

conductivities of packed beds containing various types of relatively large-size particles up to 840°C. Predicted thermal conductivities have been compared with the results of their experimental study in Figures 6 and 7. Good comparisons have been obtained, particularly when it is noted that at the highest temperature the radiation contribution accounts for about 80% of the total thermal conductivity.

CONCLUSIONS

Correlations have been developed for the effect of pressure, temperature, and particle size on the thermal conductivity of packed beds. An apparent thermal conductivity of the gas phase can be calculated from Equation (8) for use in Deissler and Eian's correlation, Figure 1, to predict the thermal conductivity of the bed. The correlation has been successfully checked with the data of various investigators. However it is not applicable to fine dusts, such as carbon black, because they contain particle agglomerates and have very large void fractions. For higher temperatures and relatively large particles a radiation contribution should be added. The radiation contribution can be calculated from Equations (16) and (17). Predicted values compared well with experimental data.

NOTATION

b = distance over which conduction takes place, ft.
 C, C_1 = empirical constants
 c_v = specific heat at constant volume, B.t.u./ (lb.) (°F.)
 c_p = specific heat at constant pressure, B.t.u./ (lb.) (°F.)
 D_p = particle diameter, ft.

h_r = heat transfer coefficient for radiation, B.t.u./ (hr.) (sq. ft.) (°F.)
 j = jump distance of temperature discontinuity, ft.
 k = thermal conductivity of packed bed, B.t.u./ (hr.) (sq. ft.) (°F./ft.)
 k_B = Boltzmann constant, 0.565×10^{-28} (ft.) (lb.) / °R.
 k_g = thermal conductivity of gas, B.t.u./ (hr.) (sq. ft.) (°F./ft.)
 k_g = apparent thermal conductivity of gas phase, B.t.u./ (hr.) (sq. ft.) (°F./ft.)
 k_p = contact thermal conductivity between particles, B.t.u./ (hr.) (sq. ft.) (°F./ft.)
 k_r = radiation contribution to thermal conductivity of powder, B.t.u./ (hr.) (sq. ft.) (°F./ft.)
 k_r = $h_r D_p$, B.t.u./ (hr.) (sq. ft.) (°F./ft.)
 k_s = thermal conductivity of solid, B.t.u./ (hr.) (sq. ft.) (°F./ft.)
 L = mean free path of molecules, ft.
 N_{Pr} = $c_p \mu / k_g$ = Prandtl number
 P = pressure, lb./sq. ft.
 P_b = break-away pressure, lb./sq. ft.
 q = rate of heat transfer, B.t.u./ hr.
 r = radius of particle, ft.
 s = element of particle perimeter, ft.
 t_1, t_2 = surface temperature of solid particle, °F.
 t_g = gas temperature, °F.
 T = absolute temperature, °R.
 x = coordinate in direction of heat flow, ft.
 z = coordinate perpendicular to direction of heat flow, ft.

Greek letters

α = accommodation coefficient
 γ = c_p / c_v = specific heat ratio
 δ = void fraction
 ϵ = emissivity
 μ = viscosity, lb./ (ft.) (hr.)
 σ = mean diameter of gas molecules, ft.

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Calculation of Equilibrium Flash Vaporization Curves by an Integration Method

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Techniques for solving the equilibrium flash problem are reviewed and a new technique especially adapted for use on automatic digital computers is developed. Instead of a single equation with multiple roots being solved by an iterative process, as is usually done, the problem is rephrased as a differential equation, and a numerical integration is made. The isothermal and isobaric flash problems can be handled with essentially the same equations. The method is particularly advantageous when a complete flash curve is required and the equations can be used when a linear model of the equilibrium flash process is required.

If a homogenous hydrocarbon mixture is subjected to a change in pres-

sure or temperature so that the resultant conditions are in the two-phase region, and if the two phases are in equilibrium, then the process is known

as an *equilibrium flash vaporization*. This paper deals with the problem of tracing the pressure-temperature-percentage-flashed history of a mixture of known over-all composition. The inverse problem of determining the phase compositions when an equilibrium flash curve is given is more difficult. Some of the methods of solving this latter

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problem have been reviewed and tested by Filak and Sandlin (1).

