

## LITERATURE CITED

1. Beenakker, J. J. M., B. van Eijnsbergen, M. Knoester, K. W. Taconis, and P. Zandbergen, "Advances in Thermophysical Properties at Extreme Temperature and Pressure," p. 114, Am. Soc. Mech. Engrs., New York (1965).
2. de Boer, J., and A. Michels, *Physica*, **5**, 945 (1938).
3. de Boer, J., *ibid.*, **14**, 139 (1948).
4. ———, and B. S. Blaisse, *ibid.*, **14**, 149 (1948).
5. de Boer, J., and R. J. Lunbeck, *ibid.*, **14**, 520 (1948).
6. Canfield, F. B., T. W. Leland, and Riki Kobayashi, *J. Chem. Eng. Data*, **10**, 92 (1965).
7. Curl, R. F., Jr., and K. S. Pitzer, *Ind. Eng. Chem.*, **50**, 265 (1958).
8. Dean, J. W., *Natl. Bureau Standards Tech. Note* 120, (Nov., 1961).
9. Johnston, H. L., and D. White, *Trans. Am. Soc. Mech. Engrs.*, **70**, 651 (1948).
10. Jones, M. L., Jr., D. T. Mage, R. C. Faulkner, Jr., and D. L. Katz, *Chem. Eng. Progr. Symposium Ser. No. 44*, **59**, 52 (1963).
11. Kay, W. B., *Ind. Eng. Chem.*, **28**, 1014 (1936).
12. Leland, T. W., Jr., Patsy S. Chapplear, and J. W. Leach, *U.S. Bureau Mines Rept.* (Apr., 1965).
13. Mage, D. T., and D. L. Katz, *A.I.Ch.E. J.*, **12**, 137 (1966).
14. Mann, D. B., *Natl. Bureau Standards Tech. Note* 154A (1962).
15. Michels, A., J. M. Levelt, and G. J. Wolkers, *Physica*, **25**, 1097 (1959).
16. Mueller, W. H., T. W. Leland, Jr., and Riki Kobayashi, *A.I.Ch.E. J.*, **7**, 267 (1961).
17. Newton, R. H., *Ind. Eng. Chem.*, **27**, 302 (1935).
18. Pfenning, D. B., and F. B. Canfield, *Document No. 7864*, Am. Documentation Inst., Lib. Congr., Washington, D. C.
19. Pitzer, K. S., *J. Am. Chem. Soc.*, **77**, 3427 (1955).
20. ———, D. Lippman, R. F. Curl, Jr., C. M. Huggins, and D. E. Petersen, *ibid.*, 3433 (1955).
21. Prausnitz, J. M., and R. D. Gunn, *A.I.Ch.E. J.*, **4**, 494 (1958).
22. Reid, R. C., and T. W. Leland, Jr., *ibid.*, **11**, 228 (1965).
23. Satter, A., and J. M. Campbell, *Soc. Petrol. Engrs. J.*, **3**, 333 (1963).

Manuscript received April 19, 1966; revision received June 16, 1966; paper accepted June 20, 1966.

# Statistical Models for Surface Renewal in Heat and Mass Transfer: Part I. Dependence of Average Transport Coefficients on Age Distribution

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Average transport coefficients for different transport models are computed by using widely varying distributions of residence times of elements at the transporting surface. It is shown that for typical transport models, the shapes of the residence time and age distributions have an insignificant effect on the average transport coefficient. Operational techniques for calculating average coefficients and for constructing delayed versions of age distributions are presented.

## DISCUSSION OF PREVIOUS WORK

The concept of surface renewal as a model for heat and mass transfer was first introduced by Higbie (3). In this early work, Higbie visualized heat transport as occurring by the arrival at the transport surface of fresh transporting elements from the bulk fluid, followed by unsteady state transport by conduction or diffusion during the residence of the fluid element at the surface, and eventual replacement of the stale element by a fresh fluid element. Higbie assumed that the fluid elements which arrived at the transporting surface had identical residence times. Danckwerts (2) later extended Higbie's work by assuming an age distribution of surface fluid elements with an exponential form. This is equivalent to the assumption that the probability of replacement of a surface fluid element is independent of its age. Zwieterling (8) derived a general relation between the age distribution of elements at the surface and the residence time distribution of elements which are about to enter the transporting surface. A constant total flow of elements to and from the surface was assumed in making this derivation. Perlmutter (5) investigated the effects of different age distribu-

tions upon the calculated average transport coefficients, and introduced the concept of a dead time effect resulting from stagnant pockets at the surface.

