0 = axial position

= evaluation of the function at  $R_{\mathsf{o}}$  $R = R_0$ 

 $\mathcal{S}$ = sphere

### LITERATURE CITED

- 1. Brenner, Howard, unpublished papers. —, and John Ĥappel, J.  $\hat{F}luid$
- Mechanics, to be published.

  3. Burke, S. P., and W. B. Plummer, Ind.
- Eng. Chem., 20, 1196 (1928). 4. Dow, W. M., and Max Jakob, Chem.
- Eng. Progr., 47, 637 (1951).
   Gamson, B. W., ibid., 47, 19 (1951).
   Gohr, E. J., in "Fluidization in Practice," by D. F. Othmer, D. Van Nostrand, New York (1954).

- 7. Happel, John, Ind. Eng. Chem., 41, 1161 (1949).
- and B. J. Byrne, ibid., 46, 1181 (1954).
- 9. Happel, John, and Norman Epstein, ibid., 1187.
- 10. Hariu, O. H., and M. C. Molstad, ibid., 41, 1148 (1949).
- 11. Leva, Max et al., Chem. Eng. Progr., 44, 511 (1948).
- Leva, Max, et al., *ibid.*, 45, 563 (1949).
   Lewis, E. W., and E. W. Bowerman, ibid., 48, 603 (1952).
- 14. Lewis, W. K., E. R. Gilliland, and W. C. Bauer, Ind. Eng. Chem., 41, 1104 (1949).
- 15. McCune, L. K., and R. H. Wilhelm, *ibid.*, 41, 1124 (1949).

- 16. Miller, C. O., and A. K. Logwinuk, *ibid.*, **43**, 1220 (1951).
- 17. Parent, J. D., N. Yagel, and C. S. Steiner, Chem. Eng. Progr., 43, 429 (1947).
- 18. Simha, Robert, Kolloid. Z. 76, 16 (1936).
- 19. Toomey, R. D., and H. F. Johnstone, Chem. Eng. Progr., 48, 220 (1952).
- 20. Van Heerden, C., A. P. P. Nobel, and D. W. van Krevelen, Ind. Eng. Chem., **45**, 1237 (1953).
- 21. Wilhelm, R. H., "Proceedings of Second Midwestern Conference on Fluid Mechanics," Ohio State Univ. (1952).
- and Mooson Kwauk, Chem. Eng. Progr., 44, 201 (1948), discussion by L. J. Friend, on p. 218.

# Unsteady State Heat Transfer in Stationary Packed Beds

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A new solution is presented of the differential equations describing unsteady state heat transfer in stationary beds of small granular solid particles through which a fluid is flowing. Arbitrary initial solid temperature distribution and arbitrary variation of inlet gas temperature are allowed. The solution presented appears easier to apply in practice than those previously published and affords an example of the versatility of Fourier integrals and series. An application of the solution to the regeneration of Dow type-B butylene dehydrogenation catalyst is described.

Passage of a fluid through a bed of granular solid is of common occurrence in chemical engineering practice. Several mathematical treatments of unsteady state heat transfer in this situation have been published. Schumann (1) developed the basic differential equations and presented a solution for the case of uniform initial solid temperature and constant entering fluid temperature. His work was extended by Furnes (2), Goldstein (3), and others. More general cases have been covered by Amundson (4, 5). One (4) considers the unsteady state with arbitrary initial solid temperature and also arbitrary inlet fluid temperature at any time. This publication was presented as a solution to a problem in adsorption, but the mathematical statement is identical with that for the heat transfer problem. A more recent publication by Amundson (5) covers much more general cases where heat may be produced or absorbed in the bed, simultaneously transferred through the wells of the containing vessel, etc.

All solutions published to date have been in terms of Bessel functions or other functions which are published for limited ranges of variables or at best for relatively widely separated values. Also, in most practical cases the more general solutions are quite laborious to apply. Presented here are two solutions of the same problem as that treated by Amundson (4), but they are derived from Fourier integrals and Fourier series. The resulting mathematical forms are consequently easy to apply. Convergence is rapid in most cases. The series form of solution is an approximation but in most practical cases it is a very good one. In general, it is easier to use than the integral form, which however is exact.

