

# A Method of Finding Simultaneously the Values of the Heat Transfer Coefficient, the Dispersion Coefficient, and the Thermal Conductivity of the Packing in a Packed Bed of Spheres: Part I. Mathematical Analysis

G. A. TURNER

University of Waterloo, Waterloo, Ontario, Canada

The response of a packed bed to a sine wave of temperature in a stream of fluid through it will depend upon the amount of dispersion in the fluid, the resistance to transfer between fluid and solid, and the thermal properties of the solid. A method is presented that allows the effects of these three phenomena on the amplitude and phase angle to be unravelled and hence all their magnitudes to be computed simultaneously. It thus presents a way of determining these three quantities in situations where they were previously obtainable either with great uncertainty or not at all. The method requires measurements of the relative amplitudes and phase angles at three frequencies, and (preferably) at  $\omega \rightarrow 0$  as well, all to be carried out with high accuracy.

The harmonic response of a physicochemical system has been used for finding values of, for example, heat transfer coefficients (1 to 3, 18, 19), diffusion coefficients of matter into solids (4), equilibrium constants (4, 5), longitudinal dispersion coefficients of mixing in packed beds (6 to 10), reaction rate constants (5), and the thickness of a laminar layer in a tube (11).

In a determination of this kind one measures the frequency response of a system that involves the interaction between a flowing phase and another (usually stationary) phase. All the systems considered by the above authors were simplifications; either the mathematical model contained a restricted number of unknowns (as any model must, to a greater or less extent) or the subsequent analysis contained approximations or restrictions on the range of the variables.

Turner (12, 13) showed that there need be no restriction on the number of quantities whose values can be found simultaneously by harmonic response, provided that an appropriate mathematical model could be set up. The method was demonstrated by showing how size distributions of parameters that define a chosen model of a packed bed could be obtained. In the present paper there are three unknown parameters: the thermal diffusivity of the solid packing, the heat transfer coefficient between the fluid and this solid packing, and the longitudinal dispersion coefficient in the bed. The packing is assumed to be uniformly sized solid spheres. (The method was originally devised to determine the conductivities of solids too small and inconsistent to have their values determined by the classical methods of taking measurements on an individual solid particle.) There are no restrictions, in principle, on the size of the packing. This model is considered to be the simplest one that is close to reality. To use it the physical system has to be made to agree with the model as closely as possible—the reverse of the usual problem.

## THE ARGUMENT

The frequency response of a system that consists of a steady stream of fluid passing through a packed bed of uniform spheres is taken to be the solution of

$$P \frac{\partial^2 T}{\partial \chi^2} - \frac{\partial T}{\partial \chi} - \frac{\epsilon}{\epsilon'} \frac{\partial T}{\partial \theta} - \left( \frac{1 - \epsilon}{\epsilon'} \right) \frac{(\rho c)_s}{(\rho c)_g} \frac{\partial \bar{T}}{\partial \theta} = 0 \quad (1)$$

$$T(0) = A(0)e^{i\Omega\theta} \quad \text{at } \chi = 0$$

$$T(\infty) = 0 \quad \text{at } \chi = \infty$$

for the fluid temperature

$$T = A(\chi) \sin [\Omega\theta - \psi(\chi)]$$

where

$$\frac{\partial \bar{T}}{\partial \theta} = A(\chi) [3Lh/U r_o (\rho c)_s] (Q + iR) \exp i(\Omega\theta - \psi(\chi) + \phi(r_o)) \quad (2)$$

gives the rate of change of the mean temperature, that is

$$\bar{\tau} = (3/r_o^3) \int_0^{r_o} \tau(r) r^2 dr$$

in a solid sphere, situated at distance  $\chi$  downstream of the origin, being heated and cooled by convection (14). It is assumed that the temperature in any sphere depends only on the radius; that is, that the solid is isotropic and that the heat transfer coefficient is uniform over the surface of the sphere. After the substitution for  $\partial \bar{T} / \partial \theta$  by Equation (2), the solution of Equation (1) for  $\psi(0) = 0$  and  $\chi = 1$  is, by the method of Deisler (15)

$$\Pi(1) \equiv \ln [A(1)/A(0)] = 1/(2P) - [(s + b)/2]^{1/2} \quad (3)$$

$$\psi(1) = -[(s - b)/2]^{1/2} \quad (4)$$

as shown in Appendix A.\* From Equations (3) and (4)