The present work consists of two parts: In Part I we review and systemize this earlier work, make appropriate modifications in the derivations to extend validity to transport in systems where a constant total flow cannot be assumed (such as in fluidized beds), point out some inconsistencies in interpretation of this previous work, justify the conclusion that for much of the work in transport phenomena only the average residence time (and not the shape of the residence time distribution) is needed for prediction of transport coefficients, and finally apply the results to an example situation which involves prediction of heat transfer coefficients at surfaces in a fluidized bed. In Part II, two simple but accurate statistical techniques are derived for computing the average residence time from information only on the total number of tagged elements present at the transporting surface as a function of time. (Interest in the average residence time only is justified by Part I.) The method is applied both to computer-simulated residence time distributions and to experimental data generated by a system of black and white fluidized particles. For exceptional cases wherein knowledge of only the average residence time is insufficient, a more

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general technique is derived for estimating the residence time distribution. This is applicable to systems in which the form of the instantaneous transport coefficient leads to a sensitive dependence of the average transport coefficient on the shape of the residence time distribution. This method is also based upon use of only the total number of tagged elements present vs. time, and is applicable to arbitrary shape of residence time distribution. Application to the computer-simulated and fluidized-bed systems is given.

## RELATION BETWEEN RESIDENCE AND AGE DISTRIBUTION

Since a major objective of the present work is a technique applicable to a variety of systems, including fluidized beds, the assumption of constant flow into and out of the transporting surface is unduly restrictive. In the present section, we derive Zwieterling's relationship between residence time and age distributions without making this assumption. Conforming as much as possible to notations used by previous investigators, we define the residence time distribution  $f(\tau)$ , so that  $f(\tau)d\tau$  is the fraction of all elements entering the transporting surface which will have residence times between  $\tau$  and  $\tau + d\tau$  at that surface. Similarly,  $F(\tau)$  is defined as the cumulative distribution resulting from the density function  $f(\tau)$ . The age distribution function  $\phi(a)$  is defined so that  $\phi(a)da$  is the fraction of all elements present at the surface at any given time, which are of ages between  $a$  and  $a + da$ .

Now it is clear that an element of age  $a$  corresponds to one whose residence time will be at least  $a$ . If we hypothesize that the fraction of elements of age  $a$  at the surface at any instant is proportional to the probability that an element entering at any instant will attain a residence time of at least  $a$ , then the resulting relation is

$$\phi(a)da = \beta[1 - F(a)]da$$

where  $\beta da$  is the constant of proportionality. We must choose  $\beta$  so that  $\phi(a)$  has an area of unity:

$$\int_0^\infty \phi(a)da = 1$$

This restriction may immediately be used to calculate the necessary value of  $\beta$  and results in

$$\phi(a) = \frac{1 - F(a)}{\bar{\tau}} \quad (1)$$

where  $\bar{\tau}$  is the first moment of  $f(\tau)$ , that is, the average residence time; this is identical to Zwieterling's relationship. Note that in the derivation of Equation (1) it is necessary to assume

$$\lim_{\tau \rightarrow \infty} \tau^2 f(\tau) = 0$$

This restriction is met by most residence time distribution functions; in fact, the existence of a second moment for the residence time distribution function is sufficient (but not necessary) to guarantee that this restriction will be satisfied. Existence of the mean  $\bar{\tau}$  is necessary and sufficient. The existence of a finite average residence time  $\bar{\tau}$  will be tacitly assumed throughout the present work. Admittedly, for some special cases such as a particularly fluidized bed (usually a liquid-solid system), such an assumption may be invalid (6). However, it should be valid for the great majority of heat and mass transfer processes.

