

y = ratio of R_2 to R_1
 z = ratio of the derivative time constant to γ_1/γ_2 ; see Equation (7d)
 X = positive constant used to multiply each process time constant in a set

Greek Letters

α_1 = defined by Equation (7b)
 α_2 = defined by Equation (7c), s
 γ_1 = product of the process time constants; $\tau_1 \cdot \tau_2 \cdot \tau_3$, s
 γ_2 = $\tau_1 \tau_2 + \tau_1 \tau_3 + \tau_2 \tau_3$, s²
 γ_3 = sum of process time constants; $\tau_1 + \tau_2 + \tau_3$, s
 τ_1 = smallest process time constant, s
 τ_2 = second largest process time constant, s
 τ_3 = largest process time constant, s
 τ_i = reset or integral action time constant, s
 τ_D = derivative action time constant, s
 τ_{IZ-N} ; τ_{DZ-N} = values of τ_i and τ_D suggested by the Ziegler-Nichol's rules, s

τ_{ib} = all lower values of τ_i will lead to operation in the split stability region; defined by Equation (10), s
 τ_{ic} = lowest value of τ_i for $z = 1$ for which any value of K_c will lead to stable operation, s
 ω_c = critical frequency (that is, 180 deg phase lag), s⁻¹

LITERATURE CITED

Buckley, P. S., *Techniques of Process Control*, Wiley, New York (1964).
 Coughanowr, D. R., and L. B. Koppel, *Process Systems Analysis and Control*, McGraw-Hill, New York (1965).
 Harriott, P., *Process Control*, McGraw-Hill, New York (1965).
 Weast, R. C., and S. M. Selby, *Handbook of Tables for Mathematics*, 4 ed., CRC Press, Cleveland, Ohio (1975).

Manuscript received May 10, 1978; revision received August 2 and accepted August 25, 1978.

Kinetics of Fixed-Bed Adsorption: A New Solution

C. H. LIAW, J. S. P. WANG, R. A. GREENKORN, and K. C. CHAO

School of Chemical Engineering
 Purdue University, West Lafayette, Indiana 47907

A new solution is obtained to the kinetics of a fixed-bed adsorber in response to a step change in feed concentration for a linear equilibrium system with consideration for the resistance to mass transfer in both the mobile and stationary phases. The differential equations of continuity and mass transfer are integrated upon simulating the intraparticle concentrations with a parabola. The results agree with Rosen's rigorous but complex solution in the range of conditions of practical interest. The present solution is more convenient for computer application for the calculation of breakthrough curves.

Adsorption in packed columns is in wide industrial use for the removal or recovery of dilute components from a fluid stream. The operation is gaining in importance in processes for the removal of pollutants from wastewater and exhaust gases.

A central problem in the design of these processes is the dynamic response of the adsorption column to a step change in input. A number of investigators have studied the problem, and the solutions that have been obtained can be broadly classified into two general types: the equilibrium and the nonequilibrium theories. In the equilibrium theories, the local concentrations of the adsorbate in the mobile and stationary phases are assumed to be at equilibrium. The results have been found to describe laboratory analytical columns in which the packing particles are small, and the fluid flow rates are low. However, the equilibrium theories do not quantitatively apply to industrial adsorbers owing to significant resistance to mass transfer in both the mobile and stationary phases.

The nonequilibrium theories take into account the finite resistance to mass transfer in the mobile and stationary phases and are capable of giving a quantitative description of industrial columns. Thomas (1944) and Edeskuty and Amundson (1952) presented results obtained by consider-

ing intraparticle diffusion resistance but ignoring fluid to particle resistance. Masamune and Smith (1965) considered the finite rates of surface adsorption in conjunction with either intraparticle diffusion or external diffusion. Rosen (1952) presented an analytical solution of the combined effects of intraparticle and external diffusion for linear equilibrium systems.

A uniform temperature and pressure are considered to prevail in the column in all the analytical solutions. This condition is closely approximated in liquid adsorption systems in which the heat of adsorption is small. It is approximated in a gas adsorption system only when the feed gas is highly diluted in the adsorbate.

We present here a new solution to the same problem that Rosen addressed. In Rosen's work, the partial differential equation of Fick's law describing intraparticle diffusion is integrated in conjunction with the external mass transfer equation and the continuity equation of flow. In this work, a parabola simulates the concentration profile in the particle that is developed as a result of diffusion. Integration is then carried out with the external mass transfer equation and the continuity equation of flow.

It is a common mathematical procedure to approximate an arbitrary function with a polynomial, and examples are too many to enumerate. We mention only von Karman's (1921) expression of the velocity profile in a boundary layer in terms of a polynomial of distance from the solid surface. The solution obtained is in good agreement with Blasius' (1908) more rigorous results. For the present

Correspondence concerning this note should be addressed to K. C. Chao. C. H. Liaw is with the Chung-Shan Institute of Science and Technology, Taiwan. J. S. P. Wang is with the School of Mathematics, Purdue University.

