Thermal Conductivity of Packed Beds

WILLIAM SCHOTTE

E. I. du Pont de Nemours and Company, Wilmington, Delaware

A correlation has been developed to predict the thermal conductivity of packed beds for various conditions of pressure, temperature, and particle size. The correlation takes into account the reduction in thermal conductivity of the gas phase at lower pressures when the mean free path of the gas molecules is of the same order as the distance between particles which are effective in transferring heat. Radiation becomes significant for large particles and high temperatures. A new relationship for the radiation contribution to the thermal conductivity of packed beds has been developed.

Packed beds containing refractory materials are often used as insulation to minimize heat losses from furnaces and reactors. A number of correlations have been reported in the literature to predict the thermal conductivity of packed beds. Although the conduction process through such a two-phase system with undefined internal boundaries is extremely complex, simple correlations have been quite successful. Generally the thermal conductivity of the bed is expressed as a function of the thermal conductivity of the gas, the thermal conductivity of the solid, and the void fraction of the bed. The influence of particle size, pressure, and temperature has not been accounted for except insofar as they affect the primary variables mentioned above.

It has been recognized, however, that packed beds may have much lower thermal conductivities than would normally be predicted. When the effective gas spaces have dimensions which are of the same order as the mean free path of the gas molecules, the thermal conductivity of the gas phase is reduced. Since most of the conduction takes place near points of contact between particles, this reduction starts to occur when the particle diameter is about one thousand times the length of the mean free path. Deissler and Eian (*3*) have measured so-called "break-away pressures," below which the thermal conductivity of the packed bed will decrease with a decrease in pressure. They have also developed a correlation to predict the break-away pressure for a particular system. The break-away pressure may be quite high; for example Deissler and Eian have shown that a value of 41 lb./sq. in. abs. is obtained for 0.2 mm. magnesium oxide particles and helium at 400°F. However methods for the prediction of the thermal conductivity at pressures below the break-away value were not found in the literature. A relationship which correlates available data has been developed and is presented in this paper.

At high temperatures radiation heat transfer between particles becomes important. Damköhler (*2*) analyzed the contribution due to radiation but did not suggest a definite equation for its effect. Later workers have interpreted Damköhler's analysis in various ways to arrive at definite relationships. For instance McAdams (*8*) suggests adding the following radiation contribution to the thermal conductivity of the packed bed:

$$k_r = 0.692\,\delta\, D_p \frac{T^3}{10^8} \qquad (1)$$

Other correlations are similar but include different factors to account for the emissivity of the particles. Only recently have some data become available in the literature to evaluate the contribution due to radiation. The work of Yagi and Kunii (*14*) has shown that Equation (1) cannot be used. They have developed a single relationship to account for all modes of heat transfer which contribute to the conductivity of packed beds, but this correlation does not give satisfactory results when the apparent thermal conductivity of the gas phase is used for the case of small particles. A new correlation for the radiation contribution is presented in this paper.

The case of fine dusts with void fractions above 0.6, such as carbon black, has not been considered here for reasons which will be discussed later.

APPARENT THERMAL CONDUCTIVITY OF THE GAS PHASE

It has been known for a long time that the thermal conductivity of a gas is reduced when low pressures are reached. This occurs when the distance over which the conduction takes place becomes of the same order as the mean free path of the gas molecules. Since molecules can cross from one surface to another before reaching thermal equilibrium, there is an abrupt change of temperature from the surface to the gas instead of the gradual transition which occurs at higher pressures. Many authors (for example, reference *6*) have related the apparent thermal conductivity of the gas, to the normal thermal conductivity with the following expression:

$$k_g = \frac{k_g^0}{1 + 2\dfrac{j}{b}} \qquad (2)$$

Kinetic theory (*6*) gives also the following equation for j:

$$j = \left(\frac{2-\alpha}{\alpha}\right)\left(\frac{2}{1+\gamma}\right)\left(\frac{k_g^0 L}{\mu c_v}\right) \qquad (3)$$

which can be rewritten to give

$$j = \left(\frac{2-\alpha}{\alpha}\right)\left(\frac{2\gamma}{1+\gamma}\right)\left(\frac{k_B T}{\pi\sqrt{2}P\sigma^2 N_{Pr}}\right) = 2.54\times 10^{-24}\left(\frac{2-\alpha}{\alpha}\right)\left(\frac{\gamma}{1+\gamma}\right)\left(\frac{T}{P\sigma^2 N_{Pr}}\right) \qquad (4)$$