The problem concerns a bed in the shape of a right porous prism (or cylinder) of granular material the initial temperature of which is an arbitrary function of distance into the bed. A fluid, the inlet temperature of which is an arbitrary function of time, is allowed to pass lengthwise through the bed at a uniform rate of flow, the sides of the bed being impervious to the fluid and to heat. The problem is to find the distribution of temperature in the solid material and in the fluid for all time if it is assumed that

1. The gas and solid temperature are uniform across any section perpendicular to the axis of the prism.

- 2. The solid particles are so small or have such high thermal diffusivity that any given particle may be considered as being at a uniform temperature at any instant.
- 3. Compared with the transfer of heat from fluid to solid, the transfer of heat by conduction, convection, or mixing in the fluid itself or in the solid itself is small and may be neglected.
- 4. The rate of heat transfer from fluid to solid at any point is proportional to the difference in temperature between fluid and solid at that point.
- 5. Change in volume of fluid and solid due to change in temperature may be neglected.
- 6. The thermal constants are independent of the temperature.
- 7. There is no generation or absorption of heat as latent heat, heat of chemical reaction, etc.

# **DERIVATION OF EQUATIONS**

The basic differential equations describing the problem, as derived by Schumann (1), are

$$\frac{\partial t_{g}}{\partial \theta} + \frac{g}{\rho_{g} f} \frac{\partial t_{g}}{\partial x} = -\frac{hs}{C_{g} \rho_{g} f} (t_{g} - t_{s}) \quad (1)$$

$$\frac{\partial t_s}{\partial \theta} = \frac{hs}{\rho_s C_s} (t_s - t_s) \tag{2}$$

Changing the independent variables to yand z gives

$$\frac{\partial t_g}{\partial y} = t_s - t_g \tag{3}$$

$$\frac{\partial t_s}{\partial z} = t_g - t_s \tag{4}$$

and from these

$$\frac{\partial t_s}{\partial z} = -\frac{\partial t_g}{\partial y} \tag{5}$$

By differentiating (3) with respect to z and (4) with respect to y and substituting (5) in both cases, one obtains the following two equations:

$$-\frac{\partial^2 t_g}{\partial y \partial z} = \frac{\partial t_g}{\partial y} + \frac{\partial t_g}{\partial z} \qquad (6)$$

$$-\frac{\partial^2 t_s}{\partial y \partial z} = \frac{\partial t_s}{\partial y} + \frac{\partial t_s}{\partial z} \qquad (7)$$

## **BOUNDARY CONDITIONS**

The boundary conditions are that the initial temperature distribution of the solid is an arbitrary function of position in the bed and that after the datum point in time the temperature of the gas entering the bed is another arbitrary function of time. Expressed mathematically, these

$$t_s = F_1(y); \qquad z = 0$$
 (8)

$$t_g = F_2(z); y = 0 (9)$$

### SOLUTION

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The solution is obtained by first finding a solution for both  $t_s$  and  $t_g$  consistent with boundary condition (8) and assuming datum inlet gas temperature. Another solution is found consistent with boundary condition (9) and assuming datum initial solid temperature. The

sum of these two solutions satisfies both boundary conditions for any temperature datum

A solution of Equation (7) is found, by the method of separation of variables, to be

$$t_s = \left[ a \cos c \left( y - \frac{z}{1+c^2} \right) + b \sin c \left( y - \frac{z}{1+c^2} \right) \right] \exp \frac{-c^2 z}{1+c^2}$$

where a, b, and c are arbitrary constants. When z = 0, this reduces to  $t_s = a \cos cy + b \sin cy$ . This form is the correct one for the formation of a Fourier integral which will converge to  $F_1(y)$  at z = 0 as required by boundary condition (8). It can be shown that this expansion is normally permissible under the conditions of this problem.