$$\nu(\omega_j)P - Zh\lambda(\omega_j) = \Pi(\omega_j), \quad j = 1, 2, 3, \dots \quad (5)$$

where  $\nu = \Pi^2 - \psi^2$ , and  $\lambda$  is a known function of  $h$ ,  $k$ ,  $r_0$ ,  $(\rho c)_s$ , and  $\omega_j$  (Notation) but whose values are as yet unknown.† Measurements at three different frequencies will give rise to three equations of type (5), namely, (5a), (5b), (5c), but in these neither the three unknowns ( $h$ ,  $k$ , and  $P$ ) nor the constants ( $\lambda_1$ ,  $\lambda_2$ ,  $\lambda_3$ ) are known.

However the following deductions can be made:

If a value of  $\lambda_1$  is found (or guessed) the values of  $\lambda_2$ ,  $\lambda_3$ , . . . are thereby fixed, since only  $\omega_1$ ,  $\omega_2$ , and  $\omega_3$  differ, and the values of these are known.

The eliminant of Equations (5a), (5b), and (5c) must be zero, that is,  $P$  and  $h$  being unknowns

$$f_1(h, k) \equiv K_1(\lambda_1/\lambda_3) + K_2(\lambda_2/\lambda_3) + 1 = 0 \quad (6)$$

The elimination of  $P$  between two equations at a time from Equations (5a), (5b), and (5c) gives

$$(f_2)_E \equiv (\nu_3\lambda_2 - \nu_2\lambda_3)(hZ/E) - 1 = 0 \quad (7a)$$

$$(f_2)_F \equiv (\nu_3\lambda_1 - \nu_1\lambda_3)(hZ/F) - 1 = 0 \quad (7b)$$

$$(f_2)_G \equiv (\nu_2\lambda_1 - \nu_1\lambda_2)(hZ/G) - 1 = 0 \quad (7c)$$

of which only two are linearly independent.

Equations (5a), (5b), and (5c) each must be satisfied also.

## PROCEDURE

Any combination of values of  $h$  and  $k$  is tried. This allows the values of  $\lambda_1$ —and so of  $\lambda_2$  and  $\lambda_3$ —to be calculated. Thus the values of  $f_1$ ,  $(f_2)_E$ ,  $(f_2)_F$ , and  $(f_2)_G$  are calculated from Equations (6), (7a), (7b), and (7c), respectively, since  $K_1$ ,  $K_2$ ,  $E$ ,  $F$ , and  $G$  are functions of  $\Pi$  and  $\psi$  only. In this way four surfaces can be generated in  $f$ ,  $h$ ,  $k$  space. The intersection of these with the 0,  $h$ ,  $k$  plane gives curves intersecting one another. [Only two are needed: their intersection gives values of the two unknowns  $h_i$  and  $k_i$  that satisfy simultaneously any two of

\* End effects and the influence of thermometers, etc., on the frequency response are assumed to be avoided by taking measurements either on two beds of different lengths or with the bed removed, and the response found by the arguments in references 6, 9, and 13.

† Dimensional and dimensionless quantities are used here as convenient. For example  $\omega$  can be measured with high accuracy but  $\Omega$  ( $= \omega L/U$ ) cannot; hence the computations are done with  $\omega$ .

Equations (6), (7a), (7b), and (7c).] The substitution of the value  $h_i$ ,  $k_i$  and the corresponding values of  $\lambda_j$  into one of Equations (5) gives a value of  $P_i$ .

In this way at least one combination of values of  $h$ ,  $k$ , and  $P$  is found that satisfies the original equations (5a), (5b), and (5c); that is, generally,  $i = 1, 2, \dots$ . In fact two combinations,  $h_1$ ,  $k_1$  and  $h_2$ ,  $k_2$ , were obtained that satisfied Equations (5). If other values of  $\omega$  were used then the surfaces in  $(f, h, k)$  space were altered, and so the lines  $f_1 = 0$ ,  $(f_2)_E = 0$ ,  $(f_2)_F = 0$ , and  $(f_2)_G = 0$  moved in such a way that one of these two common points of intersection, namely  $(h_1, k_1)$ , was unaltered, while the other,  $(h_2, k_2)$ , shifted. It was found that when  $(h_1, k_1)$  was substituted into one of Equations (5) a positive and so physically possible value of  $D/UL$  was obtained, while the substitution of  $(h_2, k_2)$  led not only to a negative value of  $D/UL$ , but also to one whose magnitude depended on the choice of the three values of  $\omega$ . Thus, means of choosing the correct values  $(h_1, k_1)$  were available.