The method presented here for computing equilibrium flash curves uses a high-speed digital computer. In the following section the necessary equations are developed to express the flash problems as a differential equation relating percentage flashed, temperature, and pressure. This equation can then be numerically integrated to give a series of values of temperature (isobaric flash) or pressure (isothermal flash) vs. percentage flashed in increments of percentage flashed. The problem has been programmed for the Remington Rand Univac Scientific Model 1103 computer with the assumption that equilibrium vaporization ratios are available in the form

$$\ln K_i p = A_i + B_i T + C_i \left(\frac{1}{T} \right) +$$

$$D_i p + E_i p^2 + F_i \left(\frac{p}{T} \right) + G_i \frac{p^2}{T}$$

although other equations can be handled. The calculation of enthalpies is incorporated as an option in the program and gives an enthalpy-percentage-flashed curve (particularly useful in adiabatic flash calculations).

DERIVATIONS OF EQUATIONS

One mole of liquid feed containing n components is fed into an apparatus in which it is partially vaporized; the resulting liquid and vapor streams leave the apparatus in equilibrium at a known pressure and temperature. The following material-balance equations may be written, of which n are independent:

$$L + V = 1 \quad (1)$$

$$Lx_i + Vy_i = z_i \quad i = 1, 2, \dots, n \quad (2)$$

These are rearranged and combined with the equilibrium relationship

$$y_i = K_i x_i \quad (3)$$

to give

$$x_i = \frac{z_i}{V(K_i - 1) + 1} \quad i = 1, 2, \dots, n \quad (4)$$

One common procedure is to use the requirement that

$$\sum_{i=1}^n x_i = 1.0$$

and so

$$\sum_{i=1}^n \frac{z_i}{V(K_i - 1) + 1} = 1.0 \quad (5)$$

If the criterion $\sum_{i=1}^n y_i = 1.0$ is used,

$$\sum_{i=1}^n \frac{K_i z_i}{V(K_i - 1) + 1} = 1.0 \quad (6)$$

$$-dx_i = \frac{z_i (K_i - 1)}{[V(K_i - 1) + 1]^2} dV + \frac{z_i V \left(\frac{\partial K_i}{\partial T} \right)_p}{[V(K_i - 1) + 1]^2} dT + \frac{z_i V \left(\frac{\partial K_i}{\partial p} \right)_T}{[V(K_i - 1) + 1]^2} dp \quad (10)$$

Rachford (10) suggested that it is convenient to combine Equations (5) and

suitable material-balance relations, one may convert Equation (9) into a form involving only p , T , V , x_i , K_i , and z_i . If the equation is solved for dx_i ,

and, since $\sum_{i=1}^n dx_i = 0$

$$\sum_{i=1}^n \frac{(K_i - 1) z_i dV}{[1 + V(K_i - 1)]^2} = - \sum_{i=1}^n \frac{V z_i \left(\frac{\partial K_i}{\partial T} \right)_p dT}{[1 + V(K_i - 1)]^2} - \sum_{i=1}^n \frac{V z_i \left(\frac{\partial K_i}{\partial p} \right)_T dp}{[1 + V(K_i - 1)]^2} \quad (11)$$

(6) to give

$$\phi(V, T) = \sum_{i=1}^n \frac{(K_i - 1) z_i}{V(K_i - 1) + 1} = 0 \quad (7)$$

Alternately, x_i may be eliminated and the criterion $\sum_{i=1}^n dy_i = 0$ employed to give

$$\sum_{i=1}^n \frac{K_i (K_i - 1) z_i dV}{[1 + V(K_i - 1)]^2} = \sum_{i=1}^n \frac{(1 - V) z_i \left(\frac{\partial K_i}{\partial T} \right)_p dT}{[1 + V(K_i - 1)]^2} + \sum_{i=1}^n \frac{(1 - V) z_i \left(\frac{\partial K_i}{\partial p} \right)_T dp}{[1 + V(K_i - 1)]^2} \quad (12)$$

Montross (7) has presented a method which can be reduced to the equation

Finally, subtracting Equation (11) from (12) gives

$$\sum_{i=1}^n \frac{(K_i - 1)^2 z_i dV}{[1 + V(K_i - 1)]^2} = \sum_{i=1}^n \frac{z_i \left(\frac{\partial K_i}{\partial T} \right)_p dT}{[1 + V(K_i - 1)]^2} + \sum_{i=1}^n \frac{z_i \left(\frac{\partial K_i}{\partial p} \right)_T dp}{[1 + V(K_i - 1)]^2} \quad (13)$$

$$\sum_{i=1}^n \frac{V z_i (K_i - 1)}{V(K_i - 1) + 1} = 0 \quad (8)$$

which is equivalent to Equation (7). Several useful rules of thumb are given by Montross to facilitate making the calculations involved by hand.