0001-1541/79-1887-0376-\$00.75. © The American Institute of Chemical Engineers, 1979.

problem, polynomials of various degrees may be used in principle, but the parabola is the lowest polynomial that is acceptable from symmetry considerations. An interesting note to the parabolic concentration profile is the mass transfer rate equation it gives as a function of concentration at the external particle surface and the average intraparticle concentration. The rate equation is linear in the difference between these two concentrations, which is the same as in Glueckauf's equation (1955), and the linear coefficient is also the same.

An equation for the rate of mass transfer to the particle is readily derived by starting with the continuity equation for the adsorbate in the porous solid

$$\frac{\partial q}{\partial t} = \frac{D}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial q}{\partial r} \right) \quad (1)$$

The average intraparticle concentration is

$$\bar{q} = \frac{3}{R^3} \int_0^R q r^2 dr \quad (2)$$

The rate of change of \bar{q} is obtained upon differentiation of Equation (2):

$$\frac{\partial \bar{q}}{\partial t} = \frac{3}{R^3} \int_0^R \left(\frac{\partial q}{\partial t} \right) r^2 dr \quad (3)$$

Substituting Equation (1) into (3), we get

$$\frac{\partial \bar{q}}{\partial t} = \frac{3}{R^3} \int_0^R D \frac{\partial}{\partial r} \left(r^2 \frac{\partial q}{\partial r} \right) dr \quad (4)$$

$$= \frac{3D}{R} \left(\frac{\partial q}{\partial r} \right) \Big|_{r=R} \quad (5)$$

Let the concentration q be approximated by a parabola

$$q = a_0 + a_2 r^2 \quad (6)$$

where a_0 and a_2 are functions of time t and axial distance q , but not of r . The linear term has been left out from Equation (6) on account of spherical symmetry.

From Equation (6) it follows that

$$\left(\frac{\partial q}{\partial r} \right) \Big|_{r=R} = \frac{5}{R} (q_R - \bar{q}) \quad (7)$$

Substituting Equation (7) into (5), we get

$$\frac{\partial \bar{q}}{\partial t} = \frac{15D}{R^2} (q_R - \bar{q}) \quad (8)$$

Equation (8) expresses the time rate of change of average intraparticle concentration to be linearly dependent on the difference $(q_R - \bar{q})$.

Glueckauf obtained the same results by considering an arbitrary variation of the surface concentration of the packing particle. The general solution was obtained as an integral of an infinite series which simplifies to Equation (8) for large values of Dt/R^2 . Equation (8) is in wide use and is discussed by Vermeulen et al. (1973).

We now proceed to the dynamics of the adsorber bed with the use of Equation (6).

THE DIFFERENTIAL RATE EQUATIONS

The equation of continuity of the mobile fluid phase is expressed by

$$v \frac{\partial c}{\partial z} + \frac{\partial c}{\partial t} = - \left(\frac{\partial \bar{q}}{\partial t} \right) / \eta \quad (9)$$

where $\eta = \epsilon/(1 - \epsilon)$.

The usual assumptions are made in connection with Equation (9):

1. The velocity and concentration of the mobile fluid phase is independent of radial position at any cross section of the bed.

2. Axial diffusion is negligible.

3. Temperature and pressure are uniform in the bed.

We introduce the change of variables

$$x = z/(\eta v) \quad (10)$$

and

$$\theta = t - z/v \quad (11)$$

Equation (9) is simplified when expressed in terms of the new variables

$$\frac{\partial c}{\partial x} = - \frac{\partial \bar{q}}{\partial \theta} \quad (12)$$

The rate of interphase mass transfer is described by

$$D \frac{\partial q}{\partial r} \Big|_{r=R} = k (c - q_R/K) \quad (13)$$

and

$$\frac{\partial \bar{q}}{\partial \theta} = \frac{3k}{R} (c - q_R/K) \quad (14)$$

Both D and k are constant as a result of the uniformity of temperature and pressure and low concentrations of solute throughout the bed.

Equations (12), (13), and (14) make up a set of simultaneous equations for c and q . We eliminate q from the equations to obtain a partial differential equation for c only upon simulating q with a parabola as in Equation (6). We obtain from Equation (6)

$$q_R = a_0 + a_2 R^2 \quad (15)$$

$$\frac{\partial q}{\partial r} \Big|_{r=R} = 2a_2 R \quad (16)$$

$$\bar{q} = a_0 + (3/5) a_2 R^2 \quad (17)$$

Combining Equations (13), (15), (16), and (17), we get

$$\bar{q} = \left(1 + \frac{Rk}{5DK} \right) q_R - \frac{Rk}{5D} c \quad (18)$$

Differentiation of Equation (18) gives

$$\frac{\partial \bar{q}}{\partial \theta} = \left(1 + \frac{Rk}{5DK} \right) \frac{\partial q_R}{\partial \theta} - \frac{Rk}{5D} \frac{\partial c}{\partial \theta} \quad (19)$$