The values of  $h(Z)$ ,  $k(Z)$  [given by the simultaneous solution of two of Equations (7a), (7b), and (7c)] will depend on  $Z$ ; in fact these values give a line in the  $(h, k)$  plane as  $Z$  is varied, and  $h = h(Z)$ ,  $k = k(Z)$  are the parametric equations of this line. This is Equation (6), which is thus obtained by eliminating  $Z$  between two of Equations (7a), (7b), and (7c).

## THE PERMISSIBLE RANGE OF $\lambda$

When  $\lambda$  is small it tends to the function  $4n^4k/9r_0h(0.2 + k/r_0h)$  and so, by Equation (6),  $f_1$  becomes independent of  $h$  and  $k$  and by Equation (7)  $f_2$  becomes independent of  $k$ , and so the method breaks down. When  $\lambda$  is large it tends to 1.0, and again the method breaks down. A suggested practicable range is  $0.15 < \lambda < 0.95$ , which means that the range of permissible values of  $\omega$  is bounded.

The physical meaning of the statements in the preceding paragraph is that at high values of  $\lambda$  the surface and average temperature waves do not penetrate any significant distance into the particle; at low values dispersion and conductivity are relatively unimportant.

## THE EFFECTS OF CHANGES OF PARAMETERS

1.  $Z$ : The positions taken up by the lines  $(f_2)_E$  (etc.)  $= 0$  in the  $(h, k)$  plane were, in the examples so far con-

TABLE 1. ACTUAL VALUES AND COMPUTED VALUES OF THE THREE QUANTITIES  $h$ ,  $k$ , AND  $P$

Trial No.	$U$	$D$	$P$	$h \times 10^3$	$k \times 10^3$	Value of $Z$ used	Remarks
15	$2.04 \times 10^3$	306.4	0.060	14.25	2.20	—	Actual values
			0.0608	13.99	2.432	188.6	} Computed values
			0.0603	14.28	2.184	189.0	
			0.0598	14.57	1.997	189.4	
17	$0.51 \times 10^3$	76.6	0.060	14.25	2.20	—	Actual values
			0.0592	14.26	2.155	755.0	} Computed values
			0.0603	14.25	2.212	756.0	
			0.0614	14.23	2.277	757.0	
18	$2.04 \times 10^3$	76.6	0.01500	14.25	2.20	—	Actual values
			0.00863	14.06	2.172	188.93	} Computed values
			0.01221	14.17	2.186	188.94	
			0.01517	14.25	2.201	188.95	
			0.01783	14.33	2.218	188.96	

Example: Run 18

$\omega_1 = 0.60000$ ;  $\omega_2 = 0.80000$ ;  $\omega_3 = 1.0000$  rad./sec.  
 $\Pi_1 = -1.3521$ ;  $\Pi_2 = -1.6132$ ;  $\Pi_3 = -1.7734$   
 $\psi_1 = -1.0626$ ;  $\psi_2 = -0.9650$ ;  $\psi_3 = -0.8641$  rad.  
 $t_{18g} = 4.7204$  sec.

$Z = 188.997$  [Calculated from  $t_{18g}$ ; as will be seen from the results above, the correct values of  $P$ ,  $h$ , and  $k$  would not be exactly recaptured with this value of  $Z$ . Units are c.g.s. throughout.]

sidered, very sensitive to small changes in  $Z$ , as illustrated by the examples below. The value of this parameter may be found in one of two ways: from its definition if values of the other quantities in its makeup can be found accurately—a doubtful supposition; or from Appendix B, namely,  $Z \approx 3t_{\text{lag}}/r_o(\rho c)_s$ , where

$$t_{\text{lag}} = \lim (\psi(\omega)/\omega)_{\omega \rightarrow 0}$$

It is essential that the value of  $Z$  be known with high accuracy.

2.  $L$ : The shallower the bed the larger and so more accurately measured is the output amplitude, and the smaller is the phase angle. The radial velocity profile will depend on  $L$ .

