqs. (6) with $s = 1, 2, \cdots, S$. With D relabeled as W_S, θ_0 relabeled as θ_S and $(d_i/b_i)_c$ relabeled as $(w_{iS}/b_i)_c$. This solution will be denoted as $\Theta^*(0) \equiv [0, \theta_1^*(0), \theta_2^*(0), \theta_3^*(0), \cdots, \theta_S^*(0)]^T$. Thus, the overhead product in the case of $S - 1$ side-draws is to become the Sth side-draw in the case of S side-draws, with the additional stream being incorporated as a new overhead product stream. Hence, when there are S side-draws, $\Theta^*(0)$ satisfies all but the first of Eqs. (1) with $s = 1, 2, \cdots, S$. Now each of Eqs. (1) is, in the region of interest, a continuous function of each θ_s, $s = 0, 1, \cdots, S$. Therefore $\Theta^*(0)$ is a point on the continuous curve

$$\Theta^*(\theta_0) \equiv [\theta_0, \theta_1^*(\theta_0), \theta_2^*(\theta_0), \cdots, \theta_S^*(\theta_0)]^T,$$

where each point on $\Theta^*(\theta_0)$ satisfies all except perhaps the first of Eqs. (1) with $s = 1, 2, 3, \cdots, S$. $\Theta^*(\theta_0)$ exists since it contains $\Theta^*(0)$; the question which must be answered is "does $\Theta^*(\theta_0)$ intersect the surface $G_0 = 0$?" To demonstrate that it does four conditions will be established concerning Eqs. (1). They are

A. The continuous surface $G_0 = 0$ is restricted to a finite part of the first orthant.
B. $\Theta^*(\theta_0)$ is positive except at the point $\Theta^*(0)$ where it is non-negative.
C. $\Theta^*(0)$ lies below the surface $G_0 = 0$, that is $G_0[\Theta^*(0)] < 0$.
D. The directional derivative of G_0 in the direction of $\Theta^*(\theta_0)$ is positive and bounded away from zero so long as $\Theta^*(\theta_0)$ lies below or on $G_0 = 0$; that is $G_0[\Theta^*(\theta_0)] \leq 0$.

Condition A. To show that the surface $G^0 = 0$ is confined to a finite part of the first orthant note first that Eqs. (1) [or Eqs. (6)] satisfy the conditions of the implicit function theorem [see for instance Flemming[7]] in the region of interest. Hence, for fixed values of G_s, in particular $G_s = 0, s = 0, 1, 2, \cdots, S$ each of Eqs. (1) defines one of the θ_s in terms of the remaining θ_p. Thus, using Eqs. (4)

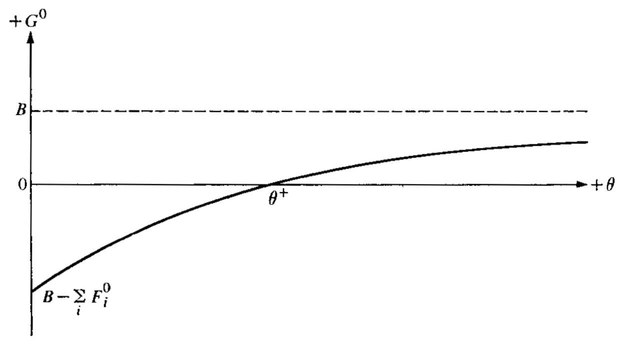


Figure 2 The function $G_0{}^0$.

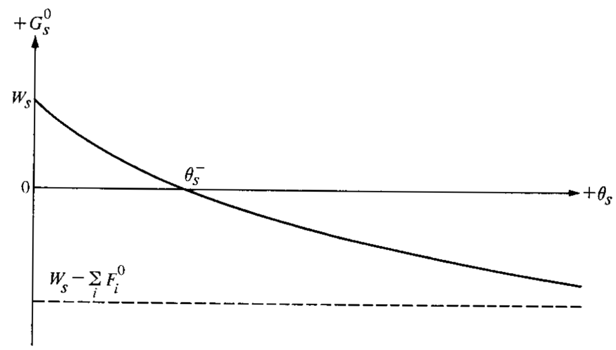


Figure 4 The function $G_s{}^0$.