The use of any of Equations (5) to (8), given the K_i at the equilibrium conditions, involves a trial-and-error solution; values of V are assumed between 0 and 1.0 until the criterion is met, for instance $\phi = 0$ in Rachford's method [Equation (7)]. A systematic procedure for these calculations is available (10), and the method has further been elaborated by Organick and Meyer (9).

The above methods give V , x_i , and y_i for a certain feed composition, temperature, and pressure. In order to obtain a flash curve of temperature vs. fraction vaporized, it is necessary to repeat the calculations at an arbitrary number of temperatures between the bubble and dew points. The following procedure will give the entire flash curve by a single numerical integration.

If a differential amount of liquid is vaporized, Equation (3) becomes

$$d(Lx_i) + d(Vy_i) = 0 \quad (9)$$

By using the equilibrium relations and

and at constant pressure

$$\left(\frac{\partial T}{\partial V} \right)_p = \frac{\sum_{i=1}^n \frac{(K_i - 1)^2 z_i}{[1 + V(K_i - 1)]^2}}{\sum_{i=1}^n \frac{z_i \left(\frac{\partial K_i}{\partial T} \right)_p}{[1 + V(K_i - 1)]^2}} = f(V, T) \quad (14)$$

At constant pressure Equation (14) is an ordinary, linear, first-order differential equation subject to the initial conditions that $T = T_b$ when $V = 0$. The equation may be integrated to give

$$T = T_b + \int_0^V f(V, T) dV \quad (15)$$

A check on the integration, which must be done numerically, is given by the requirement that at $V = 1$, $T = T_d$.

It is easy to verify that Equation (14) can also be found from Equation (7) by the relation

$$\left(\frac{\partial T}{\partial V} \right)_p = - \frac{\left(\frac{\partial \phi}{\partial V} \right)_{p,T}}{\left(\frac{\partial \phi}{\partial T} \right)_{p,V}} \quad (16)$$

when one recalls that $\left(\frac{\partial T}{\partial V} \right)_p$ is evaluated along a path of $\phi = 0$.

TABLE 1. CONSTANTS FOR CALCULATION OF EQUILIBRIUM-VAPORIZATION RATIOS

Component	$K_i = \frac{1}{p} \exp \left(A_i + B_i T + C_i \frac{1}{T} + D_i p + E_i p^2 + F_i \frac{p}{T} + G_i \frac{p^2}{T} \right)$						
	A	B	C	D	E	F	G
Multi- ply by	($\times 10^1$)	($\times 10^{-8}$)	($\times 10^8$)	($\times 10^{-8}$)	($\times 10^{-6}$)	($\times 10^0$)	($\times 10^{-4}$)
C ₈	2.0336	-5.039	-10.055	-2.03	4.48	3.6876	-42.25
C ₇	2.0047	-4.980	-9.4402	-1.596	2.659	2.9829	-21.263
C ₆	1.8610	-4.287	-8.1754	-1.334	0.6002	2.0253	0.739
nC ₆	1.6789	-3.448	-6.8436	-1.493	1.263	2.0316	-8.974
iC ₆	1.5087	-2.341	-6.0943	-2.095	1.496	2.3574	-9.493
nC ₅	1.4169	-1.879	-5.2275	-1.873	-0.02911	2.2136	-3.358
iC ₅	1.2931	-1.058	-4.610	-1.428	1.80	1.6756	-9.531
C ₅	1.3137	-1.43	-4.117	-1.55	2.90	1.663	-25.02
C ₄	1.2535	-1.03	-3.168	4.28	-8.795	-3.607	76.51
C ₃	1.0173	0.623	-1.436	1.89	-3.46	-1.880	34.23

In the preceding derivations it is assumed that K_i is a function of p and T only, that is, that the solutions are ideal. This requirement restricts the use of the method to pressures low enough so that K_i is not affected by composition, below 400 to 500 lb./sq. in. abs. for most mixtures. The K_i values used were obtained (8) and correlated by the use of

$$\ln K_i p = A_i + B_i T + C_i \left(\frac{1}{T} \right) + D_i p + E_i p^2 + F_i \left(\frac{p}{T} \right) + G_i \frac{p^2}{T} \quad (17)$$

This form is especially convenient for digital computation and represents the data well. Values for the constants are given in Table 1.