3.  $U$ : The greater the velocity the greater is the output amplitudes and the smaller is the resistance to heat transference from air to packing; that is, the significance of the conductivity of the solid is increased. By virtue of the last effect, all curves  $f_1 = 0$  and  $f_2 = 0$  are inclined at greater angles both to one another and to the  $k$  axis in the  $(h, k)$  plane, and the point of intersection is more readily found. However, too large a value of  $h$  will cause  $\lambda$  to decrease below its permitted value. An increase in  $U$  also increases  $D$ ; however the response curves are affected thereby only at the higher frequencies. It will be seen from Table 1 (trials 17 and 18) that a higher value of  $U$ , by itself, leads to a greater sensitivity of the results to values of  $Z$ .

4. The effects of changes of  $r_o$ ,  $(\rho c)_s$ ,  $(\rho c)_g$ ,  $\epsilon$  and  $k$  on the described method are difficult to determine and have not yet been investigated closely.

Note: The model does not take into account any axial conduction in the solid phase. In experimental work this could be guarded against by not having the maximum longitudinal temperature gradient (namely,  $2A\omega/\pi U$  approximately) large.

## THE COMPUTER PROGRAMS

The number of iterations and the accuracy of computation that are necessary have required the use of a digital computer. This has been programmed in two ways:

Phase I (for IBM 7094 computer) computes values of  $f_1$  and  $(f_2)_E$ ,  $(f_2)_F$ , and  $(f_2)_G$ . This is useful in determining the shape of the curves  $f_1 = 0$  and  $f_2 = 0$  in the  $(h, k)$  plane and the extent of likely search areas.

Phase II (for IBM 7094 and IBM 7040 computers) computes, by the Newton-Raphson method, the point  $h, k$  that sets  $f_1$  and one of  $(f_2)_E$ ,  $(f_2)_F$  and  $(f_2)_G$  simultaneously less than a specified quantity.

In addition, another program (phase III, for IBM 7094 computer) computes either the frequency response of a model governed by Equation (1) or the frequency response in a solid sphere of finite conductivity when heat is transferred from the medium by convection, from Equation (A1) in Appendix A. In the second case the program can compute either the responses at two radii in the sphere or the response at any radius relative to the wave in the medium.

## SIMULATED EXAMPLES

The values of  $h$ ,  $k$ , and  $P$  found in three simulations are given in Table 1, together with the values used in the simulation, that is, the correct ones (which were extracted from the literature as being of the right order). The values of the other quantities, common to all three simulations, are  $r_o = 0.15$ ,  $(\rho c)_s = 0.50$ ,  $(\rho c)_g = 3.1 \times 10^{-4}$ ,  $\epsilon = \epsilon' = 0.295$ ,  $L = 2.5$  (units are c.g.s. throughout).

## DISCUSSION

The method could be valuable for measuring heat transfer coefficients between (in particular) gases and spherical packing, a determination that in the past has been restricted to rather large sizes of packing. It can also be used to determine the thermal conductivity of solids too small in size or too inconsistent in structure (for example fertilizers, sinter, vegetables, and sweets and candies) to have it measured in conventional ways, for this value needs to be known if a rational design of equipment for heating or cooling solids is to be achieved (as in references 16 and 17 for example).

## APPARATUS

Apparatus to measure sinusoidal temperature waves with high accuracy (in a physical model resembling the above mathematical model as closely as possible) was set up at Bramford Development Station, Ipswich, England. It was inadequate because of too low an air velocity and too low an upper limit of frequency. A second apparatus is being built at the University of Waterloo, incorporating and improving on features of the first one.

## ACKNOWLEDGMENT

This work was carried out while the author was with Fisons Fertilizers, Ltd., Bramford Development Station, Ipswich, England. My thanks are due to the directors for permission to publish this paper.

My grateful thanks are due to Professor P. L. Ponzo, Mathematics Department, University of Waterloo, for his help and advice.