Figure 3 The surface $G_0 = 0$.

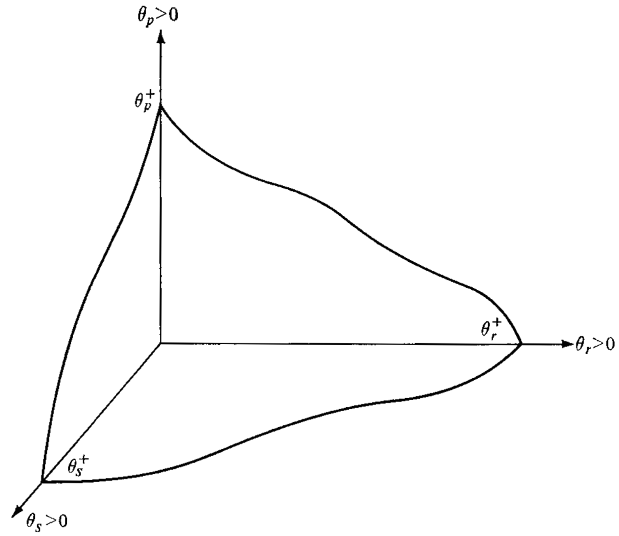
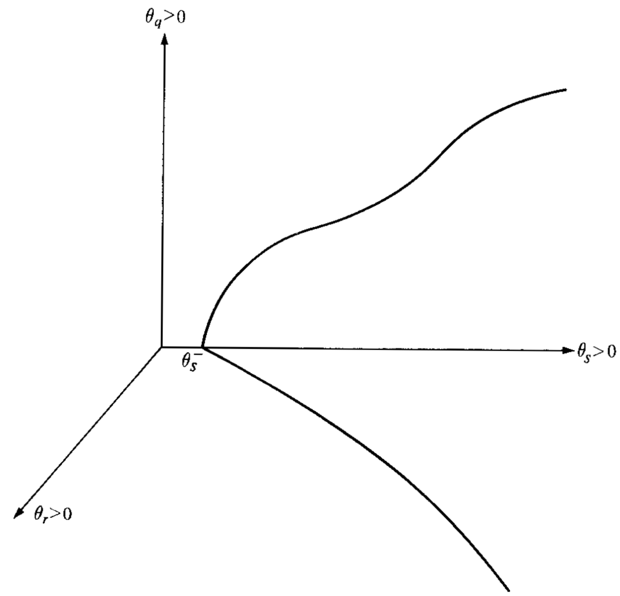


Figure 5 The surface $G_s = 0$.



$$\left.\frac{\partial \theta_q}{\partial \theta_r}\right|_{G_0=0} = -\frac{\partial G_0/\partial \theta_r}{\partial G_0/\partial \theta_q} < 0 \tag{7}$$

on the level surface $G_0 = 0$, or any level surface of G_0 for that matter. Furthermore, on the θ_s-axis, $\theta_p = 0$ for all $p \neq s$ so that G_0 reduces to

$$G_0^0(\theta_s) \equiv B - \sum_i \frac{F_i^0}{1 + (w_{is}/b_i)_c \theta_s} \tag{8}$$

with appropriate changes in nomenclature when $s = 0$. Equations (4) and (8) show that

(a) $\partial G_0/\partial \theta_s > 0$ for $\theta_s \geq 0$,
(b) $G_0^0\,(\pm\,\infty) = B > 0$,
(c) $G_0^0\,(0) = B - \sum_i F_i^0 < 0$,
(d) $d^2G_0^0/d\theta_s^2 < 0$ for $\theta_s \geq 0$,
(e) G_0^0 is continuous except at $\theta_s = -(b_i/w_{is})_c < 0$.
(f) In view of (a) through (e) G_0^0 has the form shown in Fig. 2, and $G_0^0 = 0$ has exactly one positive root, θ_s^+.