## BASIC MODELS FOR RESIDENCE TIME DISTRIBUTIONS

### Gamma Model

Physically appealing models for the residence time distribution may be generated by the gamma form:

$$f(\tau; \bar{\tau}) = \frac{s^{\alpha+1}}{\alpha!} \tau^\alpha e^{-s\tau} u(\tau) \quad (2)$$

where

$$s = \frac{\alpha + 1}{\bar{\tau}} \quad (3)$$

and  $u(\tau)$  is the unit step function, used to indicate that negative values of residence time are not allowed. Note that the parameter  $\bar{\tau}$  is included as an argument of the residence time distribution, as well as of the age distribution to be discussed below. This is to avoid the misinterpretation of the effects of model parameters upon average transport coefficients which was made in earlier work. Figure 1 is a dimensionless plot of  $f(\tau; \bar{\tau})$  and shows that the shape of the gamma form agrees with one's physical intuition for the shape of the residence time distribution.

The gamma model offers the following advantages for representation of residence time distributions: (1) the residence times are defined over an interval from zero to infinity; (2) as shown in Figure 1, great changes in the shape of the residence time distribution may be effected by varying the parameter  $\alpha$ ; and (3) this distribution will be shown below to offer convenient operational properties for calculating the average transport coefficient.

Figure 1 effectively shows the dependence of the gamma distribution upon the shape parameter  $\alpha$  at a constant value of the average residence time  $\bar{\tau}$ . The value  $\alpha = 0$  corresponds to the distribution suggested by Danckwerts, and the value  $\alpha = \infty$  corresponds to that suggested by Higbie. Values of  $\alpha$  between these extremes give residence time distributions which pass through a peak and decay to zero. It is permissible for  $\alpha$  to take on all values greater than  $-1$ . Values of  $\alpha$  less than 0 lead to residence time distributions which are infinite at zero time. However, the age distributions resulting from these residence time distributions are quite acceptable; there are no physical inconsistencies in having  $f(0)$  approach infinity.

To calculate the age distribution for the gamma model, we use Equations (1) and (2), which give for  $\alpha$  zero or a positive integer:

$$\phi(a; \bar{\tau}) = \frac{se^{-sa}}{\alpha + 1} \sum_{i=0}^{\alpha} \frac{(sa)^i}{i!} \quad (4)$$

For negative or nonintegral values of  $\alpha$ , the integration of  $f(\tau)$  required in the calculation of  $\phi(a)$  is not easily

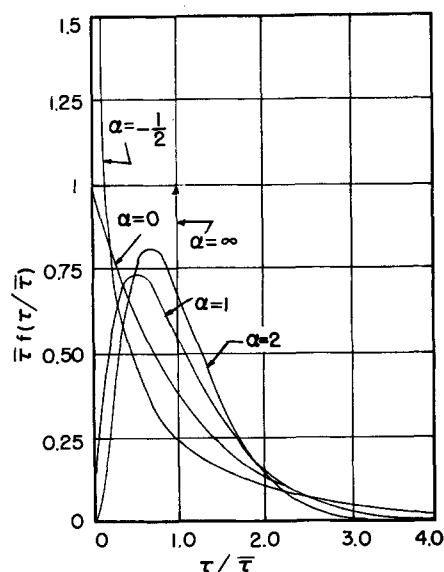


Fig. 1. Gamma distribution for various  $\alpha$ .

performed, with the exception of the value  $\alpha = -\frac{1}{2}$  for which the result is

$$\phi(a; \bar{\tau}) = \frac{1}{\bar{\tau}} \operatorname{erfc} \left[ \sqrt{\frac{a}{\bar{\tau}}} \right]; \alpha = -\frac{1}{2} \quad (5)$$

These age distributions are plotted for various values of the shape parameter  $\alpha$  in Figure 2. Note that as  $\alpha \rightarrow -1$ , the age distribution of Figure 2 will approach a pulse function concentrated around an age of zero, but with a long tail at high values of age. Figure 2 shows that the effect of decreasing  $\alpha$  is to increase the proportion of old elements at the surface at any given instant of time. The case  $\alpha = \infty$ , corresponding to Higbie's assumptions, leads to a uniform distribution of ages of elements at the surface.

#### Dead Time Model

Perlmutter (5) observed that the presence of stagnant fluid elements at the transporting surface would introduce a different form to the residence time distribution. In particular, he introduced the concept of a delayed version of the exponential distribution, resulting from stagnant pockets at the surface. An alternate interpretation of the presence of a dead time in the residence time distribution is that there is a minimum residence time for all fluid elements. (This is similar to the mixing delay which occurs in a real agitated vessel. While in the ideal case it is possible for a fluid element to reach the outlet pipe of a perfectly stirred vessel instantaneously after its entrance to the vessel, this obviously cannot happen in a real stirred vessel.)