## NOTATION

- $A$  = amplitude of a sine wave (middle to peak), °C.
- $a_1(r) = + (2)^{-1/2} (\cosh 2m - \cos 2m)^{1/2}$ , dimensionless
- $a_2(r_o) = n[\Gamma + (r_o h/k - 1)^2 \gamma/2n^2 + \Sigma(r_o h/k - 1)/n]^{1/2}$ , dimensionless
- $b = (1/2P)^2 + W\lambda r_o h/(\rho c)_s D$ , dimensionless
- $c$  = specific heat, cal./(g.) (°C.)
- $D$  = longitudinal dispersion coefficient, sq.cm./sec.
- $E = \Pi_3 \nu_2 - \Pi_2 \nu_3$ , dimensionless
- $\bar{E} = [A(1)/A(0)] \exp(-i\psi)$ , dimensionless
- $F = \Pi_1 \nu_3 - \Pi_3 \nu_1$ , dimensionless
- $f_1 = K_1(\lambda_1/\lambda_3) + K_2(\lambda_2/\lambda_3) + 1$ , dimensionless
- $(f_2)_E = (\nu_3 \lambda_2 - \nu_2 \lambda_3) hZ/E - 1$ , dimensionless
- $(f_2)_F = (\nu_3 \lambda_1 - \nu_1 \lambda_3) hZ/F - 1$ , dimensionless
- $(f_2)_G = (\nu_2 \lambda_1 - \nu_1 \lambda_2) hZ/G - 1$ , dimensionless
- $G = \Pi_2 \nu_1 - \Pi_1 \nu_2$ , dimensionless
- $g = \epsilon \Omega/\epsilon' P + W X r_o h/(\rho c)_s D$ , dimensionless
- $h$  = heat transfer coefficient, air to surface of packing, cal./(sec.) (sq.cm.) (°C.)
- $i = \sqrt{-1}$
- $K_1 = (\Pi_3 \nu_2 - \Pi_2 \nu_3)/(\Pi_2 \nu_1 - \Pi_1 \nu_2)$ , dimensionless
- $K_2 = (\Pi_1 \nu_3 - \Pi_3 \nu_1)/(\Pi_2 \nu_1 - \Pi_1 \nu_2)$ , dimensionless
- $k$  = thermal conductivity of individual spheres in packed bed, cal./(cm.) (sec.) (°C.)
- $L$  = length of bed, cm.
- $m = r(\omega/2\alpha)^{1/2}$ , dimensionless
- $n = r_o(\omega/2\alpha)^{1/2}$ , dimensionless
- $P = D/UL$  (the inverse of the Peclet number), dimensionless
- $Q = (n\Sigma - \gamma)/[(2\gamma)^{1/2} a_2]$ , dimensionless
- $R = n\sigma/[(2\gamma)^{1/2} a_2]$ , dimensionless
- $s = (b^2 + g^2)^{1/2}$ , dimensionless
- $t$  = time, sec.
- $t_{\text{lag}} = \lim (\psi_o/\omega)_{\omega \rightarrow 0}$ , sec.
- $U$  = velocity through bed (interstitial), cm./sec.
- $W = 3(L^2/r_o^2)/[(\rho c)_s/(\rho c)_g] [(1 - \epsilon)/\epsilon']$ , dimensionless

$X = Q \sin \phi + R \cos \phi = I[(Q + iR) \exp i\phi]$ , dimensionless  
 $Z = 3[(1 - \epsilon)/\epsilon'] (L/r_o) [1/U(\rho c)_g]$ , (sec.) (sq. cm.) (°C.) / cal.  
 $z$  = distance down bed, cm.

#### Greek Letters

$\alpha = k/(\rho c)_s$  = thermal diffusivity, sq.cm./sec.  
 $\Gamma = \cosh 2n + \cos 2n$ , dimensionless  
 $\gamma = \cosh 2n - \cos 2n$ , dimensionless  
 $\epsilon$  = volume porosity of bed = (free space/total volume), dimensionless  
 $\epsilon'$  = cross-sectional area porosity of bed = (free area/total area), dimensionless  
 $\theta$  = dimensionless time =  $Ut/L$ , dimensionless  
 $\lambda = Q \cos \phi - R \sin \phi = R[(Q + iR) \exp i\phi]$ , dimensionless  
 $\nu = \Pi^2 - \psi^2$ , dimensionless  
 $\Pi = \ln A(1)/A(0)$ , dimensionless  
 $\rho$  = density, g./cc.  
 $(\rho c)$  = volumetric specific heat, cal./cc.) (°C.)  
 $\Sigma = \sinh 2n + \sin 2n$ , dimensionless  
 $\sigma = \sinh 2n - \sin 2n$ , dimensionless  
 $\tau(r, t)$  = temperature in sphere at radius  $r$ , °C.  
 $\bar{\tau}(t)$  = mean temperature in sphere, °C.  
 $\phi(r) = \phi_1(r) - \phi_2$ , dimensionless  
 $\phi_1(r) = \tan^{-1}(\coth m \tan m)$ , dimensionless  
 $\phi_2 = \tan^{-1}\{[1 + \tanh n \tan n + (r_o h/k - 1)(\tan n)/n]/[1 - \tanh n \tan n + (r_o h/k - 1)(\tanh n)/n]\}$ , dimensionless  
 $\chi$  = dimensionless distance =  $z/L$ , dimensionless  
 $\psi(z)$  = phase angle in gas stream relative to inlet temperature wave, rad., dimensionless  
 $\psi_o$  = limiting value of  $\psi$  as  $\omega \rightarrow 0$ , rad., dimensionless  
 $\omega$  = angular frequency, rad./sec.  
 $\Omega = \omega L/U$ , dimensionless