Thus, the $G_0 = 0$ surface intersects each coordinate axis exactly once. This, together with Eq. (7), shows the surface $G_0 = 0$ in the first orthant must lie within the region for which $\theta_s \leq \theta_s^+$, $s = 0, 1, 2, \cdots, S$. Indeed any 3-dimensional cross-section of $G_0 = 0$ must have the general form depicted in Fig. 3.

Condition B. To show that $\Theta^*(\theta_0)$ is positive except at the point $\Theta^*(0)$ where it is non-negative requires examination of the somewhat more complicated surfaces $G_s = 0$, $s \neq 0$. It is first noted that the surface $G_s = 0$ can intersect no hyperplane on which $\theta_s = 0$ since at $\theta_s = 0$, $G_s = W_s > 0$. Consequently, $G_s = 0$ can intersect no coordinate axis except perhaps the θ_s-axis. On this axis G_s becomes

$$G_s^0(\theta_s) \equiv W_s - \sum_i \frac{(w_{is}/b_i)_c \theta_s F_i^0}{1 + (w_{is}/b_i)_c \theta_s}.$$

Inspection of this relation, together with Eqs. (4), shows

(a) $\partial G_s^0/\partial \theta_s < 0$ for $\theta \geq 0$,
(b) $G_s^0(\pm \infty) = W_s - \sum_i F_i^0 < 0$,
(c) $G_s^0(0) = W_s > 0$,
(d) $d^2G_s^0/d\theta_s^2 > 0$ for $\theta_s \geq 0$,
(e) G_s^0 is continuous except at $\theta_s = -(b_i/w_{is})_c < 0$.
(f) In view of (a) through (e) G_s^0 has the form shown in Figure 4, and $G_s^0 = 0$ has exactly one positive root, θ_s^-.

Thus the $G_s = 0$ surface intersects the θ_s-axis exactly once.

Further examination of the surface $G_s = 0$, $s \neq 0$ shows that it cannot exist in the positive orthant for $\theta_s < \theta_s^-$. To see this consider

$$\left.\frac{\partial \theta_r}{\partial \theta_s}\right|_{G_s=0} = -\frac{\partial G_s/\partial \theta_s}{\partial G_s/\partial \theta_r}$$

$$= -\left[-\frac{\theta_0}{\theta_s}\frac{\partial G_s}{\partial \theta_0} - \frac{\theta_r}{\theta_s}\frac{\partial G_s}{\partial \theta_r} - \sum_{p;r\neq p\neq s} \frac{\partial G_p}{\partial \theta_s}\right]$$

$$\cdot\left(\frac{\partial G_s}{\partial \theta_r}\right)^{-1} > \frac{\theta_r}{\theta_s} \geq 0, \quad \text{for} \quad \theta_r \geq 0 < \theta_s.$$

Thus $\Theta^*(\theta_0)$ is confined to the region $\theta_s \geq \theta_s^-$, $s = 1, 2, \cdots, S$. Hence $\Theta^*(\theta_0)$ is positive except when $\theta_0 = 0$.

To complete the picture of the surface $G_s = 0$ one notes with the aid of Eqs. (4) that

$$\left.\frac{\partial \theta_p}{\partial \theta_q}\right|_{G_s=0} = -\frac{\partial G_s/\partial \theta_q}{\partial G_s/\partial \theta_p} < 0$$

so that any 3-dimensional cross section of $G_s = 0$ is as depicted in Fig. 5 if the cross section is not orthogonal to the θ_s-axis and is as depicted in Fig. 3 otherwise.

Condition C. To show that $\Theta^*(0)$ lies below the surface $G_0 = 0$ requires a somewhat more involved construction. Consider then the line from the origin, $\theta_s = 0$, $s = 0, 1, \cdots, S$, through the point $\Theta^*(0)$ as is shown in Fig. 6 for the case $S = 2$. The tangent to this line is the vector $\boldsymbol{\sigma}$, where

$$\sigma_s = \theta_s^*(0)/[\Theta^*(0) \cdot \Theta^*(0)]^{\frac{1}{2}}$$

is the sth component of $\boldsymbol{\sigma}$ and the dot product is the usual vector inner product. The gradient of G_0 is the vector, $\mathbf{G}_0$, of partial derivatives of G_0. Now

$$\sigma_s \frac{\partial G_0}{\partial \theta_s} \begin{cases} = 0, & s = 0 \\ > 0, & s \neq 0 \quad \text{since} \quad \theta_s^* \geq \theta_s^- \end{cases}$$

so that the derivative, $dG_0/d\sigma$, of G_0 in the direction of $\boldsymbol{\sigma}$ is positive, and

$$dG_0/d\sigma = [\boldsymbol{\sigma} \cdot \mathbf{G}_0]^{\frac{1}{2}} > 0. \tag{9}$$