To eliminate $\partial q_R / \partial \theta$ from Equation (19), we substitute Equation (12) into (14) and differentiate to obtain

$$\frac{\partial q_R}{\partial \theta} = \frac{KR}{3k} \frac{\partial^2 c}{\partial x \partial \theta} + K \frac{\partial c}{\partial \theta} \quad (20)$$

substituting Equations (20) and (12) into (19)

$$\frac{\partial c}{\partial x} = -K \frac{\partial c}{\partial \theta} - \xi \frac{\partial^2 c}{\partial x \partial \theta} \quad (21)$$

where

$$\xi = R \left(\frac{K}{3k} + \frac{R}{15D} \right) \quad (22)$$

In Equation (21), we have succeeded in eliminating q and obtained an equation for c only. The concentration is more conveniently expressed in a reduced form $u \equiv c/c_0$, and Equation (21) becomes

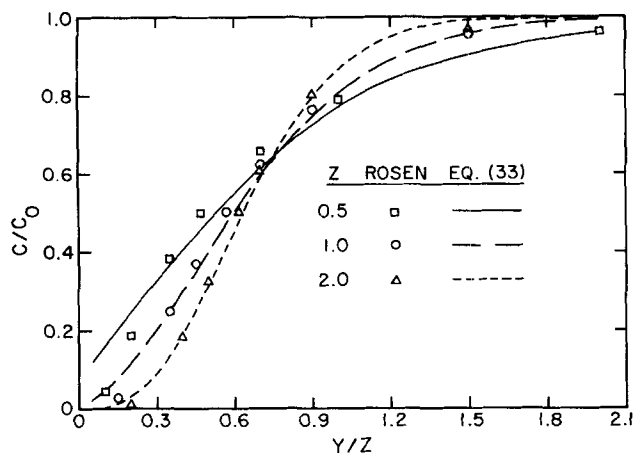


Fig. 1. Breakthrough curves for selected values of bed length Z with film resistance parameter $\nu/Z = 0.0$.

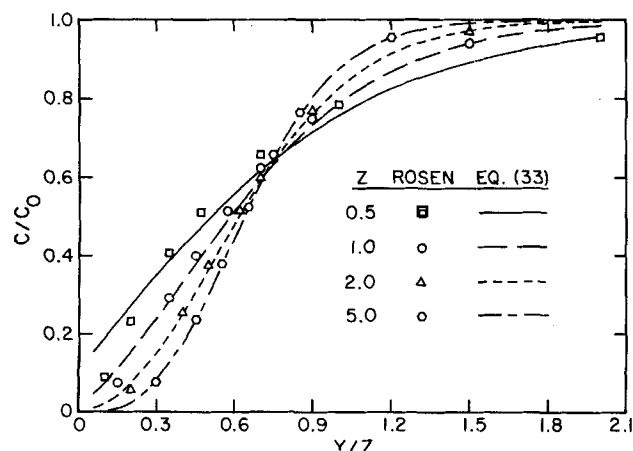


Fig. 3. Breakthrough curves for selected values of bed length Z with film resistance parameter $\nu/Z = 0.05$.

$$\frac{\partial u}{\partial x} = -K \frac{\partial u}{\partial \theta} - \xi \frac{\partial^2 u}{\partial x \partial \theta} \quad (23)$$

Equation (23) is now to be solved with the initial and boundary conditions:

$$u = 0 \quad \text{at } \theta \leq 0 \quad \text{and all } x \quad (24)$$

$$u = 1 \quad \text{at } \theta > 0 \quad \text{and } x = 0 \quad (25)$$

INTEGRATION OF THE RATE EQUATION

To integrate Equation (23), we take its Laplace transform

$$\frac{\partial U}{\partial x} = \frac{-Ks}{1 + \xi s} U \quad (26)$$

where we have made use of the initial condition Equation (24). The equation is integrated with respect to x

$$U = \frac{1}{s} \exp[-Ksx/(1 + \xi s)] \quad (27)$$

where the constant of integration has been determined from the transform of Equation (25).