An isothermal flash curve can be obtained from Equation (13):

$$\left(\frac{\partial p}{\partial V} \right)_T = \frac{\sum_{i=1}^n \frac{(K_i - 1)^2 z_i}{[1 + V(K_i - 1)]^2}}{\sum_{i=1}^n \frac{z_i \left(\frac{\partial K_i}{\partial p} \right)_T}{[1 + V(K_i - 1)]^2}} = g(V, p) \quad (18)$$

and similarly the isometric equation is

$$-\left(\frac{\partial p}{\partial T} \right)_V = \frac{\sum_{i=1}^n \frac{z_i \left(\frac{\partial K_i}{\partial T} \right)}{[1 + V(K_i - 1)]^2}}{\sum_{i=1}^n \frac{z_i \left(\frac{\partial K_i}{\partial p} \right)_T}{[1 + V(K_i - 1)]^2}} \quad (19)$$

INTEGRATION PROCEDURE

Equation (15) may be written over an interval, ΔV , as

$$T(V + \Delta V) = T(V) + \int_V^{V+\Delta V} T'(V) dV \quad (20)$$

when

$$T'(V) = f(V, T) \quad (21)$$

It has been suggested that the integral in Equation (20) can be accurately evaluated numerically by Gaussian quadrature (2). This integration leads to

$$T(V + \Delta V) = T(V) + \frac{\Delta V}{18} \left\{ 5T' \left[V + \frac{\Delta V}{2} \left(1 - \frac{\sqrt{15}}{5} \right) \right] + 8T' \left[V + \frac{\Delta V}{2} \right] + 5T' \left[V + \frac{\Delta V}{2} \left(1 + \frac{\sqrt{15}}{5} \right) \right] \right\} \quad (22)$$

The Runge-Kutta method of fourth-order accuracy (4) can be used three times to give the values of T at $[V + \Delta V/2(1 - \sqrt{15}/5)]$, $[V + \Delta V/2]$, and $[V + \Delta V/2(1 + \sqrt{15}/5)]$. $T'(V)$ is then evaluated at each of these points to obtain $T(V + \Delta V)$ from Equation (22). For certain cases tested (2), this procedure had an error about one-tenth that of the Runge-Kutta-Gill method (3). It is a self-starting method, in contrast to the various Adams procedures (5).

For the equilibrium flash problem the interval from $V = 0$ to $V = 1$ is divided into $1/\Delta V$ increments. A check is made after the first integration, and if $\sum_{i=1}^n x_i$ at $V = 1.0$ is not equal to $1.0 \pm \epsilon$, ΔV is halved and the process repeated until the criterion is met, on the assumption that ϵ is not chosen to be so small that round-off errors become important.

ENTHALPY BALANCE AND BUBBLE-POINT CALCULATION

The enthalpy balance for a flow process can be written as

$$h_f + Q = (1 - V)h + VH \quad (23)$$

where

$$h = \sum_{i=1}^n x_i h_i \quad (24)$$

$$h_f = \sum_{i=1}^n z_i h_{fi} \quad (25)$$

$$H = \sum_{i=1}^n y_i H_i \quad (26)$$

Both h_i and H_i are considered here as functions of temperature only (6). For a given feed h_f is calculated by Equation (25); then for the conditions in the flash chamber h and H are calculated by Equations (24) and (26). Equation (23) is thus solved for Q , so that a curve of Q vs. V is obtained. The temperature and fraction vaporized in an adiabatic flash are obtained from the T vs. V curve and the value of V where $Q = 0$.

Since the bubble point is needed as the initial point of the integration, a word on the calculation of the bubble point is included. At this condition the following criterion must be satisfied:

$$\sum_{i=1}^n K_i z_i = 1.0$$

For a given pressure the bubble-point temperature can be found by the use of a Newton-Raphson iteration as follows:

$$T_{Bj+1} = T_{Bj} + \frac{- \left[\left(\sum_{i=1}^n K_i z_i \right)_j - 1 \right]}{\left[\sum_{i=1}^n z_i \left(\frac{\partial K_i}{\partial T} \right)_p \right]} \quad (27)$$

Setting $j = 0$, one can estimate an initial value of the bubble point and calculate T_{B1} by Equation (27). This process is then repeated, and j is advanced each time until T_{Bj+1} is arbitrarily close to T_{Bj} .