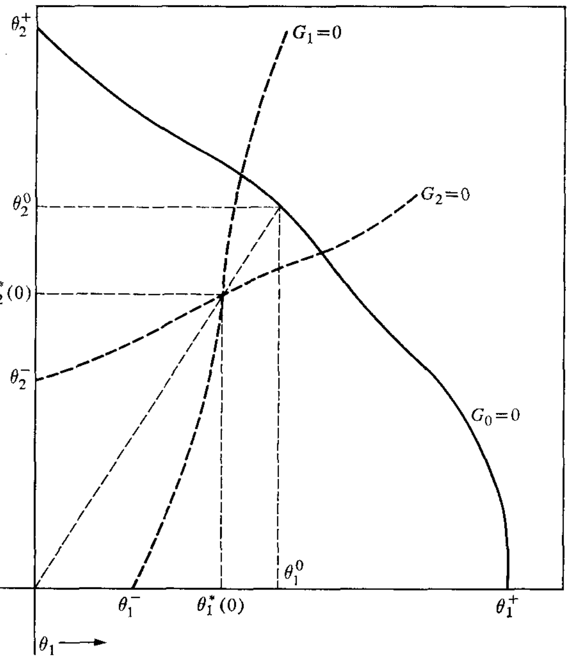


Figure 6 Intersection of surfaces $G_s = 0$ with the hyperplane $\theta_0 = 0$.

Furthermore, this quantity is a continuous function of each θ so that it assumes in the region $0 \leq \theta_s \leq \theta_s^+$ a minimum value, $(dG_0/d\sigma)^-$, which, in view of $\partial^2 G_0/\partial\theta_s^2$ from Eqs. (4), occurs at $\Theta^+ \equiv (\theta_0^+, \theta_1^+, \cdots, \theta_S^+)^T$ (see for instance Hyder and Simpson[8]). At the origin $G_0 = B - \sum_i F_i^0 < 0$ so that at a distance $(-B + \sum_i F_i^0)/(dG_0/d\sigma)^-$ from the origin on the line through $\Theta^*(0)$, $G_0 \geq 0$. This line must then intersect $G_0 = 0$ exactly once because of the continuity of G_0 and the constant sign of $dG_0/d\sigma$ (see for instance Hyder and Simpson[8]). Denote this point of intersection by $\Theta^0 \equiv (0, \theta_1^0, \cdots, \theta_S^0)^T$.

It now must be shown that $\Theta^*(0)$ is nearer the origin than Θ^0, that is $\theta_s^0 = \alpha\theta_s^*(0)$ where $\alpha > 1$. To this end, note that $G_0(\Theta^0) = 0 = G_s[\Theta^*(0)]$ by construction so that according to Eq. (2)

$$-G_0(\Theta^0) + B + \sum_{s=1}^{S} \{-G_s[\Theta^*(0)] + W_s\}$$

$$= -D + \sum_i F_i^0 < \sum_i F_i^0.$$

Then by use of Eqs. (1) and (3) there is

$$\sum_i F_i^0\beta_i(\Theta^0) + \sum_{s=1}^{S}\sum_i (w_{is}/b_i)_c \theta_s^*(0) F_i^0 \beta_i[\Theta^*(0)]$$

$$< \sum_i F_i^0$$

which, since the F_i^0 are independently positive, requires

$$\beta_i(\Theta^0) - 1 + \beta_i[\Theta^*(0)] \sum_{s=1}^{S} (w_{is}/b_i)_c \theta_s^*(0) < 0$$