#### Subscripts

$g$  = gas  
 $i$  = particular combination of  $h$  and  $k$   
 $s$  = solid

19. Shearer, C. J., *Rept. No. 55, Natl. Eng. Lab., East Kilbride, Scotland* (1964).

#### APPENDIX A

The temperature  $\tau(r, t)$  in a sphere of conductivity  $k$  and diffusivity  $\alpha$  due to a sinusoidal temperature  $A \sin(\omega t - \psi)$  is (14)

$$\tau(r, t) = A(r_o h/k) (r_o a_1/r a_2) \sin[(\omega t - \psi + \phi(r))] \quad (A1)$$

where  $\phi(r) = \phi_1 - \phi_2$   
 $a_1 \exp(i\phi_1) = \sinh m \cos m + i \cosh m \sin m$ ;  
 $a_2 \exp(i\phi_2) = n(1+i) \cosh[n(1+i)] + (r_o h/k - 1) \sinh[n(1+i)]$ ;  
 $m = r(\omega/2\alpha)^{1/2}$ ; and  $n = r_o(\omega/2\alpha)^{1/2}$

Equation (2) is derived from Equation (A1) by the method of Deisler (15) by using

$$\frac{d\bar{\tau}(t)}{dt} = (3\alpha/r_o) \frac{d\tau(r_o)}{dr}$$

(converted to dimensionless time  $\theta$  by putting  $t = \theta L/U$ ). Substitution of Equation (2) into Equation (1) gives

$$\exp(i\omega t) \left[ P \frac{d^2 \bar{E}}{d\chi^2} - \frac{d\bar{E}}{d\chi} - i[(\epsilon\omega L)/(U\epsilon')] \bar{E} - 3[(1-\epsilon)h/L]/[U\epsilon' r_o (\rho c)_g] Q + iR \bar{E} \exp i\theta \right] = 0 \text{ at } \chi = 1$$

where  $\bar{E} = [A(1)/A(0)] \exp(-i\psi)$ . This is solved in the complex variable  $E$  to give Equations (3) and (4).

#### APPENDIX B

Equations (3) and (4) are differentiated with respect to  $n$  to give, respectively

$$\frac{d\Pi}{dn} = \text{function}(g, b) = \text{function}(\lambda, X) = \text{function}(Q, R, \theta) \quad (B1)$$

and similarly

$$\frac{d\psi}{dn} = \text{function}(Q, R, \theta) \quad (B2)$$

The limiting values of  $Q$ ,  $R$ , and  $\phi$ , and of their first derivatives with respect to  $n$ , as  $n \rightarrow 0$  and  $n \rightarrow \infty$  by virtue of  $\omega \rightarrow 0$  and  $\omega \rightarrow \infty$ , respectively, can be found after expansion in series of their defining equations; these limiting values can be substituted into Equations (B1) and (B2) to give

$$\left( \frac{d\Pi}{dn} \right)_{n \rightarrow 0}, \left( \frac{d\psi}{dn} \right)_{n \rightarrow 0}, \left( \frac{d\Pi}{dn} \right)_{n \rightarrow \infty}, \left( \frac{d\psi}{dn} \right)_{n \rightarrow \infty} \text{ as func-}$$