The exponential function of Equation (27) is expanded into a series and then inverted term by term to give the solution in the time domain

$$u = 1 + e^{\Theta} \sum_{n=0}^D \sum_{i=0}^n \frac{X^{n+1}\Theta^i}{(n+1)[(n-i)!](i!)^2} \quad (28)$$

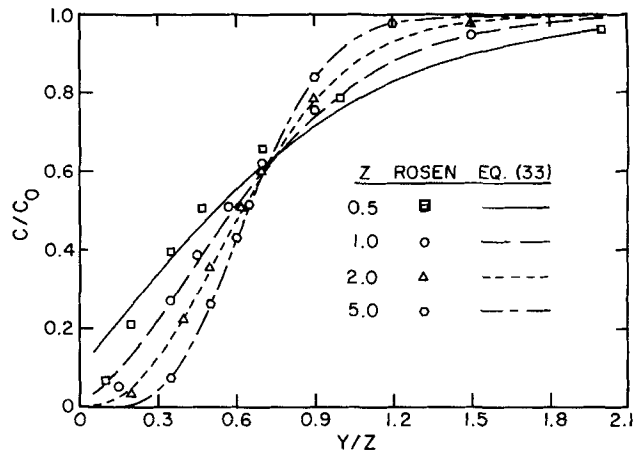


Fig. 2. Breakthrough curves for selected values of bed length Z with film resistance parameter $\nu/Z = 0.025$.

where Θ and X are reduced dimensionless time and distance, respectively, defined by

$$\Theta = -\theta/\xi \quad (29)$$

$$X = -Kx/\xi \quad (30)$$

The infinite series in Equation (28) is convergent, and the proof is given by Liaw (1977). Therefore, the series can be evaluated as the sum of a finite number of terms

$$u_N = 1 + e^{\Theta} \sum_{n=0}^N \sum_{i=0}^n \frac{X^{n+1}\Theta^i}{(n+1)[(n-i)!](i!)^2} \quad (31)$$

where N is a suitably large number. For $|\Theta| \leq 50$ and $|X| \leq 18$, an error of not more than 0.1% is incurred with $N = 80$. But more terms may have to be summed for larger absolute values of either Θ or X , and the sum becomes difficult or impossible to evaluate on computers on account of the very large values of $(i!)^2$ that will be encountered. To avoid evaluating a large factorial, we reexpress Equation (28) in a computationally more efficient form:

$$u = e^{\Theta+X} \left[1 + \sum_{i=1}^{\infty} \left[\prod_{k=1}^i \frac{|\Theta|}{k} \right] \left[1 + \sum_{j=1}^i \prod_{k=1}^j \frac{|X|}{k} \right] \right] \quad (32)$$

The derivation is given by Liaw (1977). Again, the infinite series may be replaced by the sum of the first N terms:

$$u_N = e^{\Theta+X} \left[1 + \sum_{i=1}^N \left[\prod_{k=1}^i \frac{|\Theta|}{k} \right] \left[1 + \sum_{j=1}^i \prod_{m=1}^j \frac{|X|}{m} \right] \right] \quad (33)$$

The remainder of Equation (33) may be shown (Liaw, 1977) to be

$$R_N = u - u_N < \frac{u_N}{2^{N-\text{Max}(2\bar{\Theta}, 2\bar{X})} - 1} \quad (34)$$

where $\bar{\Theta}$ and \bar{X} are positive integers, $|\Theta| \leq \bar{\Theta} < |\Theta| + 1$, $|X| \leq \bar{X} < |X| + 1$. The error incurred by setting $N = 20 + \text{Max}(2\bar{\Theta}, 2\bar{X})$ is estimated by Equation (34) to be

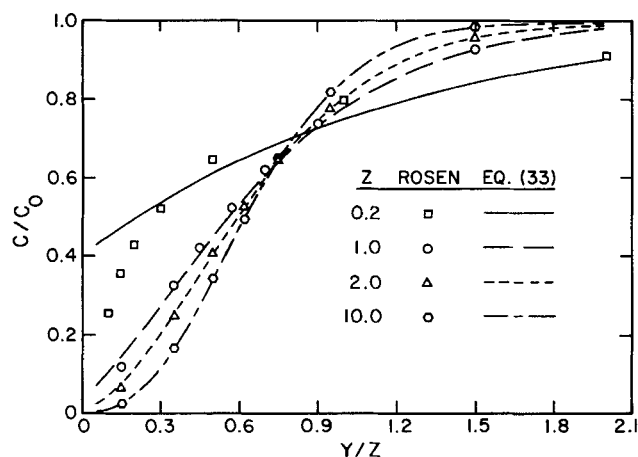


Fig. 4. Breakthrough curves for selected values of bed length Z with film resistance parameter $\nu/Z = 0.1$.

$$R_N/u_N < 1/2^{19} = 1.9 \times 10^{-6}$$

Equation (33) is the working formula expressing concentration of the adsorbate in the fluid as a function of time and axial distance. It is well suited for computer application for all values of Θ and X .

RESULTS AND DISCUSSION

The breakthrough curve of an adsorption column in response to a step change in input calculated from Equation (33) is now compared with those by Rosen (1954). The comparison requires that the breakthrough curves from the two works be expressed in terms of the same dimensionless variables. Rosen presented his results in terms of the following dimensionless variables:

$$\nu = Dk/Rk_f \quad (35)$$

$$Z = 3Dkz/\eta\nu R^2 \quad (36)$$

$$Y = 2D(t - z/\nu)/R^2 \quad (37)$$

For a given value of ν/Z , the effluent concentration is presented as a function of the ratio Y/Z , with Z as a parameter. The ratios Y/Z and ν/Z can be interpreted as dimensionless time and film resistance, respectively.