An equation for the apparent thermal conductivity of the gas phase is obtained by the substitution of Equation (4) into Equation (2):

$$k_g = \frac{k_g^0}{1+5.08\times 10^{-24}\left(\dfrac{2-\alpha}{\alpha}\right)\left(\dfrac{\gamma}{1+\gamma}\right)\left(\dfrac{T}{Pb\sigma^2 N_{Pr}}\right)} \qquad (5)$$

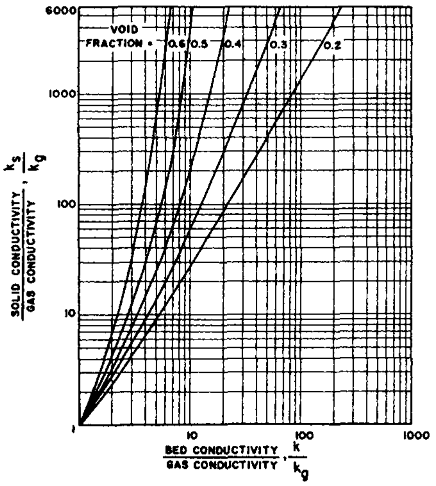

Fig. 1. Deissler-Eian correlation for the thermal conductivity of packed beds.

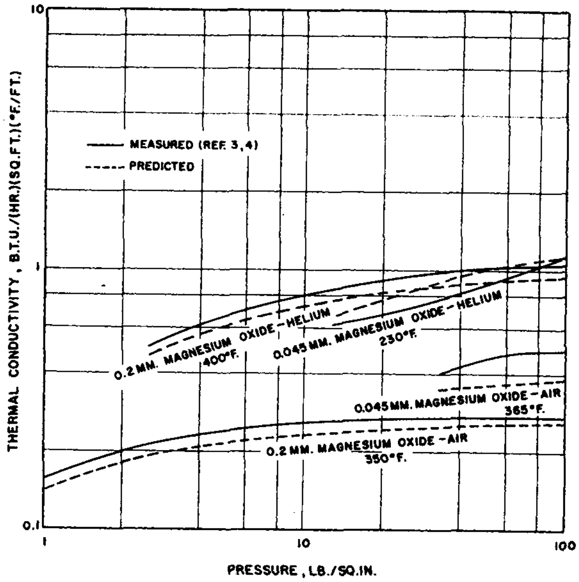

Fig. 2. Effect of pressure on the thermal conductivity of packed beds.

A useful solution of Equation (5) will require a knowledge of the accommodation coefficient and the effective distance over which the conduction takes place. Only few experimental values of the accommodation coefficient are available, and even for a given solid-gas system values differ widely depending on the condition of the surface. An exact relationship for the effective distance is also difficult to determine, since most of the conduction will take place near points of contact between the particles. However the success of Deissler and Eian in predicting break-away pressures indicates that an empirical constant may be used to include the accommodation coefficient and a proportionality factor for the evaluation of b. Actually b is equal to the heat transfer-average distance between particle surfaces. As a simplification however it will be assumed that b is proportional to the length-average distance between particle surfaces.

If one considers spherical particles in a unit cube, the number of particles within the cube will be $(1-\delta)/(\pi/6\,D_p^3)$. In a given direction these particles will present a total interfering area which is equal to the sum of their cross-sectional areas:

$$\left(\frac{\pi}{4}D_p^2\right)\left(\frac{1-\delta}{\frac{\pi}{6}D_p^3}\right) = 1.5\left(\frac{1-\delta}{D_p}\right)$$

The average interparticle distance is equal to the free volume divided by the total interfering area:

$$\frac{2}{3}\left(\frac{\delta}{1-\delta}\right)D_p$$

Then

$$b = C_1\left(\frac{\delta}{1-\delta}\right)D_p \qquad (6)$$

The final expression for the apparent thermal conductivity is obtained by the substitution of Equation (6) into (5) and by the inclusion of the term $[(2-\alpha)/\alpha]$ in C:

$$k_g = \frac{k_g^\circ}{1 + C\left(\frac{\gamma}{1+\gamma}\right)\left(\frac{1-\delta}{\delta}\right)\left(\frac{T}{PD_p\sigma^2N_{Pr}}\right)} \qquad (7)$$

A value of $C = 2.03 \times 10^{-22}$ has been determined by a comparison of the results from Equation (7) with experimental data of a number of investigators. This will be discussed below.