The solution shown above has no term containing  $t_{\sigma}$ . In order to produce a solution of this type consistent with datum inlet gas temperature, a means is used which can best be explained from a physical point of view. A bed of solid, infinitely long, and initially at uniform datum temperature throughout is imagined to lie ahead of the real solid bed. This will have the effect of adjusting the gas temperature entering the real bed to the datum temperature. The mathematical justification for this step will be indicated later. Also, to make the Fourier integral converge, the solid from the outlet end of the real bed to infinity in the positive direction is considered to be at datum temperature. Boundary condition (8) can then be expressed, in complete form, as follows:

$$t_s(y, 0) = 0$$
  $y < 0$   
=  $F_1(y)$   $0 < y < Y$   
=  $0$   $Y < y$ 

INLET FLUID TEMPERATURE = 1150 °F.

1250

INITIAL TEMPERATURE DISTRIBUTION TEMPERATURES AFTER 45 MINS. TEMPERATURES AFTER 45 MINS.

1250

1210

1210

1110

Fig. 1. Calculated temperatures of solid packing. Inlet fluid temperature = 1,150°F.

A solution of Equation (7) consistent with this condition can be made by properly choosing the constants a, b, and c to expand the solution into Fourier integral form, which is

$$t_{s} = \frac{1}{\pi} \int_{0}^{\infty} \left[ g(\alpha) \cos \left( y\alpha - \frac{z\alpha}{1 + \alpha^{2}} \right) + h(\alpha) \sin \left( y\alpha - \frac{z\alpha}{1 + \alpha^{2}} \right) \right] \cdot \exp \left( -\frac{z\alpha^{2}}{1 + \alpha^{2}} d\alpha \right)$$
(10)

where

$$g(\alpha) = \int_0^Y F_1(n) \cos \alpha \eta \ d\eta$$

and

$$h(\alpha) = \int_0^Y F_1(n) \sin \alpha \eta \ d\eta$$

If Equation (10) is differentiated with respect to z and substituted into Equation (4), the following expression for  $t_{\sigma}$  results.

$$t_{g} = \frac{1}{\pi} \int_{0}^{\infty} \left[ \frac{g(\alpha) - \alpha h(\alpha)}{1 + \alpha^{2}} \right]$$

$$\cdot \cos \left( y\alpha - \frac{z\alpha}{1 + \alpha^{2}} \right)$$

$$+ \frac{h(\alpha) + \alpha g(\alpha)}{1 + \alpha^{2}}$$

$$\cdot \sin \left( y\alpha - \frac{z\alpha}{1 + \alpha^{2}} \right)$$

$$\cdot \exp \left( -\frac{z\alpha^{2}}{1 + \alpha^{2}} d\alpha \right)$$
 (11)

It can be shown that for all positive z, the preceding expression converges to  $t_{q} = 0$  at y = 0. This is the mathematical justification for the assumption of an infinite imaginary bed initially at datum temperature ahead of the real one.

By a method similar to that used to develop Equations (10) and (11), expressions for  $t_s$  and  $t_g$  can be developed for datum initial solid temperature based on Equation (6) and boundary condition (9). The condition of datum initial solid temperature is achieved by a device analogous to the imaginary bed used previously. Here it is assumed that gas of datum temperature has flowed through the actual bed for an infinite time before the zero of time. The following equations result:

$$t_{s} = \frac{1}{\pi} \int_{0}^{\infty} \left[ \frac{p(\alpha) - \alpha q(\alpha)}{1 + \alpha^{2}} \right]$$

$$\cdot \cos \left( z\alpha - \frac{y\alpha}{1 + \alpha^{2}} \right)$$

$$+ \frac{q(\alpha) + \alpha p(\alpha)}{1 + \alpha^{2}}$$

 $p(\alpha)$  and  $q(\alpha)$  are as defined below.