We present here general relations for constructing delayed residence time and age distributions from originally nondelayed models. A delayed residence time distribution may be constructed from any undelayed model by the relation

$$f_d(\tau; \bar{\tau}) = u(\tau - \bar{\tau}d) f[\tau - \bar{\tau}d; \bar{\tau}(1-d)] \quad (6)$$

where  $f_d(\tau; \bar{\tau})$  is the delayed residence time distribution which has the same average residence time  $\bar{\tau}$  as does the undelayed residence time distribution  $f(\tau; \bar{\tau})$ . The quantity  $d$  is the dead time expressed as a fraction of the average residence time  $\bar{\tau}$ , and  $u(\tau)$  is the unit step function. It is easily verified that the first moment of the distribution defined by Equation (6) is indeed  $\bar{\tau}$ . The major difference between this relation and that of Perlmutter is that in Equation (6) we have insisted that, regardless of the amount of dead time to be added to the model, the average residence time must remain constant. The reason for this is that the average residence time is a fundamental property of the time scale for the transport process. It will be essential to be able to separate the dependence of the average transport coefficients on the average residence time from the dependence upon the shape of the age distribution.

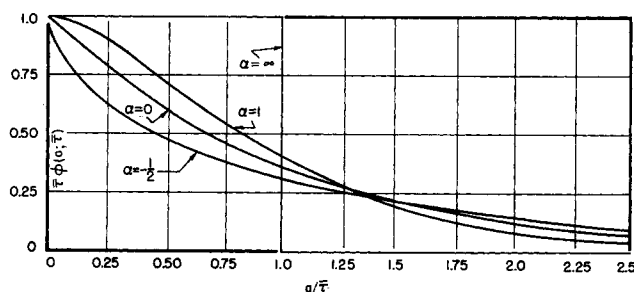


Fig. 2. Age distributions resulting from gamma residence time distribution.

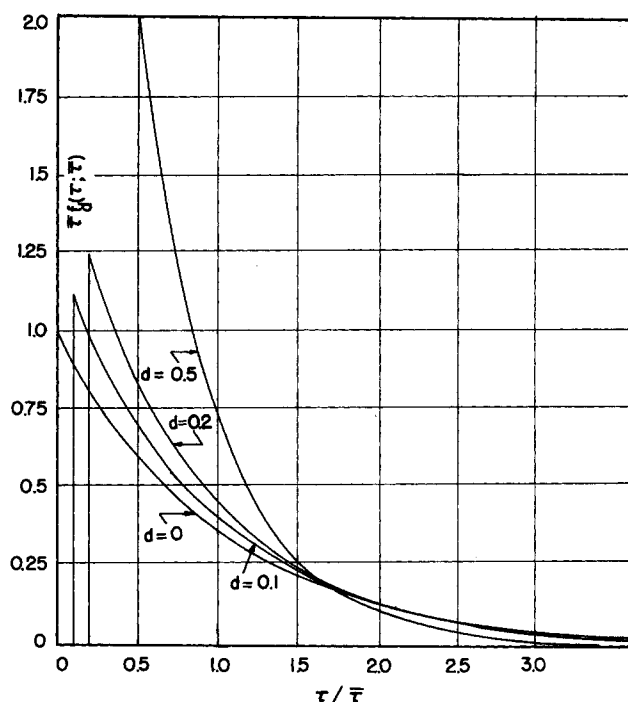


Fig. 3. Delayed exponential distribution for various  $d$ .

Examples of the delayed distribution resulting from the exponential case are shown in Figure 3 for varying amounts of the dead time  $\bar{\tau}d$ . Note that as  $d$  approaches unity, the residence time distribution approaches a unit impulse located at  $\tau = \bar{\tau}$ , which shows that the distribution becomes identical to that used by Higbie, as must be true from the physical considerations. Note also that  $d$  may never exceed unity, because it would then be impossible to have an average residence time of  $\bar{\tau}$ . The advantage of using a delayed model for the residence time distribution is that such a model may be more descriptive of the physical situation than, for example, any of the distributions shown in Figure 1.

