R & D NOTES

On the Optimum Choice of Feed Stage in Staged Equilibrium Processes

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This communication has its origin in a classroom comment by E. R. Gilliland, made more than 20 years ago. We had encountered one of those situations in a distillation problem in which the feed could be introduced on either of two stages without affecting the total number of stages required. To a man, the class concluded that the choice of feed stage was immaterial, and Professor Gilliland characteristically remarked that our engineering consciences ought to be disturbed if we increased the entropy of the universe more than necessary. However, strictly thermodynamic criteria for feed stage location seem to be lacking—or at least unused. Feed stage location is apparently most often based on matching the ratios of the light and heavy key components in the feed and on the stage in question, or on minimizing the sum of the squares of the differences between feed and stage mol fractions. Neither method is thermodynamic in other than an intuitive sense, and the second can lead to serious error if applied blindly. In this communication a simplified criterion based on minimizing the irreversibility (entropy production) consequent to introducing the feed is developed. The criterion has been applied to a simple 3-component example in a stage-to-stage calculation using the Lewis assumptions.

The starting point in the development is an expression for the inequality of Clausius applicable to systems of variable mass (Van Wylen and Sonntag, 1965):

$$dS + \sum_0 \bar{S}_0 \delta m_0 - \sum_i \bar{S}_i \delta m_i - (\delta Q/T) \geqq 0 \qquad (1)$$

where:

S = the entropy of the system
$\bar{S}_0$, $\bar{S}_i$ = entropies per mole of streams leaving and entering the system respectively
δQ = increment of heat transmitted to the system
T = temperature of that part of the system to which heat is transmitted
δm_i, δm_0 = the mol increments entering and leaving the system respectively.

The extent to which the inequality of Equation (1) is honored is a quantitative measure of the irreversibility of the process, called the entropy production. Equation (1) may be divided by the time increment δt to yield an expression for the rate of entropy production, namely,

$$dS/\delta t + \sum_0 \bar{S}_0 \dot{m}_0 - \sum_i \bar{S}_i \dot{m}_i - \dot{Q}/T \geqq 0 \qquad (2)$$

where the dotted symbols denote rates.

Equation (2) is applied to the feed stage of a counterflow, steady state, adiabatic, staged equilibrium process. The liquid, vapor, and feed flow rates are denoted by L, G, and F respectively, with subscripts to denote the stage from which the stream comes and its phase. The expression is normalized by dividing each term by the product of the gas constant and the feed rate.

$$\dot{R}_s = \frac{L_f \bar{S}_{L,f}}{RF} + \frac{G_f \bar{S}_{G,f}}{RF} - \frac{L_{f+1} \bar{S}_{L,f+1}}{RF} - \frac{G_{f-1} \bar{S}_{G,f-1}}{RF} - \frac{\bar{S}_F}{R} \geqq 0 \qquad (3)$$

where $\dot{R}_s$ is the normalized rate of entropy production on the feed stage and is positive definite. Were one to omit the feed from the stage, there would still be an irreversibility (primed symbols) given by

$$\dot{R}_s' = \frac{L_f' \bar{S}'_{L,f}}{RF} + \frac{G_f' \bar{S}'_{G,f}}{RF} - \frac{L'_{f+1} \bar{S}'_{L,f+1}}{RF} - \frac{G'_{f-1} \bar{S}'_{G,f-1}}{RF} \geqq 0 \qquad (4)$$

Obviously the feed stage would be that stage for which the difference between the two positive definite quantities (entropy production consequent to introducing the feed) is a minimum, that is, $\dot{R}_s - \dot{R}_s' =$ minimum.

While the foregoing is a sufficient criterion by itself, and in a sufficiently sophisticated calculation in which one keeps track of all the thermodynamic properties on each stage one could compute the arguments by Equations (3) and (4), a more accessible formulation is essential for first cut calculations. This simpler formulation can be obtained by application of the material and energy balances to which the quantities in Equations (3) and (4) are subject.

The energy balance is

$$-L_f \bar{H}_{L,f} - G_f \bar{H}_{G,f} + L_{f+1} \bar{H}_{L,f+1} + G_{f-1} \bar{H}_{G,f-1} + F \bar{H}_F = 0 \qquad (5)$$

where the quantities $\bar{H}$ are the molal enthalpies. The material balance for component j is

$$-L_f x_{j,L,f} - G_f x_{j,G,f} + L_{f+1} x_{j,L,f+1} + G_{f-1} x_{j,G,f-1} + F x_{j,F} = 0 \quad (j = 1, 2, \ldots, J) \qquad (6)$$

where χ_j are the mol fractions.

Equations (5) and (6) may be normalized by dividing both sides by RFT_f and F respectively, T_f being the temperature of the stage. If the normalized forms of Equations (6) each be multiplied by the normalized chemical potentials, $\mu_{j,L,f}/(RT_f)$ and subtracted from Equation (3) and the normalized form of Equation (5) be added to the result, one may obtain