For any temperature datum, the expressions for  $t_s$  will be the sum of Equations (10) and (12). Similarly,  $t_g$  is given by the sum of Equations (11) and (13). The final solution, then, which satisfies Equations (6) and (7) and boundary conditions (8) and (9) for any temperature datum is

$$t_{s} = \frac{1}{\pi} \int_{0}^{\infty} \left[ g(\alpha) \cos \left( y\alpha - \frac{z\alpha}{1 + \alpha^{2}} \right) + h(\alpha) \sin \left( y\alpha - \frac{z\alpha}{1 + \alpha^{2}} \right) \right]$$

$$+ \exp \left[ -\frac{z\alpha^{2}}{1 + \alpha^{2}} d\alpha \right]$$

$$+ \frac{1}{\pi} \int_{0}^{\infty} \left[ \frac{p(\alpha) - \alpha q(\alpha)}{1 + \alpha^{2}} \right]$$

$$+ \cos \left( z\alpha - \frac{y\alpha}{1 + \alpha^{2}} \right)$$

$$+ \frac{q(\alpha) + \alpha p(\alpha)}{1 + \alpha^{2}}$$

$$\cdot \sin \left( z\alpha - \frac{y\alpha}{1 + \alpha^{2}} \right) \right]$$

$$\exp \left[ -\frac{y\alpha^{2}}{1 + \alpha^{2}} d\alpha \right]$$
 (14)

$$t_{g} = \frac{1}{\pi} \int_{0}^{\infty} \left[ p(\alpha) \cos \left( z\alpha - \frac{y\alpha}{1 + \alpha^{2}} \right) + q(\alpha) \sin \left( z\alpha - \frac{y\alpha}{1 + \alpha^{2}} \right) \right]$$

$$\cdot \exp \left[ -\frac{y\alpha^{2}}{1 + \alpha^{2}} d\alpha \right]$$

$$+ \frac{1}{\pi} \int_{0}^{\infty} \left[ \frac{g(\alpha) - \alpha h(\alpha)}{1 + \alpha^{2}} \right]$$

$$\cdot \cos \left( y\alpha - \frac{z\alpha}{1 + \alpha^{2}} \right)$$

$$+ \frac{h(\alpha) + \alpha g(\alpha)}{1 + \alpha^{2}}$$

$$\cdot \sin \left( y\alpha - \frac{z\alpha}{1 + \alpha^{2}} \right) \right]$$

$$\cdot \exp \left[ -\frac{z\alpha^{2}}{1 + \alpha^{2}} d\alpha \right]$$
(15)

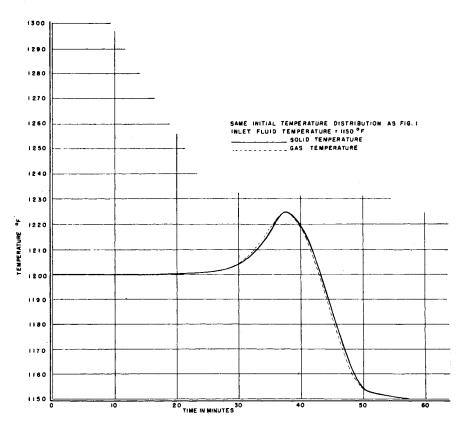


Fig. 2. Calculated temperature of solid packing and gas at a point 5 ft. from fluid entrance into bed. Same initial temperature distribution as Figure 1. Inlet fluid temperature = 1,150°F. ————— solid temperature, - - - - - gas temperature.

where

$$g(\alpha) = \int_0^Y F_1(\eta) \cos \alpha \eta \, d\eta$$

$$p(\alpha) = \int_0^Z F_2(\gamma) \cos \alpha \gamma \, d\gamma$$

$$h(\alpha) = \int_0^Y F_1(\eta) \sin \alpha \eta \, d\eta$$

$$q(\alpha) = \int_0^Z F_2(\gamma) \sin \alpha \gamma \, d\gamma$$

This solution can be shown to be mathematically identical to that of Amundson (4), and Schumann's solution (1) can be shown to be a special case of that above.

In practice, the integrals involved in the solution are usually best evaluated graphically.

# FOURIER-SERIES FORMS

An approach similar to those used in obtaining the foregoing exact forms can be used to produce approximate solutions derived from Fourier series. A solution for  $t_s$  and  $t_g$  is found for datum inlet gas temperature as before by assuming an imaginary bed of solid at datum temperature ahead of the real bed. Because of the finite range of convergence of Fourier series this bed can be of only finite length, and so the gas is brought only approximately to datum temperature. In actual cases the approximation is very good for y and z less than Y. For

y and z greater than Y the approximation fails completely. In practice, this is not usually an important limitation because values of y greater than Y are never of interest and those of z are not usually interesting.