THERMAL CONDUCTIVITY OF A PACKED BED

A relationship is needed to predict the thermal conductivity of the packed bed. Various methods have been proposed in the literature. The most suitable one appears to be that of Deissler and Eian (3), who have presented the semiempirical relationship shown in Figure 1. The ratio of the thermal conductivity of the bed to that of the gas phase is plotted against the ratio of the thermal conductivity of the solid to that of the gas phase with the void fraction used as a parameter.

The accuracy of the correlation of Deissler and Eian, or those of other workers, is difficult to ascertain. It appears that many of the experimental data reported in the literature are somewhat inaccurate; for example, data for the system glass spheres—air at nearly the same temperature and with almost the same void fraction differ widely, as shown in Table 1. The data differ by as much as a factor of 2.4, and as a result some data which appeared to be greatly in error were not considered in this study.

TABLE 1. COMPARISON OF DATA FOR GLASS-AIR SYSTEM*

Reference	t, °C.	D_p, mm.	δ	k/k_g
(5)	10	0.32	0.35	6.62
(7)	60	4.06	0.40	12.0
(12)	80	6.77	0.40	5.35
	80	10.0	0.40	5.0
(9)	40	0.34	0.40	9.4

* All measurements made with glass spheres and at atmospheric pressure.

Deissler and Eian's correlation (3) for the break-away pressure

$$P_b = 1.77 \times 10^{-21}\frac{T}{D_p\sigma^2}$$

can be used to determine whether pressure, temperature, or particle size will affect the gas-phase thermal conductivity. If the pressure is below the break-away pressure, the gas-phase thermal conductivity should be calculated by the use of Equation (8):

$$k_g = \frac{k_g^\circ}{1+2.03\times10^{-22}\left(\frac{\gamma}{1+\gamma}\right)\left(\frac{1-\delta}{\delta}\right)\left(\frac{T}{PD_p\sigma^2N_{Pr}}\right)} \qquad (8)$$

This value of k_g is used to calculate the ratio of k_s to k_g at the average bed temperature. Figure 1 then gives the ratio of k to k_g from which the bed conductivity can be determined. This procedure, but in the reverse order, was applied to calculate values of the apparent gas-phase conductivity from experimental data for the thermal conductivity of packed beds in order to arrive at values for C in Equation (7). Values of C varied by a factor of 2 possibly owing to variations in the accommodation coefficient but perhaps to an even greater extent to inaccuracies of the experimental data and to the approximations in the method of Deissler and Eian.

COMPARISON WITH EXPERIMENTAL DATA

Thermal conductivities of packed beds at different pressures have been measured by a number of investigators. For the same systems Equation (8) and Figure 1 were used to obtain predicted thermal conductivities. The results are compared in Figures 2 to 4. In addition, predicted results have been compared with data of Schumann and Voss (*11*) and have been deposited with the American Documentation Institute.*

Predicted and experimental curves compare quite well, particularly if one considers the curves on a relative rather than an absolute basis. Deviations are about as large at higher pressures, where the apparent gas-phase conductivity is nearly equal to the normal thermal conductivity of the gas, as at lower pressures. It is therefore believed that the differences are caused mainly by inaccuracies of the experimental data and by the simplified approximation of Deissler and Eian.

Theoretically there may be no lower limit for application of Equation (8). It has been reported in the literature that at very low pressures the thermal conductivity of the gas phase is proportional to the pressure. Equation (8) shows this also, since the first term, 1, in the denominator on the right-hand side of the equation becomes negligible compared with the second term at low pressures. In practice however there are limits to the use of Equation (8) and Figure 1 for the prediction of thermal conductivities of packed beds. It is therefore not advisable to extend Figure 1 beyond its limits, that is outside of $0.2 < \delta < 0.6$ or beyond $k_s/k_g = 6{,}000$. Although Deissler and Eian have included the whole range of void fractions from 0 to 1 in their correlation, the experimental values for which the correlation was tested fell in the range $0.2 < \delta < 0.6$. Fine dusts have large void fractions, up to values of 0.98, but known correlations do not apply. This is at least partly due to the formation of agglomerates. For very large values of k_s/k_g, solid conduction at contact points becomes significant. This has not been accounted for in Deissler and Eian's correlation. Wilhelm (*13*) has presented a correlation for this contact conductivity:

$$\log k_p = -1.76 + 0.0129 \frac{k_s}{\delta}$$

Since this correlation does not account for the particle size, the thermal conductivity contribution due to contact conductivity is the same for larger and smaller particles; however more contact resistances must be added in series for smaller particles to obtain the conductivity over a unit distance. As a result calculations can show that predicted values may be an order of magnitude larger than experimental values for very small particles. Development of a new correlation for the contact conductivity will be difficult until more data become available.