In this work, the effluent concentration is expressed by Equation (33) as a function of Θ and X . These variables can be expressed in terms of Rosen's Z , ν/Z , and Y/Z as follows:

$$\Theta = \frac{-15(Y/Z)}{2[5(\nu/Z) + (1/Z)]} \quad (38)$$

$$X = \frac{-5}{5(\nu/Z) + (1/Z)} \quad (39)$$

Thus, at all conditions studied by Rosen, we can compute Θ and X by Equations (38) and (39) and then the effluent concentration by Equation (33).

Figures 1 through 5 show the comparison of our results with Rosen's tabulated values. Each figure is for a specific value of ν/Z (dimensionless film resistance) given by Rosen; the five figures exhaust all his tabulated ν/Z values.

The curves show the effluent concentration as a function of time (Y/Z) at a specified axial distance (Z).

In all the figures, excellent agreement is obtained at all times for large values of axial distance. The agreement gradually falls off as axial distance is reduced, but remains quantitative as long as the breakthrough curve retains its typical S shape. At small axial distances, the break-

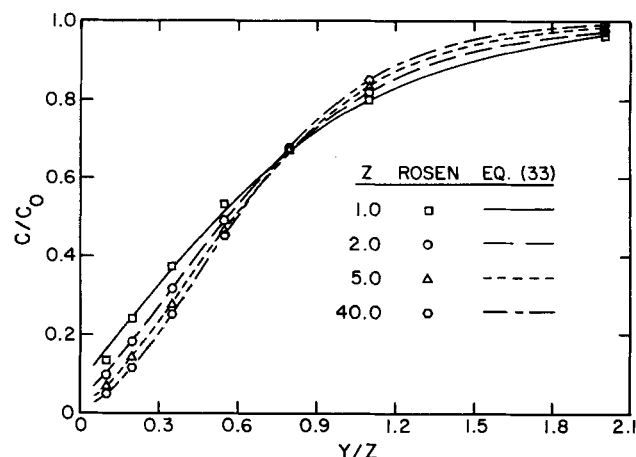


Fig. 5. Breakthrough curves for selected values of bed length Z with film resistance parameter $\nu/Z = 0.2$.

through curves are not S shaped, and there is a rapid buildup of effluent concentration after a short on-stream time. At these conditions, the parabolic approximation may not be accurately representing the concentration gradient at the particle surface, leading to discrepancy with the rigorous solution. The widely used Thomas (1944) solution also diverges from Rosen for shallow beds. However, these conditions are not likely encountered in real adsorber operation and design.

It appears that Equation (33) can be recommended for the calculation of effluent concentration of industrial adsorbers in place of Rosen's complex function. The new result is simpler and is well suited for application on computers.

NOTATION

- a_0, a_2 = parameters in Equation (6)
- c = concentration of adsorbate in fluid phase, (gmole/cm³)
- c_0 = concentration of adsorbate in feed stream, (gmole/cm³)
- D = effective diffusivity inside the solid particle, (cm²/s)
- k = mass transfer coefficient
- k_f = particle to fluid mass transfer coefficient, (cm/s)
- K = adsorption equilibrium constant = q/c
- q = concentration of adsorbate in solid phase, (gmole/cm³ particle)
- q_R = concentration of adsorbate at the particle surface, (gmole/cm³ particle)
- r = radial distance from center of particle
- s = frequency
- t = time variable, (s)
- u = c/c_0 = reduced concentration, dimensionless
- U = Laplace transform of u
- v = interstitial velocity, (cm/s)
- x = $z/\eta\nu$, (s)
- X = $-Kx/\xi$, dimensionless
- \bar{X} = integer, $|X| \leq \bar{X} \leq |X| + 1$
- Y = $2D(t - z/\nu)/R^2$, Rosen's time parameter
- z = bed length, (cm)
- Z = $3Dkz/\eta\nu R^2$, Rosen's bed length parameter

Greek Letters

- ϵ = void fraction of the packed bed, dimensionless
- η = $\epsilon/1 - \epsilon$, dimensionless
- θ = $t - z/v$, (cm)
- Θ = $-\theta/\xi$, dimensionless
- $\bar{\Theta}$ = integer, $|\Theta| \leq \bar{\Theta} \leq |\Theta| + 1$

$\nu = DK/Rk_f$, Rosen's film resistance parameter
 $\xi = R(K/3k_f + R/15D)$, (s)