Similarly, the device of obtaining datum initial solid temperature by treatment of the solid with gas of datum temperature is true only as a good approximation and for y and z less than Y.

The Fourier series forms are

$$t_{s} = \frac{g_{0}}{2} + \sum_{n=1}^{\infty} \left[ g_{n} \cos \left( c_{n} y - \frac{c_{n} z}{1 + c_{n}^{2}} \right) + h_{n} \sin \left( c_{n} y - \frac{c_{n} z}{1 + c_{n}^{2}} \right) \right]$$

$$+ \exp \left[ -\frac{c_{n}^{2} z}{1 + c_{n}^{2}} + \sum_{n=1}^{\infty} \left[ \frac{p_{n} - c_{n} q_{n}}{1 + c_{n}^{2}} + \frac{q_{n} + c_{n} p_{n}}{1 + c_{n}^{2}} + \frac{c_{n} z}{1 + c_{n}^{2}} + \frac{q_{n} + c_{n} p_{n}}{1 + c_{n}^{2}} \right] \right]$$

$$+ \exp \left[ -\frac{c_{n} y}{1 + c_{n}^{2}} \right]$$

$$+ \exp \left[ -\frac{c_{n}^{2} y}{1 + c_{n}^{2}} \right]$$

$$+ q_{n} \sin \left( c_{n} z - \frac{c_{n} y}{1 + c_{n}^{2}} \right) \right]$$

$$+ q_{n} \sin \left( c_{n} z - \frac{c_{n} y}{1 + c_{n}^{2}} \right)$$

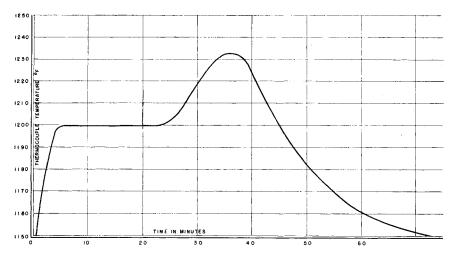


Fig. 3. Typical regeneration pattern for Dow type-B catalyst (observed). Thermocouple 5 ft. down in bed.

$$\exp -\frac{c_{n}^{2}y}{1+c_{n}^{2}}$$

$$+\frac{g_{0}}{2} + \sum_{n=1}^{\infty} \left[ \frac{g_{n} - c_{n}h_{n}}{1+c_{n}^{2}} \right]$$

$$\cdot \cos \left( c_{n}y - \frac{c_{n}z}{1+c_{n}^{2}} \right) + \frac{h_{n} + c_{n}g_{n}}{1+c_{n}^{2}}$$

$$\cdot \sin \left( c_{n}y - \frac{c_{n}z}{1+c_{n}^{2}} \right)$$

$$\cdot \exp -\frac{c_{n}^{2}z}{1+c_{n}^{2}}$$

$$(17)$$

$$c_n = \frac{n\pi}{Y} \quad p_n = \frac{1}{Y} \int_0^Y F_2(\gamma) \cos c_n \gamma \ d\gamma$$

$$g_n = \frac{1}{Y} \int_0^Y F_1(\eta) \cos c_n \eta \ d\eta$$

$$q_n = \frac{1}{Y} \int_0^Y F_2(\gamma) \sin c_n \gamma \ d\gamma$$

$$h_n = \frac{1}{Y} \int_0^Y F_1(\eta) \sin c_n \eta \ d\eta$$

## APPLICATION TO A COMMERCIAL REACTOR

The series form of solution has been applied to a system in which a mixture of steam and air is flowing downward through a bed of  $\frac{3}{16}$  in. cylindrical pellets of Dow type B butylene dehydrogenation catalyst under the following conditions:

$$C_{g} = 0.48^{\text{-}}\text{B.t.u./(lb.)(°F.)}$$

f = 0.366

X = 6 ft.