for each i. From Eq. (3) it is seen that

$$\sum_{s=1}^{S} (w_{is}/b_i)_c \theta_s^*(0) = \{\beta_i[\Theta^*(0)]\}^{-1} - 1,$$

so that the last previous inequality becomes

$$\beta_i(\Theta^0) - \beta_i[\Theta^*(0)] < 0,$$

or since $\theta_0 = 0$ in both Θ^0 and $\Theta^*(0)$,

$$\left[1 + \sum_{s=1}^{S} \left(\frac{w_{is}}{b_i}\right)_c \alpha\theta_s^*(0)\right]^{-1} < \left[1 + \sum_{s=1}^{S} \left(\frac{w_{is}}{b_i}\right)_c \theta_s^*(0)\right]^{-1}.$$

Term by term inspection of this expression shows $\alpha > 1$. Hence $\Theta^*(0)$ is nearer the origin than Θ^0. Consequently, Eq. (9) shows that $G_0[\Theta^*(0)] < 0$. This had been assumed in a previous paper, Ref. 6.

Condition D. The intersection of all surfaces $G_s = 0$, $s = 1, 2, \cdots, S$ is represented as a function of θ_0 by the curve $\Theta^*(\theta_0)$. The existence of this curve has been established in the region $G_0 < 0$, $\theta_s \geq \theta_s^-$, $s = 1, 2, \cdots, S$. Because of the continuity of all functions involved, $\Theta^*(\theta_0)$ is a continuous vector function of θ_0. It is conceivable, however, that this curve never leaves the region $G_0 < 0$. In this case it would not intersect the surface $G_0 = 0$ and consequently no solution to Eqs. (1) would exist. The behavior of $\Theta^*(\theta_0)$ is investigated in the following paragraphs by determining the sign of the derivative of G_0 in the direction of the tangent to $\Theta^*(\theta_0)$.

Let η_s be proportional to the cosine of the angle between the θ_s-axis and the tangent to $\Theta^*(\theta_0)$. Thus, $\eta_s = \eta_s(\theta_0)$. Now at any point $\Theta^*(\theta_0)$ must be orthogonal to the normal to each surface $G_p = 0$, $p = 1, 2, \cdots, S$. This is expressed by

$$\sum_{s=0} \eta_s\, \partial G_p/\partial\theta_s = 0, \qquad p = 1, 2, \cdots, S. \tag{10}$$

The number of unknowns, η_s, exceeds by one the number of Eqs. (10); hence, η_0 will be set equal 1 so that Eqs. (10) become

$$A\eta = -\gamma, \tag{11}$$

where

$$A \equiv \begin{bmatrix} \partial G_1/\partial\theta_1 & \cdots & \partial G_1/\partial\theta_S \\ \vdots & & \vdots \\ \partial G_S/\partial\theta_1 & \cdots & \partial G_S/\partial\theta_S \end{bmatrix}$$

$$\eta \equiv (\eta_1, \eta_2, \cdots, \eta_S)^T$$

$$\gamma \equiv (\partial G_1/\partial\theta_0, \partial G_2/\partial\theta_0, \cdots, \partial G_S/\partial\theta_0)^T$$

From Eqs. (4) it is seen that

(a) each element of γ is positive,
(b) each element of A which is not on the principal diagonal is positive,
(c) each principal diagonal element of A is negative,
(d) A is diagonally dominant by columns. That is, if $A = (a_{rs})$, then $|a_{rr}| > \sum_r' |a_{rs}|$, where $\sum'$ denotes that a_{rr} is omitted.