LITERATURE CITED

- Blasius, H., "Grenzschichten in Flüssigkeiten unit kleiner Reibung," *Z. Math. u. Phys.*, **56**, 1 (1908); *NACA Tech. Memo. No. 1256*.
 Edeskuty, F. J., and N. R. Amundson, "Effect of Intraparticle Diffusion," *Ind. Eng. Chem.*, **44**, 1968 (1952).
 Glueckauf, E., "Theory of Chromatography. Part 10—Formula for Diffusion into Spheres and Their Application to Chromatography," *Trans. Faraday Soc.*, **51**, 1540 (1955).
 Liaw, C. H., "Liquid Phase Adsorption in Fixed Beds," Ph.D. dissertation, Purdue Univ., West Lafayette, Ind. (1977).
 Masamune, S., and J. M. Smith, "Adsorption Rate Studies—Interaction of Diffusion and Surface Processes," *AIChE J.*, **11**, 34 (1965).
 Rosen, J. B., "Kinetics of a Fixed Bed System for Solid Diffusion into Spherical Particles," *J. Chem. Phys.*, **20**, 387 (1952).
 ———, "General Numerical Solution for Solid Diffusion in Fixed Beds," *Ind. Eng. Chem.*, **46**, 1590 (1954).
 Thomas, H. C., "Heterogeneous Ion Exchange in a Flowing System," *J. Am. Chem. Soc.*, **66**, 1664 (1944).
 Vermeulen, T., G. Klein, and N. K. Hiester, "Adsorption and Ion Exchange," in *Chemical Engineers' Handbook*, R. H. Perry and C. M. Chilton, ed., sect. 16, McGraw-Hill, New York (1973).
 Von Karman, Th., "Ueber Laminare and Turbulente Reibung," *Z. Angew. Math. Mech.*, **1**, 233 (1921).

APPENDIX A: CONVERGENCY OF EQUATION (28)

Absolute convergence implies convergence, as

$$\sum_n |a_n| \text{ converge} \rightarrow \sum_n a_n \text{ converge}$$

$$\sum_{n=0}^{\infty} \sum_{i=0}^n \frac{|X|^{n+1} |\theta|^i}{(n+1)(n-i)! (i!)^2} \leq \sum_{n=0}^{\infty} \sum_{i=0}^n \frac{|X|^{n+1} n! |\theta|^i}{(n+1)! (n-i)! i!} \quad (\text{A1})$$

By use of binomial series

$$(1+A)^n = 1 + nA + \frac{n(n-1)}{2!} A^2 + \dots = \sum_{i=0}^n \frac{n!}{(n-i)! i!} A^i \quad (\text{A2})$$

$$\begin{aligned} \sum_{n=0}^{\infty} \sum_{i=0}^n \frac{|X|^{n+1} n! |\theta|^i}{(n+1)! (n-i)! i!} &= \sum_{n=0}^{\infty} \frac{|X|^{n+1}}{(n+1)!} (1+|\theta|)^n \\ &= \sum_{n=0}^{\infty} \frac{[|X|(1+|\theta|)]^{n+1}}{(n+1)! (1+|\theta|)} \\ &= \frac{1}{1+|A|} \{ \exp [|X|(1+|\theta|)] - 1 \} \end{aligned} \quad (\text{A3})$$

where the series expression of $\exp [|X|(1+|\theta|)]$ is used. From (A3), it can be seen that Equation (28) converges everywhere and rapidly!

APPENDIX B: DERIVATION OF EQUATION (32)

In Equation (28), let

$$f(\theta, X) = \sum_{n=0}^{\infty} \sum_{i=0}^n \frac{X^{n+1} \theta^i}{(n+1)(n-i)! (i!)^2} \quad (\text{B1})$$

Equation (B1) can be summarized over i from zero to infinite, which gives

$$f(\theta, X) = \sum_{i=0}^{\infty} \frac{\theta^i}{(i!)^2} \sum_{n \geq i} \frac{X^{n+1}}{(n-i)! (n+1)!} \quad (\text{B2})$$

Let

$$g(x) = \sum_{n \geq i} \frac{X^{n+1}}{(n-i)! (n+1)!} \quad (\text{B3})$$

It can be seen that $g(0) = 0$. The derivative of $g(X)$ gives

$$\frac{dg(X)}{dX} = \sum_{n \geq i} \frac{X^n}{(n-i)!} = \sum_{n \geq i} \frac{X^{n-i}}{(n-i)!} X^i \quad (\text{B4})$$

Applying the equality $e^X = \sum_{i=0}^{\infty} (X^i/i!)$ to Equation (C5), we get

$$\frac{dg(X)}{dX} = X^i e^X \quad (\text{B5})$$

Integrating Equation (B5) and recalling that $g(0) = 0$, we get

$$g(X) = e^X \left[\frac{X^i}{i!} - \frac{X^{i-1}}{(i-1)!} + \dots + (-1)^i i! - (-1)^i i! \right] \quad (\text{B6})$$