tions of  $n$  and parameters. Then, since

$$\begin{aligned} \frac{d}{d\omega} \cdot &= \frac{dn}{d\omega} \frac{d}{dn} \cdot = (r_o^2/4\alpha n) \frac{d}{dn} \cdot \\ \frac{d}{d(\omega^{1/2})} \cdot &= \frac{dn}{d(\omega^{1/2})} \frac{d}{dn} \cdot = r_o/(2\alpha)^{1/2} \frac{d}{dn} \cdot \\ \frac{d}{d(\omega^2)} \cdot &= \frac{dn}{d(\omega^2)} \frac{d}{dn} \cdot = r_o^4/16\alpha^2 n^3 \frac{d}{dn} \cdot \end{aligned}$$

the following limiting values are obtained:

$$\lim \left( \frac{d\Pi}{d(\omega^2)} \right)_{\omega \rightarrow 0} = -3(1-\epsilon)Lr_o^2(\rho c)_s [4/45\alpha + 4(\rho c)_s/9r_o h]/U\epsilon'(\rho c)_g + 4P[(\rho c)_s/(\rho c)_g - \epsilon((\rho c)_s/(\rho c)_g - 1)]^2 L/(\epsilon'U)^2 \quad (B3)$$

$$\approx -(1-\epsilon)(\rho c)_s \{3Lr_o^2(4/45\alpha + 4(\rho c)_s/9r_o h)/U + 4P(\rho c)_s(1-\epsilon)L^2/\epsilon'U^2(\rho c)_g\}/(\rho c)_g \epsilon' \quad (B4)$$

#### LITERATURE CITED

1. Bell, J. C., and E. F. Katz, "Heat Transfer and Fluid Mechanics Institute," Am. Soc. Mech. Engr., Berkeley, Calif. (June 22, 1949).
2. Dayton, R. W., et al., *Rept. BMI-747*, Battelle Memorial Inst. (1952).
3. Littman, Howard, and A. P. Stone, *Chem. Eng. Progr. Symp. Ser. No. 62*, 62, 47 (1966).
4. Deisler, P. F., Jr., and R. H. Wilhelm, *Ind. Eng. Chem.*, 45, 1219 (1953).
5. Rosen, J. B., and W. E. Winsche, *J. Chem. Phys.*, 18, 1587 (1950).
6. McHenry, K. W., Jr., and R. H. Wilhelm, *AIChE J.*, 3, 83 (1957).
7. Ebach, E. A., and R. R. White, *ibid.*, 4, 2 (1958).
8. Strang, D. A., and C. J. Geankoplis, *Ind. Eng. Chem.*, 50, 1305 (1958).
9. Liles, A. W., and C. J. Geankoplis, *AIChE J.*, 6, 591 (1960).
10. Stahel, E. P., and C. J. Geankoplis, *ibid.*, 10, 174 (1964).
11. Keyes, J. J., Jr., *ibid.*, 1, 305 (1955).
12. Turner, G. A., *Chem. Eng. Sci.*, 7, 156 (1958).
13. *ibid.*, 10, 14 (1959).
14. Carslaw, H. S., and J. C. Jaeger, "Conduction of Heat in Solids," Clarendon, Oxford (1959).
15. Deisler, P. F., Jr., Ph.D. dissertation, Princeton Univ., N. J. (1952).
16. Turner, G. A., *Can. J. Chem. Eng.*, 44, 13 (1966).
17. Porter, S. J., *Trans. Inst. Chem. Eng. (London)*, 41, 272 (1963).
18. Meek, R. M. G., *Rept. No. 54, Natl. Eng. Lab., East Kilbride, Scotland* (1962).

Also

$$t_{lag} = \lim (\psi_0/\omega)_{\omega \rightarrow 0} = \lim \left( \frac{d\psi}{d\omega} \right)_{\omega \rightarrow 0} = - [(\rho c)_s/(\rho c)_g - \epsilon ((\rho c)_s/(\rho c)_g - 1)] L/\epsilon' U \quad (B5)$$

$$\approx - (\rho c)_s (1 - \epsilon) L/(\rho c)_g \epsilon' U \quad (B6)$$

[The approximate forms, Equations (B4) and (B6), are permitted if  $(\rho c)_s/(\rho c)_g \approx 1,000$ ]. From the definition of  $Z$  it follows from Equation (B6) that  $Z \approx 3t_{lag}/r_0 (\rho c)_s$ . Again