It is desired to show that η is positive. To this end define the diagonal matrix, $M = (m_{rs})$, to have $m_{ss} = -\partial G_s/\partial\theta_s$ (> 0) and $m_{rs} = 0$ for $r \neq s$. Equation (11) may then be manipulated as follows.

$$[-M + (A + M)]\eta = -\gamma,$$

$$[I - (A + M)M^{-1}](-M)\eta = -\gamma.$$

Now all principal diagonal elements of $A + M$ are zero. In view of the diagonal dominance of A and the fact that only the principal diagonal elements of M^{-1} are non-zero, each being the reciprocal of the corresponding element of M, the product $(A + M)\, M^{-1} = (c_{rs})$ has

$$\max_{1 \leq s \leq S} \sum_{r=1}^{S} |c_{rs}| < 1.$$

It is known from matrix theory (see Varga[9], pp. 17, 20, 84) that the spectral radius does not exceed the left side of this relation and that if the spectral radius of an irreducable matrix, say $(A + M)\, M^{-1}$, is less than unity then $[I - (A + M)M^{-1}]^{-1}$ exists and

$$[I - (A + M)M^{-1}]^{-1} = I + \sum_{k=1}^{\infty} [(A + M)M^{-1}]^k.$$

With this relation Eq. (11) becomes

$$\eta = M^{-1}\left\{I + \sum_{k=1}^{\infty} [(A + M)M^{-1}]^k\right\}\gamma. \tag{12}$$

Since each element in every factor on the right side of Eq. (12) is non-negative no subtraction occurs and η is positive,

$$\eta_s > 0, s = 0, 1, \cdots, S. \tag{13}$$

In view of Eqs. (13) $\Theta^*(\theta_0)$ cannot enter the region $\theta_s < \theta_s^*(0)$, and moreover, it intersects the boundary of the positive orthant only once, namely at the point $\Theta^*(0)$. The tangent to $\Theta^*(\theta_0)$ is the vector $\boldsymbol{\tau}$, where

$$\tau_s = \eta_s/(\eta\cdot\eta)^{\frac{1}{2}} > 0$$

is the sth component of $\boldsymbol{\tau}$. Now then,

$$\tau_s \partial G_0/\partial\theta_s > 0, \quad s = 0, 1, \cdots, S$$

so that the derivative, $dG_0/d\tau$, of G_0 in the direction of $\boldsymbol{\tau}$ is positive, and

$$dG_0/d\tau = (\boldsymbol{\tau}\cdot \mathbf{G}_0)^{\frac{1}{2}} > 0.$$

Furthermore, this quantity is a continuous function of each θ so that in the region, $\theta_s^*(0) \leqq \theta_s \leqq \theta_s^+$, $s = 0, 1, 2, \cdots, S$, it assumes a minimum value, $(dG_0/d\tau)^-$. Recall that the continuous surface $G_0 = 0$ is confined to the region $0 \leqq \theta_s \leqq \theta_s^+$, $s = 0, 1, \cdots, S$, and therefore at a distance not exceeding $-G_0[\Theta^*(0)]/(dG_0/d\tau)^-$ from $\Theta^*(0)$ along $\Theta^*(\theta_0)$, $G_0[\Theta^*(\theta_0)] > 0$. Because of the continuity of $G_0 = 0$ and of $\Theta^*(\theta_0)$, these two must intersect. Since $dG_0/d\tau$ does not change sign, only one such intersection is possible. Denote this intersection by $\Theta^*(\theta_0^*)$ or simply Θ^*. The solution to Eqs. (1) is then Θ^* when $s = 1, 2, \cdots, S$, and this has been shown to result from the supposition that a positive solution exists for these equations when $s = 1, 2, \cdots, S - 1$.

It remains only to show that a solution exists when $s = 1 = S$. To this end, one notes first that in the case $S = 1$, the curve $\Theta^*(\theta_0)$ becomes the plane curve $G_1(\theta_0, \theta_1) = 0$. Since, as mentioned previously, G_1 satisfies the conditions of the implicit function theorem, $G_1 = 0$ specifies θ_1 as a function of θ_0. Now the only way the arguments A, B, C and D relied on the induction assumption was that it guaranteed the existence of $\Theta^*(0)$. Thus, if $G_1(0, \theta_1) = 0$ can be shown to possess exactly one solution, the preceding derivation will hold for the case $S = 1$. This has already been accomplished, however, in argument B, since $G_1(0, \theta_1)$ is the same as $G_s^0(\theta_s)$. Figure 7 depicts the situation in the case $S = 1$.

It is to be noted that this proof remains valid when the total amount of the same fixed subset of components is specified in each product stream. This is because the only requirement on the summations over components is that they include all components which have been specified in the product streams.