Substitution of $g(X)$ from Equation (B6) into Equation (B2) gives

$$\begin{aligned} f(\theta, X) &= \sum_{i=0}^{\infty} \frac{\theta^i}{(i!)^2} g(X) \\ &= -e^{-\theta} + e^X \sum_{i=0}^{\infty} \left[\frac{X^i}{i!} - \frac{X^{i-1}}{(i-1)!} + \dots + (-1)^i \right] \frac{\theta^i}{i!} \end{aligned} \quad (\text{B7})$$

Combination of Equations (B7), (B1), and (28) gives

$$\begin{aligned} u(x, \theta) &= 1 + e^{\theta} f(\theta, x) \\ &= e^{\theta+X} \sum_{i=0}^{\infty} \left[\frac{X^i}{i!} - \frac{X^{i-1}}{(i-1)!} + \dots + (-1)^i \right] \frac{\theta^i}{i!} \end{aligned} \quad (\text{B8})$$

Recall that both θ and X are negative; therefore, $X = -|X|$ and $\theta = -|\theta|$, and Equation (B8) can be written as

$$u(x, \theta) = e^{\theta+X} \sum_{i=0}^{\infty} (-1)^{2i} \left[\frac{|X|^i}{i!} + \frac{|X|^{i-1}}{(i-1)!} + \dots + 1 \right] \frac{|\theta|^i}{i!}$$

or

$$u(x, \theta) = e^{\theta+X} \sum_{i=0}^{\infty} \left[\frac{|X|^i}{i!} + \frac{|X|^{i-1}}{(i-1)!} + \dots + 1 \right] \frac{|\theta|^i}{i!} \quad (\text{B9})$$

It can be seen from Equation (B9) that $u(x, \theta) \geq 0$, and it can be also shown that $u(x, \theta) \leq 1$ as follows:

$$\begin{aligned} |u(x, \theta)| &= \left| e^{\theta+X} \sum_{i=0}^{\infty} \left(\frac{|X|^i}{i!} + \frac{|X|^{i-1}}{(i-1)!} + \dots + 1 \right) \frac{|\theta|^i}{i!} \right| \\ &\leq e^{\theta+X} \sum_{i=0}^{\infty} e^{|X|} \frac{|\theta|^i}{i!} \end{aligned}$$

$$= e^{\theta+X} e^{|X|} e^{|\theta|}$$

$$= e^0 = 1$$

Equation (B9) can be written in compact form as

$$u(x, \theta) = e^{\theta+X} \sum_{i=0}^{\infty} \left[\sum_{j=0}^i \frac{|X|^j}{j!} \right] \frac{|\theta|^i}{i!} \quad (\text{B10})$$

by use of the following equality:

$$\frac{|M|^j}{j!} = \prod_{k=1}^j \left(\frac{|M|}{k} \right)$$

Equation (B8) can be written as

$$u(x, \theta) = e^{\theta+X} \left[1 + \sum_{i=1}^{\infty} \left[\prod_{k=1}^i \frac{|\theta|}{k} \right] \left[1 + \sum_{j=1}^i \prod_{m=1}^j \frac{|X|}{m} \right] \right] \quad (32)$$

APPENDIX C: REMAINDER OF EQUATION (32)

$$u(x, \theta) = e^{\theta+X} \left[1 + \sum_{i=1}^{\infty} \left[\prod_{k=1}^i \frac{\theta}{k} \right] \left[1 + \sum_{j=1}^i \prod_{m=1}^j \frac{|X|}{m} \right] \right] \quad (32)$$

$$= e^{\theta+X} \sum_{i=0}^{\infty} \left[\frac{|X|^i}{i!} + \frac{|X|^{i-1}}{(i-1)!} + \dots + 1 \right] \frac{|\theta|^i}{i!}$$

$$= u_N + R_N \quad (\text{B9})$$

where

$$u_N = e^{\theta+X} \sum_{i=0}^N \left[\frac{|X|^i}{i!} + \frac{|X|^{i-1}}{(i-1)!} + \dots + 1 \right] \frac{|\theta|^i}{i!} \quad (\text{C1})$$

Since

$$e^{|X|} = \sum_{i=0}^{\infty} \frac{|X|^i}{i!}$$

therefore

$$R_N \leq e^{\theta+X} e^{|X|} \left[\frac{|\theta|^{N+1}}{(N+1)!} + \frac{|\theta|^{N+2}}{(N+2)!} + \dots \right]$$

$$= e^{\theta+X} e^{|X|} \frac{|\theta|^N}{N!} \left[\frac{|\theta|}{N+1} + \frac{|\theta| \cdot |\theta|}{(N+1)(N+2)} + \dots \right]$$