EFFECT OF RADIATION

Radiation heat transfer between particles should be included at high temperatures, particularly when the particles are larger. For example radiation becomes important for 1 mm. particles at temperatures above 400°C. and for 0.1-mm. particles above 1,500°C. A simplified analysis can be made for spherical particles, but experimental data indicate that the final correlation will apply also to other shapes.

The derivation presented below will be limited to opaque particles which are large compared with the wave length of radiation. Transparency of the particles would increase the rate of heat transfer and therefore the thermal conductivity of the bed. When the particles are in the micron range, scattering of radiation should be considered and the absorption of radiation is more complicated.

The analysis given here is somewhat similar to that used by Argo and Smith (*1*), but a different interpretation has led to an improved correlation. When one considers the radiation from a plane located on one side of a particle to a plane located on the far side of the

* Tabular material has been deposited as document 6137 with the American Documentation Institute, Photoduplication Service, Library of Congress, Washington 25, D. C., and may be obtained for $1.25 for photoprints or 35-mm. microfilm.

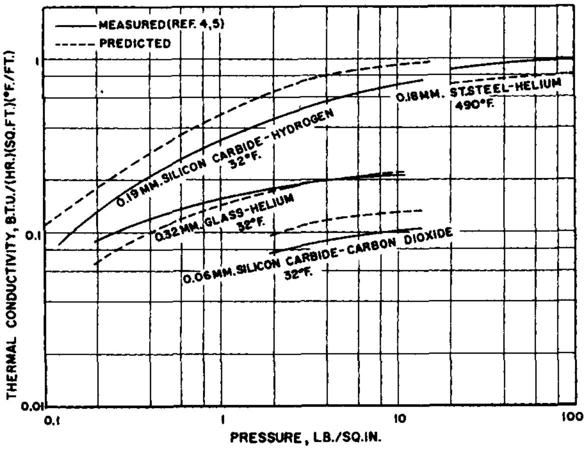

Fig. 3. Effect of pressure on the thermal conductivity of packed beds.

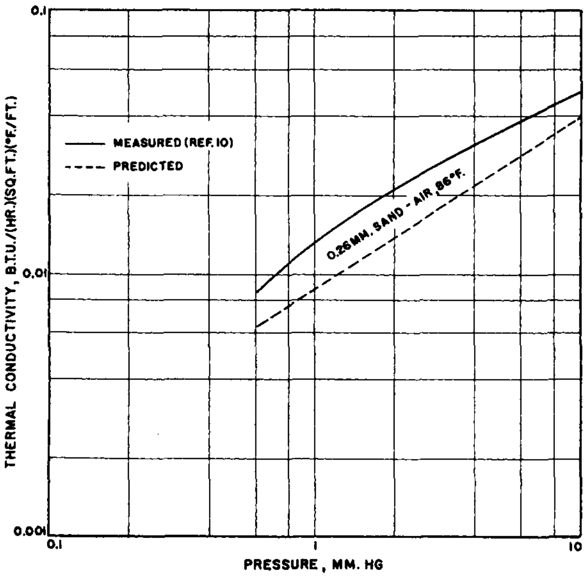

Fig. 4. Effect of pressure on the thermal conductivity of packed beds.

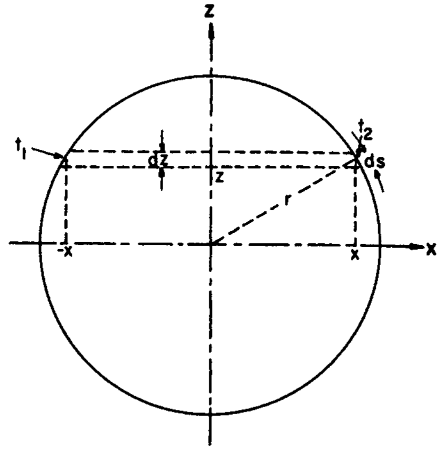

Fig. 5. Model for derivation of the radiation contribution to the thermal conductivity of a packed bed.