DISCUSSION OF SAMPLE PROBLEMS

Besides those mentioned the following computations can be obtained if desired:

1. Value of Q at each integration interval.
2. The $\sum x_i$ and $\sum y_i$ at each integration interval.
3. All values of x_i and y_i at each integration interval.
4. Values of V and T and dt/dV at the three Runge-Kutta points in each interval for the isobaric flash or V , p ,

TABLE 2. RESULTS OF SAMPLE ISOBARIC AND ISOTHERMAL EQUILIBRIUM FLASH CALCULATIONS

Type of flash	Mixture	Number of intervals	Dew point		Σx_i
			Actual	By integration	
Isobaric	A*	1	771.799 °R.	767.261 °R.	1.03918
Isobaric	A	2	771.799 °R.	770.669 °R.	1.00955
Isobaric	A	4	771.799 °R.	771.624 °R.	1.00147
Isobaric	A	8	771.799 °R.	771.788 °R.	1.00010
Isobaric	B†	1	740.359 °R.	740.909 °R.	0.99508
Isobaric	B	2	740.359 °R.	740.345 °R.	1.00013
Isothermal	B	1	492.24 lb./sq. in. abs.	474.13 °R.	0.95407
			492.24 lb./sq. in. abs.	499.77 °R.	1.00096
Isothermal	B	4	492.24 lb./sq. in. abs.	499.36 °R.	1.00022
			492.24 lb./sq. in. abs.		

* Mixture A — C₁, 1%; C₂, 5%; C₃, 11%; iC₄, 6%; nC₄, 14%; iC₅, 21%; nC₅, 20%; C₆, 10%; C₇, 9%; C₈, 3%.

† Mixture B — C₂, 35%; C₃, 10%; nC₄, 10%; nC₅, 10%; C₆, 35%.

and dp/dV for the isothermal flash. This information is useful when only a few intervals are used or when interpolation between points is desired.

The flash curve is computed by the use of four intervals ($\Delta V = 1/4$). After the curve has been computed, it can be repeated with half as large an interval. This procedure can be repeated any number of times.

To illustrate the application of this method, several sample problems were solved for two different feed compositions and with different numbers of integration intervals. The results are summarized in Table 2. It was seen in the preceding sections that an integration is started at the bubble point and can be continued until the dew point is reached. The greatest error normally occurs at the dew point. The simplest test of the accuracy of the method is to compare the value calculated by the integration method with that computed directly from the K values. These values are given in the fourth and fifth columns of Table 2, and it can be seen that, with the exception of the one-interval integration with ten components, the errors are small. As might be expected, the ten-component problem was less accurate for the same number of intervals than the five-component problem. The error decreases rapidly with increase in the number of intervals. On the basis of these results four intervals were judged sufficiently accurate in all cases examined.

Another measure of errors is the Σx_i at the dew point, listed in the last column of Table 2, and the results are consistent with the conclusions based on the dew-point comparisons. The error for the one-interval case is 3.9%, decreasing to 0.009% in the eight-interval integration.

APPLICATION OF METHOD

The method described in the previous section was developed primarily to generate a complete equilibrium flash curve. Such a curve might be of interest in itself; however a more probable application would be to use the program to generate a table which would be stored in the computer. Thus a problem calling for many references to an equilibrium flash curve would obtain them by interpolation in the table. For example, the four-interval integration used in the program would give a total of fifteen temperature-volume flashed-composition points, in addition to the bubble and dew points. Also the slopes of the flash curve would be known at these points. Interpolation between these points with the slopes known would be accurate and many times faster than computing the points directly.

In addition, the equations developed in this paper can be used to reduce the highly nonlinear equilibrium flash process to a linear process in the neighborhood of some point. This linearization technique is a highly useful simplification in many problems. The analysis of control systems, for example, is limited largely to linear systems, and it is usually necessary to describe the system in terms of linear perturbations about some point. Similarly some economic analysis techniques, such as linear programming, depend on the use of linear models. Obviously the equilibrium flash process can be described in the neighborhood of a point by the equilibrium values of temperature, pressure, composition, and the appropriate slopes as obtained from Equations (10) through (19).

Although the techniques described in this paper could be used for the computation of a single point on the equilibrium flash curve, it does not

seem likely that this would be desirable, since the conventional methods are faster if only one point is required.

In summary, then, these techniques seem to be best adapted to the machine computation of entire equilibrium flash curves, particularly when interpolation between points is desired. The equations are also important when a linear model of the equilibrium flash process is desired.

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NOTATION

h_i	= enthalpy of liquid component i , B.t.u./lb. mole
h_f	= enthalpy of liquid mixture before flashing, B.t.u./lb. mole
H_i	= enthalpy of vapor component i , B.t.u./lb. mole
K_i	= y_i/x_i at equilibrium
L	= moles of liquid per mole of feed
p	= pressure, lb./sq. in. abs.
Q	= heat added during flash, B.t.u./lb. mole of feed
T	= absolute temperature, °R.
T_{b0}	= initial value of bubble point
V	= moles of vapor per mole of feed
ΔV	= integration interval
x_i	= mole fraction component i in liquid
y_i	= mole fraction component i in vapor
z_i	= mole fraction component i in mixture prior to flashing

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