Select $N \geq \max(2\theta, 2X)$. Then

$$R_N \leq e^{\theta+X} e^{|X|} \frac{|\theta|^N}{N!} \left[\frac{1}{2} + \frac{1}{2} \cdot \frac{1}{2} + \frac{1}{2} \cdot \frac{1}{2} \cdot \frac{1}{2} + \dots \right]$$

$$= e^{\theta+X} e^{|X|} \frac{|\theta|^N}{N!}$$

(i) If $|\theta| \leq |X|$, consider the $2\bar{X}$ th term, $t_{2\bar{X}}$ in u_N (where \bar{X} is a positive integer) $|X| \leq \bar{X} < |X| + 1$:

$$t_{2\bar{X}} = e^{\theta+X} \left[\frac{|X|^{2\bar{X}}}{(2\bar{X})!} + \frac{|X|^{2\bar{X}-1}}{(2\bar{X}-1)!} + \dots + 1 \right] \frac{|\theta|^{2\bar{X}}}{(2\bar{X})!} \quad (\text{C2})$$

$$e^{|X|} = 1 + |X| + \frac{|X|^2}{2!} + \dots + \frac{|X|^{2\bar{X}}}{(2\bar{X})!} + Q_{2\bar{X}}^-$$

$$Q_{2\bar{X}}^- = \frac{|X|^{2\bar{X}}}{(2\bar{X})!} \left[\frac{|X|}{2\bar{X}+1} + \frac{|X|^2}{(2\bar{X}+2)(2\bar{X}+1)} + \frac{|X|^3}{(2\bar{X}+3)(2\bar{X}+2)(2\bar{X}+1)} + \dots \right]$$

$$< \frac{|X|^{2\bar{X}}}{(2\bar{X})!} \left(\frac{1}{2} + \frac{1}{4} + \frac{1}{8} + \dots \right)$$

$$= \frac{|X|^{2\bar{X}}}{(2\bar{X})!}$$

(ii) If $|\theta| \geq |X|$, consider the $2\bar{\theta}$ th term $t_{2\bar{\theta}}$ in u_N (where $\bar{\theta}$ is a positive integer) $|\theta| \leq \bar{\theta} < |\theta| + 1$:

$$t_{2\bar{\theta}} = e^{\theta+X} \left[\frac{|X|^{2\bar{\theta}}}{(2\bar{\theta})!} + \frac{|X|^{2\bar{\theta}-1}}{(2\bar{\theta}-1)!} + \dots + 1 \right] \frac{|\theta|^{2\bar{\theta}}}{(2\bar{\theta})!}$$

and

$$e^{|X|} = 1 + |X| + \dots + \frac{|X|^{2\bar{\theta}}}{(2\bar{\theta})!} + Q_{2\bar{\theta}}^-$$

$$Q_{2\bar{\theta}}^- = \frac{|X|^{2\bar{\theta}}}{(2\bar{\theta})!} < 1 + X + \dots + \frac{|X|^{2\bar{\theta}}}{(2\bar{\theta})!}$$

Similarly, we can obtain

$$\frac{R_N}{u_N} < \left(\frac{1}{2} \right)^{N-2\bar{\theta}-1} \quad (\text{C3})$$

Combination of (C5) and (C3) gives an expression for the estimation of remainder:

$$\frac{R_N}{u_N} < \left(\frac{1}{2} \right)^{N-\max(2\bar{\theta}, 2\bar{X})-1} \quad (34)$$

that is

$$Q_{2\bar{X}}^- < \frac{|X|^{2\bar{X}}}{(2\bar{X})!} < 1 + |X| + \dots + \frac{|X|^{2\bar{X}}}{(2\bar{X})!}$$

Therefore

$$\frac{1}{2} e^{|X|} < 1 + |X| + \dots + \frac{|X|^{2\bar{X}}}{(2\bar{X})!} \quad (\text{C4})$$

From (C2) and (C4)

$$t_{2\bar{X}}^- > \frac{1}{2} e^{\theta+X} e^{|X|} \frac{|\theta|^{2\bar{X}}}{(2\bar{X})!}$$

Consider the ratio of remainder to u_N . Since $u_N > t_{2\bar{X}}^-$, we obtained

$$\frac{R_N}{u_N} < \frac{R_N}{t_{2\bar{X}}^-} < \frac{e^{\theta+X} e^{|X|} \frac{|\theta|^N}{N!}}{\frac{1}{2} e^{\theta+X} e^{|X|} \frac{|\theta|^{2\bar{X}}}{(2\bar{X})!}} = 2 \left[\frac{\theta^{N-2\bar{X}}}{N!/(2\bar{X})!} \right]$$

$$= 2 \left(\frac{\theta}{2\bar{X}+1} \right) \left(\frac{\theta}{2\bar{X}+2} \right) \dots \left(\frac{\theta}{N} \right)$$

$$< 2 \left(\frac{1}{2} \right)^{N-2\bar{X}}$$

Therefore

$$\frac{R_N}{u_N} < \left(\frac{1}{2} \right)^{N-2\bar{X}-1} \quad (\text{C5})$$

Manuscript received February 20, 1978; revision received August 2, and accepted August 25, 1978.