Summary

For the first time it is shown that a solution to the Holland equations exists and is unique at each iteration performed to obtain the answer to a distillation problem, provided the total withdrawal rate of each product stream is specified.

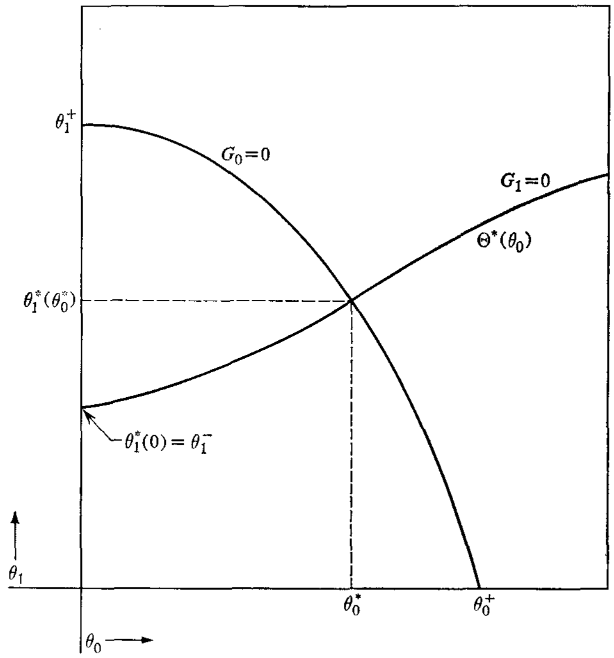


Figure 7 Intersection of curves $G_0 = 0$ and $\Theta^*(\theta_0)$ when $S = 1$.

Notation

A = matrix defined after Eq. (11)
b_i = rate of flow of component i in the bottom product stream
B = total rate of flow of the bottom product stream. $B = \sum_i b_i$
d_i = rate of flow of component i in the overhead product stream
D = total rate of flow of the overhead product stream. $D = \sum_i d_i$
f_i = feed rate of component i in a single feed
F_i^0 = total rate of feed of component i to the unit
G_0, G_s = functions defined by Eqs. (1) and (5)
G_0^0, G_s^0 = functions G_0 and G_s, respectively, evaluated with all but one θ set to zero
$\mathbf{G}_0$ = the gradient vector of G_0
$dG_0/d\sigma$ = the derivative of G_0 in the direction of $\boldsymbol{\sigma}$
$dG_0/d\tau$ = the derivative of G_0 in the direction of $\boldsymbol{\tau}$
$(dG_0/d\sigma)^-$ = the minimum value of $dG_0/d\sigma$ in the region $0 \leqq \Theta \leqq \Theta^+$
$(dG_0/d\tau)^-$ = the minimum value of $dG_0/d\tau$ in the region $\Theta^*(0) \leqq \Theta \leqq \Theta^+$
I = the unit matrix (main diagonal elements each = 1, all other elements = 0)
M = matrix defined after Eq. (11)
S = number of side-draws from the unit
w_{is} = flow rate of component i in the sth side-draw up from the bottom of the unit
W_s = total flow rate of the sth side-draw up from the bottom of the unit. $W_s = \sum_i w_{is}$

Greek letters

α = constant defined after Eq. (9)
$\beta_i, \beta_i[\Theta]$ = variable defined by Eq. (3), and evaluated at the point $(\theta_0, \theta_1, \cdots, \theta_s)$
γ = vector defined after Eq. (11).
η_s = variable defined prior to Eq. (10)
η = vector defined after Eq. (11)
θ_0 = $(d_i/b_i)/(d_i/b_i)_c$ for all components
θ_s = $(w_{is}/b_i)/(w_{is}/b_i)_c$ for all components
$\theta_s^*(\theta_0)$ = an element of the solution to Eqs. (6) with θ_0 regarded as a parameter

θ_s^+ = the value of θ_s at which the surface $G_0 = 0$ intersects the θ_s-axis

θ_s^- = the value of θ_s at which the surface $G_s = 0$ intersects the θ_s-axis; $s = 1, 2, \cdots, S$.