particle, there are two parts to be included. First, there is radiation across the void space past the particle. For a particle of cross-sectional area $\left(\frac{\pi}{4}D_p^2\right)$, the total area for heat transfer can be approximated by

$$\frac{\pi}{4}D_p^2\left(\frac{1}{1-\delta}\right) \qquad (9)$$

with a void area of

$$\frac{\pi}{4}D_p^2\frac{\delta}{1-\delta}$$

The radiation heat transfer across the void area will then be

$$q=-h_r\left(\frac{\pi}{4}D_p^2\frac{\delta}{1-\delta}\right)\left(D_p\frac{dt}{dx}\right) \qquad (10)$$

where $[D_p\,(dt/dx)]$ is the temperature difference between the planes previously mentioned.

Secondly, there is radiation from the particle in series with conduction through the particle. For an element of the particle, shown in Figure 5, the rate of heat transfer from left to right will be

$$dq=-k_s\,(2\pi z dz)\,\frac{t_s-t_1}{2x}=$$

$$h_r\,(2\pi z ds)\,(t_2-t_g) \qquad (11)$$

$$=h_r\,(2\pi z)\left(\frac{r dz}{x}\right)\,(t_2-t_g)$$

or $k_s\,(t_1-t_2)=2\,h_r r\,(t_2-t_g)$, which by elimination of t_2 gives

$$t_2=\frac{k_s t_1+2\,h_r r t_g}{k_s+2\,h_r r}$$

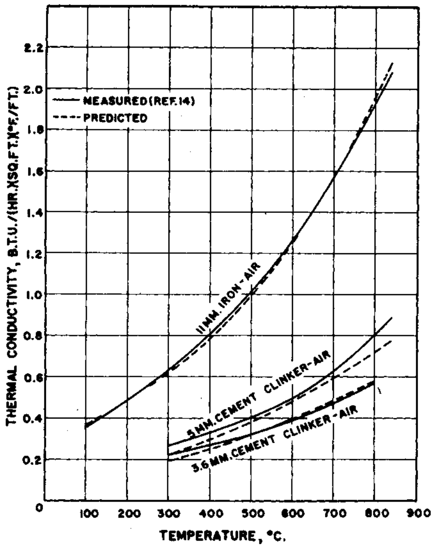

Fig. 6. Effect of radiation on the thermal conductivities of packed beds.

Substitution of this expression for t_2 into Equation (11) gives

$$dq=k_s\left(\frac{\pi z dz}{x}\right)\left(t_1-\frac{k_s t_1+2h_r r t_g}{k_s+2h_r r}\right)$$

$$=k_s\left(\frac{\pi z dz}{x}\right)\frac{h_r D_p\,(t_1-t_g)}{k_s+h_r D_p} \qquad (12)$$

where $2r$ has been replaced by D_p. For a simplified approximation it will be assumed that the temperature gradient, dt/dx, for the solid-gas system (packed bed) is constant for a distance of 1 particle diameter. Then

$$t_1-t_g=-2x\left(\frac{dt}{dx}\right)$$

and Equation (12) becomes

$$dq=-k_s\,(2\pi\, z dz)\left(\frac{h_r D_p}{k_s+h_r D_p}\right)\left(\frac{dt}{dx}\right)$$

This expression can be integrated over the whole particle:

$$q=-k_s\,(\pi r^2)\left(\frac{h_r D_p}{k_s+h_r D_p}\right)\left(\frac{dt}{dx}\right)$$

or

$$q=-\frac{\pi}{4}D_p^2\left(\frac{k_s h_r D_p}{k_s+h_r D_p}\right)\left(\frac{dt}{dx}\right) \qquad (13)$$

Adding the two parts of radiation heat transfer, Equations (10) and (13), one gets

$$q=-\frac{\pi}{4}D_p^2\left(\frac{k_s h_r D_p}{k_s+h_r D_p}\right)\left(\frac{dt}{dx}\right)-h_r\left(\frac{\pi}{4}D_p^2\frac{\delta}{1-\delta}\right)D_p\frac{dt}{dx} \qquad (14)$$

To determine the thermal-conductivity correction for radiation heat transfer the rate of heat transfer across the total area [see Equation (9)] can be written as