To calculate the age distribution resulting from the delayed residence time distribution, we apply Equation (1) to Equation (6), which results in

$$\phi_d(a; \bar{\tau}) = \frac{u(a) - F[a - \bar{\tau}d; \bar{\tau}(1-d)]}{\bar{\tau}} \quad (7)$$

As an example, for the exponential case considered by Perlmutter, the result of Equation (7) takes the specific form

$$\phi_d(a; \bar{\tau}) = \frac{1}{\bar{\tau}} \left[ u(a - \bar{\tau}d) \exp\left(\frac{d - a/\bar{\tau}}{1-d}\right) + u(a) - u(a - \bar{\tau}d) \right] \quad (8)$$

Equation (8) is plotted for various amounts of dead time in Figure 4. Note that for larger amounts of dead time, there is a smaller fraction of old elements present at the surface, as must be true in order to maintain a constant average residence time.

#### CALCULATION OF AVERAGE TRANSPORT COEFFICIENTS FROM AGE DISTRIBUTION

##### Basic Definition

For each element of fluid which approaches the transporting surface, there exists a function  $N_{Nu}(a)$  which expresses the dependence of the instantaneous transport

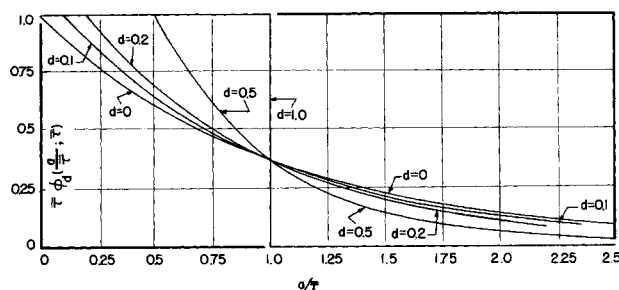


Fig. 4. Age distributions resulting from delayed exponential residence time distribution.

coefficient between the surface and that fluid element upon the age of the fluid element. The average transport coefficient  $\bar{N}_{Nu}$  is obtained by averaging the instantaneous transport coefficients over the entire surface, which in this case corresponds to averaging over the age distribution. Therefore

$$\bar{N}_{Nu}(\bar{\tau}) = \int_0^\infty \phi(a; \bar{\tau}) N_{Nu}(a) da \quad (9)$$

Note that we have indicated in Equation (9) that the average transport coefficient  $\bar{N}_{Nu}$  depends upon the average age residence time  $\bar{\tau}$ .

#### Calculation for Gamma Model

The following theorem holds for the Laplace transform operation when applied to functions which are sectionally continuous and possess a transform:

$$\mathcal{L}\{t^i h(t)\} = (-1)^i \frac{d^i h(p)}{dp^i} \quad (10)$$

where  $h(p)$  is the Laplace transform of the function  $h(t)$ . It then follows from Equations (4), (9), and (10) that for  $\alpha$  zero or a positive integer

$$\bar{N}_{Nu}(\bar{\tau}) = \sum_{i=0}^{\alpha} \frac{(-1)^i s^i}{\bar{\tau}!} \left\{ \frac{d^i}{dp^i} [N_{Nu}(p)] \right\}_{p=s} \quad (11)$$

where  $N_{Nu}(p)$  is the Laplace transform of the instantaneous transport coefficient  $N_{Nu}(a)$ . For other values of  $\alpha$ , it is necessary to calculate the average transport coefficient from the basic definition. Danckwerts (2) presented Equation (11) for the special case  $\alpha = 0$ . The result of Equation (11) represents a particularly convenient property of the gamma distribution; that is, from the Laplace transform of the instantaneous coefficient one can calculate, *without inversion*, the time average transport coefficient. Since the instantaneous rate coefficient  $N_{Nu}(a)$  will often be obtained as the solution to partial differential equations, and this will frequently be done through use of Laplace transforms, it is convenient not to have to invert the transform to obtain the desired average transport coefficient.