$$\begin{aligned}\dot{R}_s = &- V_f \sum_{j=1}^{J} \chi_{j,G,f}(\mu^+{}_{j,G,f} - \mu^+{}_{j,L,f}) \\ &+ C_{f+1} \sum_{j=1}^{J} \chi_{j,L,f+1} \Big(\mu^+{}_{j,L,f+1} - \mu^+{}_{j,L,f} \\ &- \frac{\overline{H}_{j,L,f+1}}{R}\Big(\frac{1}{T_{f+1}} - \frac{1}{T_f}\Big)\Big) \\ &+ V_{f-1} \sum_{j=1}^{J} \chi_{j,G,f-1} \Big(\mu^+{}_{j,G,f-1} - \mu^+{}_{j,L,f} \\ &- \frac{\overline{H}_{j,G,f-1}}{R}\Big(\frac{1}{T_{f-1}} - \frac{1}{T_f}\Big)\Big) \\ &+ \sum_{j=1}^{J} \chi_{j,F} \Big(\mu^+{}_{j,F} - \mu^+{}_{j,L,f} - \frac{\overline{H}_{j,F}}{R}\Big(\frac{1}{T_F} - \frac{1}{T_f}\Big)\Big)\end{aligned} \tag{7}$$

wherein the following definitions and thermodynamic relations have been used:

$$C \equiv L/F, \quad V \equiv G/F, \quad \mu_j{}^+ \equiv \mu_j/(RT)$$

$$\sum_{j=1}^{J} \mu_j\chi_j = \sum_{j=1}^{J} \overline{H}_j\chi_j - T\,\overline{S} = \overline{H} - T\,\overline{S}$$

Since the stage under consideration is an equilibrium stage, the first term on the right of Equation (7) is zero. The remaining terms, involving differences of chemical potentials are different from zero because of stage-to-stage variations in temperature, pressure, and composition. Pressure variations are usually not large enough to be significant. Temperature variations are usually significant; they may be treated by means of the thermodynamic relation:

$$\frac{\partial}{\partial T}(\mu_j/(RT)) = -\overline{H}_j/(RT^2) \tag{8}$$

As a first approximation, we assume $\overline{H}_j$ constant between any pair of stages and integrate Equation (8) to yield:

$$\int_{T_f}^{T_{f+1}} \frac{\partial}{\partial T}(\mu_{j,L}/(RT))\,dT \sim \overline{H}_{j,L}(1/T_{f+1} - 1/T_f)/R \tag{9}$$

With this approximation all the chemical potential terms in Equation (7) may be adjusted to the same temperature, that of the stage in question, so that the different subscripts reflect differences in composition only. Thus, by use of the approximate relation (9), Equation (7) takes the form:

$$\begin{aligned}\dot{R}_s = &C_{f+1} \sum_{j=1}^{J} \chi_{j,L,f+1}(\mu^+{}_{j,L,f+1} - \mu^+{}_{j,L,f})_{T_f} \\ &+ V_{f-1} \sum_{j=1}^{J} \chi_{j,G,f-1}(\mu^+{}_{j,G,f-1} - \mu^+{}_{j,G,f})_{T_f} \\ &+ \sum_{j=1}^{J} \chi_{j,F}(\mu^+{}_{j,F} - \mu^+{}_{j,L,f})_{T_f}\end{aligned} \tag{10}$$

where all the chemical potentials are evaluated at T_f, the subscripts reflecting composition variations only.

Equation (10) represents a simplified expression for the normalized rate of entropy production on the stage with the feed introduced. This must be compared with the corresponding term for the same stage without the feed. We denote the stage without the feed by single primes:

$$\begin{aligned}\dot{R}_s' = &C'_{f+1} \sum_{j=1}^{J} \chi'_{j,L,f+1}(\mu^{+\prime}{}_{j,L,f+1} - \mu^{+\prime}{}_{j,L,f})_{T_{f'}} \\ &+ V'_{f-1} \sum_{j=1}^{J} \chi'_{j,G,f-1}(\mu^{+\prime}{}_{j,G,f-1} - \mu^{+\prime}{}_{j,G,f})_{T_{f'}}\end{aligned} \tag{11}$$

The effect of introducing the feed on the irreversibility of the stage is the difference $(\dot{R}_s - \dot{R}_s')$, and it is this difference which is to be minimized by introducing the feed at the proper stage.

In a simple stage-to-stage calculation, starting from the reboiler, the terms involving V_{f-1} and V'_{f-1} will be unaffected by introduction of the feed. For this case, ideal solutions and a single phase feed, the feed stage criterion becomes

$$\begin{aligned}\dot{R}_s - \dot{R}_s' = &C_{f+1} \sum_{j=1}^{J} \chi_{j,L,f+1}\log_e(\chi_{j,L,f+1}/\chi_{j,L,f}) \\ &- C'_{f+1} \sum_{j=1}^{J} \chi'_{j,L,f+1}\log_e(\chi'_{j,L,f+1}/\chi'_{j,L,f}) \\ &+ \sum_{j=1}^{J} \chi_{j,F}\log_e(\chi_{j,F}/\chi_{j,L,f}) = \text{minimum}\end{aligned} \tag{12}$$

Equation (12) was applied to the problem of benzene-toluene-xylene separation (Robinson and Gilliland, 1950) in a stage-to-stage calculation with the feed a liquid at its boiling point. At each stage $\dot{R}_s - \dot{R}_s'$ was computed and compared with the value for the previous stage until the next stage showed an increase in value. The feed stage was thus found to be the tenth from the bottom, including the reboiler. The conditions assumed for the calculation were

Mol ratio bottoms/feed = 0.399
Mol ratio boil-up/feed = 1.803
Bottoms composition, mol fractions:
  benzene = 0.005
  toluene = 0.745
  xylene = 0.250

Lewis assumptions about overflow and vapor rates.

While this simple example does not constitute a complete test of the usefulness of the criterion, it does show that a thermodynamically sound, workable criterion can be applied.

LITERATURE CITED

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