 $\rho_s = 62.5 \text{ lb./cu. ft.}$ 

 $C_s = 0.24 \text{ B.t.u./(lb.)(°F.)}$ 

g = 225 lb./sq. ft.)(hr.)

 $\rho_a = 0.0714 \text{ lb./cu. ft.}$ 

The initial conditions used were that the solid was at 1,300°F. for the top 6 in. and 1,200°F. for the remainder of the bed.

Figure 1 shows the solid temperature throughout the bed initially and as calculated after the elapse of 25 and 45 min. with the steam-air mixture entering at 1150°F. The heat transfer constant hs used in the calculation of Figure 1 was 3,500 B.t.u./(cu. ft.)(hr./°F.). This is the approximate value indicated by the method of analogy with mass transfer quoted by McAdams (7). The broken line applies to the temperatures calculated at 45 min. and arises because z is greater than Y for this line. For this reason the broken part of the calculated line at 45 min. is incorrect.

Figure 2 is a plot of the calculated solid and gas temperatures at a point 5 ft. into the bed shown against a time base. Figure 3 shows a typical temperature history of a thermocouple shielded by a metallic well, at a point 5 ft. down in the bed under study. The solid temperature curve of Figure 2 was calculated in an attempt to explain the shape of the curve of Figure 3. There is reason to believe that very shortly after the beginning of the regeneration of the catalyst as described in a previous paper (6), the catalyst pellets are at temperatures approximately as described for the initial condition used for calculating Figures 1 and 2 and that little or no heat is produced or absorbed in the bed during the remainder of the regeneration period. The fact that the curve of Figure 3 is of the same general shape as the calculated one in Figure 2 substantiates the postulate that the temperature "surges" as shown in Figure 3 are largely the result of a band of high pellet temperature passing down through the bed by heat transfer.

## GENERALIZATION

Certain problems in mass transfer. such as some cases of chromatography, some ion exchange processes, and some cases of drying by desiccant beds, are analogous to unsteady state heat transfer in packed beds. Where the analogy holds, the solutions developed here can be applied with appropriate changes in definition of the variables.

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#### NOTATION

= specific heat of fluid, B.t.u./ (lb.)(°F.)

= specific heat of solid, B.t.u./ (lb.)(°F.)

void fraction of bed, dimension-

 $F_1(y) = t_s \text{ at } z = 0$ 

 $F_2(z) = t_g \text{ at } y = 0$ 

= mass velocity of fluid, lb./ (sq. ft.)(hr.)

surface heat transfer coefficient, B.t.u./(sq. ft.)(hr./°F.)

effective heat transfer area per unit bulk volume of solid, ft.-1

temperature of fluid, °F. above datum

= temperature of solid, °F. above datum

distance into bed in direction of fluid flow, ft.

total length of bed in direction of fluid flow, ft.

 $=\frac{hsx}{gC_{g}}$ , dimensionless

 $=\frac{hsX}{gC_g}$  dimensionless, the maximum value of y to be considered

 $= \frac{hs}{\rho_s C_s} \left( \theta - \frac{\rho_s fx}{g} \right), \text{ dimensionless}$ 

Z= maximum value of z to be con-

density of fluid lb./cu. ft.

= bulk density of solid lb./cu. ft.

= time, hr.

## LITERATURE CITED

- 1. Schumann, T. E. W., J. Franklin Inst., 208, 405 (1929).
- Furnas, C. C., Ind. Eng. Chem., 22, 721 (1930).
- 3. Goldstein, S., Proc. Royal Soc. (London), 219A, 151 (1953).
- Amundson, N. R., J. Physical & Colloid Chem., 54, 812 (1950).
- Chem., 34, 812 (1930).
   ——, Ind. Eng. Chem., 48, 26 (1956).
   Reilly, P. M., Chemistry in Canada, p. 25, (March, 1953).
   McAdams, W. H., "Heat Transmission," 3 ed., p. 294, McGraw-Hill Book Company. The New York, (1974). pany, Inc., New York, (1954).