θ_s^0 = the value of θ_s at which the line through the origin and the point $[0, \theta_1^*(0), \theta_2^*(0), \cdots, \theta_S^*(0)]$ intersects the surface $G_0 = 0$

Θ = the vector $(\theta_0, \theta_1, \theta_2, \cdots, \theta_S)^T$. This represents a point in the space spanned by the θ_s, $s = 0, 1, 2, \cdots, S$

$\Theta^*(\theta_0)$ = the vector $\{\theta_0, \theta_1^*(\theta_0), \theta_2^*(\theta_0), \cdots, \theta_S^*(\theta_0)\}^T$

Θ^+ = the vector $(\theta_0^+, \theta_1^+, \cdots, \theta_S^+)^T$

Θ^0 = the vector $(0, \theta_1^0, \theta_2^0, \cdots, \theta_S^0)^T$

σ = distance from the origin toward the point Θ^0 along the line between these points. σ is positive when Θ is non-negative

σ_s = the s-th component of the tangent to the line between the origin and Θ^0. $\sigma_s = \theta_s^*(0)/[\Theta^*(0)\cdot\Theta^*(0)]^{\frac{1}{2}}$

$\boldsymbol{\sigma}$ = the tangent to the line between the origin and Θ^0. Since the line is straight, $\boldsymbol{\sigma}$ represents a vector of unit length along this line. $\boldsymbol{\sigma} = (0, \sigma_1, \sigma_2, \cdots, \sigma_S)^T$

τ = the distance from $\Theta^*(0)$ along the curve defined by $\Theta(\theta_0)$. τ is positive when Θ is non-negative.

τ_s = the s-th component of the tangent to the curve defined by $\Theta^*(\theta_0)$. $\tau_s = \eta_s/(\eta\cdot\eta)^{\frac{1}{2}}$

$\boldsymbol{\tau}$ = the tangent to the curve defined by $\Theta^*(\theta_0)$. $\boldsymbol{\tau} = (\tau_0, \tau_1, \cdots, \tau_S)^T$

Subscripts

c denotes the last previously computed value of the quantity to which it applies

i denotes component number

p, q, r, s denote side-draw number. Except where otherwise specified these assume the values $1, 2, \cdots, S$

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Appendix

In connection with the specific example of Ref. 5, Professor Holland has pointed out to the author that the limits on the value of a purity specification for which the Holland equations are applicable are easily found. For the case of a binary mixture separated in a simple column where the mole fraction, x_1, of one component in the bottoms product is specified there is

$$b_i = f_i/[1 + (d_i/b_i)_c\theta], \quad i = 1, 2$$

$$x_1 = b_1/(b_1 + b_2),$$

so that

$$\lim_{\theta\to 0} x_1 = f_1/(f_1 + f_2),$$

$$\lim_{\theta\to\infty} x_1 = \frac{f_1/(d_1/b_1)_c}{[f_1/(d_1/b_1)_c] + [f_2/(d_2/b_2)_c]}.$$

If, for instance, the specifications are $f_1 = 80$, $f_2 = 20$ and x_1 is the mole fraction of the lighter component in the bottoms,

$$\frac{80}{100} \geq x_1 \geq \frac{80/(b_1/d_1)_c}{[80/(b_1/d_1)_c] + [20/(b_2/d_2)_c]}.$$

For the case of $x_1 = 0.1$ there is

$$[8/(d_1/b_1)_c] + [2/(d_2/b_2)_c] \geq 80/(d_1/b_1)_c,$$

from which $36\,(d_2/b_2)_c \leq (d_1/b_1)_c$, so that if on any iteration $(d_1/b_1)_c < 36\,(d_2/b_2)_c$, then $\theta < 0$. Note further that simply setting $\theta = 0$ is unsuitable since then $b_i = f_i$ and there is no distillate. Obviously if $(d_1 b_1) < 36\,(d_2/b_2)$ in the actual solution another method is required. A similar argument may be applied to situations where the product temperature is specified. Thus, the conditions given previously by Lyster, et al.[10] are seen to be necessary but not sufficient to insure a solution to the Holland equations in the case of a purity specification.

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