#### Operational Calculation of $\bar{N}_{Nu}$ for Arbitrary Age Distribution

Since the age distribution function  $\phi(a)$  is necessarily sectionally continuous, it may be represented by an expansion in orthogonal functions. The Laguerre functions are suitable for this expansion because they are defined over the appropriate interval, zero to infinity. The Laguerre functions are defined according to the relation

$$L_n(a) = u(a) \frac{e^{a/2}}{n!} \frac{d^n}{da^n} (a^n e^{-a}) \quad (12)$$

The expansion of an arbitrary distribution  $\phi(a; \bar{\tau})$  takes the form

$$\phi(a; \bar{\tau}) = \sum_{n=0}^{\infty} c_n(\bar{\tau}) L_n(a) \quad (13)$$

where the coefficients  $c_n(\bar{\tau})$  are calculated according to the relation

$$c_n(\bar{\tau}) = \int_0^\infty \phi(a; \bar{\tau}) L_n(a) da \quad (14)$$

Since the Laguerre functions involve only products of exponentials and polynomials, a form such as Equation (11) can always be found for calculation of the average transport coefficient from the expansion of the chosen age distribution. Therefore, in principle, one need never invert the Laplace transform of the instantaneous rate coefficient to calculate the average transport coefficient. However, for practical considerations it is necessary that only a few terms of the infinite series of Equation (13) be required to represent suitably the age distribution to render Equation (11) computationally useful.

#### AVERAGE TRANSPORT COEFFICIENTS FOR BASIC PENETRATION MODEL

In the basic concept of penetration theory, a fluid element is visualized as arriving at the surface, and absorbing heat or mass by unsteady state conduction into a semi-infinite medium. The surface of the element is assumed to reach instantaneously the surface condition of the transporting surface with regard to temperature or concentration. The resulting instantaneous transport coefficient for this type of transport model always has the form

$$N_{Nu}(a) = \frac{1}{\sqrt{\pi a}} \quad (15)$$

where  $a$  is the *dimensionless* age of the element. Equation (15) is generally obtained by inversion of its Laplace transform, which has the form

$$N_{Nu}(p) = \frac{1}{\sqrt{p}} \quad (16)$$

#### Gamma Distribution

For  $\alpha$  zero or a positive integer, use of Equation (11) on Equation (16) yields

$$\bar{N}_{Nu}(\bar{\tau}) = \frac{1}{\sqrt{(\alpha+1)\bar{\tau}}} \sum_{i=0}^{\alpha} \frac{\Gamma\left(i + \frac{1}{2}\right)}{i!} \quad (17)$$

where  $\Gamma$  is the gamma function. By using the identity

$$\frac{\Gamma\left(\alpha + \frac{1}{2}\right)}{\alpha!} = 2 \left[ \frac{\Gamma\left(\alpha + \frac{3}{2}\right)}{\alpha!} - \frac{\Gamma\left(\alpha + \frac{1}{2}\right)}{(\alpha-1)!} \right] \quad (18)$$

which applies for nonzero  $\alpha$ , the series in Equation (17) may be summed to yield the result

$$\bar{N}_{Nu}(\bar{\tau}) = \frac{2\Gamma\left(\alpha + \frac{3}{2}\right)}{\alpha! \sqrt{\pi \bar{\tau}} (\alpha+1)} \quad (19)$$

It is easily verified that as  $\alpha$  approaches zero,  $\bar{N}_{Nu}$  in Equation (19) approaches  $1/\sqrt{\bar{\tau}}$  and as  $\alpha$  approaches infinity,  $\bar{N}_{Nu}$  approaches  $2/\sqrt{\pi \bar{\tau}}$ . Equation (19) is plotted in Figure 5 in the form of  $\bar{N}_{Nu} \sqrt{\bar{\tau}}$  vs.  $1/(1+\alpha)$  for all values of  $\alpha$  between zero and infinity.

For the special case  $\alpha = -1/2$ , Equations (5) and (9) yield upon direct integration

$$\bar{N}_{Nu}(\bar{\tau}) = \frac{2\sqrt{2}}{\pi\sqrt{\bar{\tau}}} \frac{0.9}{\sqrt{\bar{\tau}}} \quad (20)$$

#### Dead Time Model

Using the age distribution of Equation (8) in Equation (9) and integrating, one obtains

$$\bar{N}_{Nu}(\bar{\tau}) = 2\sqrt{\frac{d}{\pi\bar{\tau}}} + \sqrt{\frac{1-d}{\bar{\tau}}} \exp\left(\frac{d}{1-d}\right) \operatorname{erfc}\left(\sqrt{\frac{d}{1-d}}\right) \quad (21)$$

Equation (21) for the average transport coefficient is plotted in the form  $\bar{N}_{Nu}\sqrt{\bar{\tau}}$  vs.  $d$  in Figure 5.