$$\lim (\psi)_{\omega \rightarrow \infty} = \lim (\Pi)_{\omega \rightarrow \infty} = \lim [(s/2)^{1/2}]_{\omega \rightarrow \infty} = - (\epsilon \omega L^2/2D\epsilon')^{1/2} \quad (B7)$$

and

$$\lim \left( \frac{d\psi}{d(\omega^{1/2})} \right)_{\omega \rightarrow \infty} = \left( \frac{d\Pi}{d(\omega^{1/2})} \right)_{\omega \rightarrow \infty} = - L(\epsilon/\epsilon'D)^{1/2}/2 \quad (B8)$$

[In principle Equation (B8) could be used for estimating  $D$ .]

*Manuscript received April 25, 1966; revision received October 26, 1966; paper accepted October 26, 1966.*

# Generalized Solution of the Tomotika Stability Analysis for a Cylindrical Jet

BERNARD J. MEISTER and GEORGE F. SCHEELE

Cornell University, Ithaca, New York

The stability of cylindrical jets in immiscible liquid systems is analyzed with the low velocity theory of Tomotika. For the first time the several limiting solutions in the literature are obtained from a general equation, so approximate restrictions on their applicability can be presented. These restrictions show that for many systems none of the limiting solutions is valid. Correlations applicable to all Newtonian liquid-liquid systems are presented for predicting the growth rate and wavelength of the most unstable disturbance.

The injection of one liquid into another is important in many industrial operations. At low injection velocities drops are formed directly at the nozzle and their size is controlled by the forces acting on the forming drop (3). At higher injection velocities a jet of liquid issues from the nozzle and then breaks into droplets in a regular pattern. This breakup of a cylinder of liquid has interested many scientists.

In 1873 Plateau (6) showed that a cylinder of liquid subject to surface forces is unstable if its length exceeds its circumference, because it can be divided into two spheres of equal volume with an accompanying decrease in surface area. This analysis indicated that surface forces are the cause of jet breakup and that the waves visible on the jet surface should have a wavelength equal to the circumference of the jet.

In 1879 Lord Rayleigh (7, 8) set forth several postulates concerning wave forms on a jet which have been the basis of most subsequent instability analyses. He assumed that disturbances corresponding to all possible wavelengths are initiated at the nozzle exit as a result of density and pressure fluctuations. The amplitude  $\xi$  of any resulting wave is given by the equation

$$\xi = \xi_0 \exp(\alpha t + ikz) \quad (1)$$

where  $\xi_0$  is the initial amplitude of the disturbance and  $\lambda$  is the disturbance wavelength which is related to the wave number  $k$  by the equation

$$\lambda = 2\pi/k \quad (2)$$

All waves with a wavelength greater than the jet circumference have a positive growth rate  $\alpha$  and amplify with time. Rayleigh assumed that all the initial disturbances are very small and of the same magnitude. Therefore the wave with the largest growth rate becomes the dominant wave on the jet surface and ultimately breaks the jet into drops. Rayleigh derived equations predicting the disturbance growth rate for a nonviscous liquid jet in a gas (7, 8), a gas jet in a nonviscous liquid (10), and an extremely viscous liquid in a gas (9). Further derivations have been made by Weber (14) for a viscous liquid jet in a gas, by Tomotika (13) for a very viscous liquid jet in a very viscous liquid medium, and by Christiansen (1, 2) for a nonviscous liquid jet in a nonviscous liquid medium. Other relevant theoretical instability studies have included non-Newtonian and viscoelastic effects (5, 16, 17) and jet contraction effects (15). All these equations are limited to jet velocities sufficiently low that the gross jet velocity does not affect the wavelength and growth rate of the dominant disturbance.

The agreement between experimental results and theoretical instability analyses has been good when experiments are performed which closely approximate the assumptions made in the theoretical development (1, 4, 11, 12). There are, however, two significant limitations to these analyses which prevent direct application to many experimental studies. In the first place, while the solutions are for limiting cases, all of the analyses except Tomotika's assume the character of the fluids at the start of the derivation, so it is not possible to set quantitative limits on the applicability of the resulting equations. This has created uncertainty as to which analysis should be applied in a particular situation because there are no estimates available of the ranges of physical properties

Bernard J. Meister is with the Dow Chemical Company, Midland, Michigan.