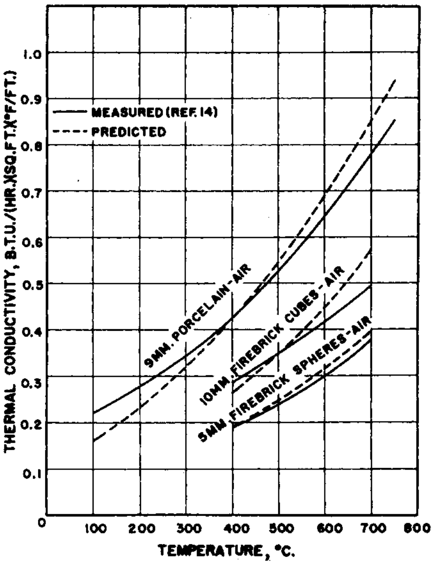

Fig. 7. Effect of radiation on the thermal conductivity of packed beds.

$$q=-k_r\left(\frac{\pi}{4}D_p^2\frac{1}{1-\delta}\right)\frac{dt}{dx} \qquad (15)$$

The radiation contribution is found by equating the right-hand sides of Equations (14) and (15).

$$k_r=\frac{1-\delta}{\frac{1}{k_s}+\frac{1}{k_r^\circ}}+\delta k_r^\circ \qquad (16)$$

where $k_r^\circ=h_r D_p$. The right-hand side of Equation (16) consists of two terms. The first term represents radiation heat transfer to the particle in series with conduction through the particle, while the second term accounts for radiation across the void space adjacent to the particle.

The radiation heat transfer coefficient between a particle and its neighbors can be approximated by

$$h_r=0.692\epsilon\frac{T^3}{10^8}$$

This approximation can be made because the angle factor is unity and the emissivity factor is close to the emissivity of the particle, since the particle under consideration is surrounded by the irregular surfare of the packed bed. Then

$$k_r^\circ=0.692\epsilon\, D_p\frac{T^3}{10^8} \qquad (17)$$

To evaluate the thermal conductivity of a packed bed it is first predicted from the correlation in Figure 1 [with Equation (8) used, if necessary], and then the radiation contribution from Equation (16) is added.

High-temperature data for which the correlation can be tested have recently become available. Yagi and Kunii (*14*) have measured thermal 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^{-23} (ft.)(lb.)/°R.
k_g^0 = 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^0 = $h_r D_p$, 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} = $c_p\mu/k_g^0$ = 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.

LITERATURE CITED

1. Argo, W. B., and J. M. Smith, *Chem. Eng. Progr.*, **49**, 443 (1953).
2. Damköhler, G., "Der Chemie Ingenieur," Eucken-Jakob, Vol. 3, Part 1, p. 445, Leipzig, Akademische Verlagsgesellschaft M.B.H. (1937).
3. Deissler, R. G., and C. S. Eian, *Natl. Advisory Comm. Aeronaut., RM E52C05*, (1952).
4. *Ibid., RM E53G03* (1953).
5. Kannuluik, W. G., and L. H. Martin, *Proc. Roy Soc. (London)*, **A141**, 144 (1933).
6. Kennard, E. H., "Kinetic Theory of Gases," p. 311, McGraw-Hill, New York (1938).
7. Kling, G., *Forsch. Geb. Ingenieurw.*, **9A**, 82 (1938).
8. McAdams, W. H., "Heat Transmission," 3ed., McGraw-Hill, New York (1954).
9. Prins, J. A., J. Schenk, and A. J. G. L. Schram, *Physica*, **16**, 379 (1950).
10. Ribaud, G., *Chaleur & ind.*, **18**, 36 (1937).
11. Schumann, T. E. W., and V. Voss, *Fuel*, **13**, 249 (1934).
12. Sugawara, S., and S. Sato, *Trans. Soc. Mech. Engrs. (Japan)*, **13**, No. 45, 164 (1947).
13. Wilhelm, R. H., W. C. Johnson, R. Wynkoop, and D. W. Collier, *Chem. Eng. Progr.*, **44**, 105 (1948).
14. Yagi, Sakae, and Daizo Kunii, *A.I.Ch.E. Journal*, **3**, 373 (1957).

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Calculation of Equilibrium Flash Vaporization Curves by an Integration Method

C. O. BENNETT, C. J. BRASKET, and J. W. TIERNEY

Remington Rand Univac, St. Paul, Minnesota

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 pressure 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

C. O. Bennett is with the Lummus Company, New York, New York.