#### Variation of $\bar{N}_{Nu}$ with Model Parameter

Figure 5 shows clearly that the value of the average transport coefficient  $\bar{N}_{Nu}$  when the instantaneous coefficient is given by Equation (15) is insensitive to changes in the shape of the residence time and age distributions. The variation indicated is only 13% over all values of  $\alpha$  and  $d$  considered in Figure 5. In the special case  $\alpha = -\frac{1}{2}$ , an additional variation of 10% may be introduced, but even this is not considered to be a significant variation in the average transport coefficient. Note the distinct difference between this result and that of Perlmutter (5), who found a relatively great dependence of the transport coefficient upon the dead time. However, when Perlmutter varied the dead time, he also varied the average residence time directly. Figure 5 shows clearly that most of the variation of the average coefficient observed by Perlmutter was due to the variation in average residence time and not to changes in the dead time.

The lack of sensitivity of the average transport coefficient occurs despite the relatively great change in the shape of the age distribution  $\phi(\alpha; \bar{\tau})$  shown in Figures 2 and 4. This is caused by the basic form of the instantaneous rate coefficient given in Equation (15). To demonstrate this, we can construct a simple example where all the transport to a fluid element occurs suddenly at a specific age and relatively little transport occurs at other times. This situation might be generated by transport due to an exothermic chemical reaction at a hot reacting surface. As the fluid element arrives at the surface, little

reaction occurs until it heats up to the necessary temperature, at which point the reaction goes quickly and subsides when the supply of reactant in the fluid element is exhausted. In the limiting case the instantaneous rate coefficient would take the form of an impulse function centered at the age required for heating to the reaction temperature. Figures 2 and 4 show that use of an impulse function in Equation (9) will cause the average transport coefficient to depend significantly upon the shape of the age distribution.

The insensitivity exhibited by instantaneous rate coefficients of the form of Equation (15) to the shape of the age distribution is a great advantage for much work in transport phenomena. The implication is that a knowledge of the average residence time  $\bar{\tau}$  is sufficient to make acceptable predictions of the average transport coefficients without knowledge of the shape of the age distribution. Part II of this paper presents techniques for estimating the average residence time, and thus provides a method for taking advantage of the insensitivity to the shape of the age distribution.

One further point should be noted. The enormous differences in the shape of the residence time distributions depicted in Figure 1 lead to much smaller differences in the shape of the age distributions shown in Figure 3, primarily because of the integration process used in calculating the age distribution. In fact, since the age distribution has a fixed intercept and must be monotonously decreasing with unit area, it is obvious that a wide variety of choice of shapes is not available. While this may be a disadvantage for study of processes of surface renewal, it must be considered an advantage for calculation of average transport coefficients, as described above. Thus, the combination of insensitivity of age distribution to the residence time distribution, and insensitivity of average transport coefficient (in the typical case considered above) to shape of the age distribution, is a significant advantage of the surface renewal model for analysis of transport processes.

#### Application to Fluidized-Bed Heat Transfer

Thus far, only one model for transport at the surface has been considered in calculating an average transport coefficient, namely, that of Equation (15). Recent studies (1, 4, 7) of heat transfer in fluidized beds have produced variations of this basic model, as well as data (4) on average residence time and average transport coefficients for comparison of these models.

The basic mechanism for these models is the same and is illustrated in Figure 3 of reference 7. Particles from the fluidized bed arrive at the heating surface, absorb heat, and then leave to transport this heat to the bulk of the fluidized bed. Mickley et al. (4) assume that the particles arrive as packets of quiescent solids, and that transport occurs by unsteady conduction into these packets. The resulting expression for the instantaneous transport coefficient is that of Equation (15). The Nusselt number given in Equation (15) is considered to be based upon the difference between the temperature  $t_w$  of the heating surface and the temperature  $t_b$  of the bulk of the fluidized bed.

Baskakov (1) replaces one of the boundary conditions of Mickley et al. with a finite resistance to heat transfer at the surface, expressed by the constant  $M$ , the Nusselt number describing the contact conductance. The solution of the boundary value problem now results in the following Laplace transform of the instantaneous rate coefficient:

$$N_{Nu}(p) = \frac{M}{\sqrt{p}(M + \sqrt{p})} \quad (22)$$

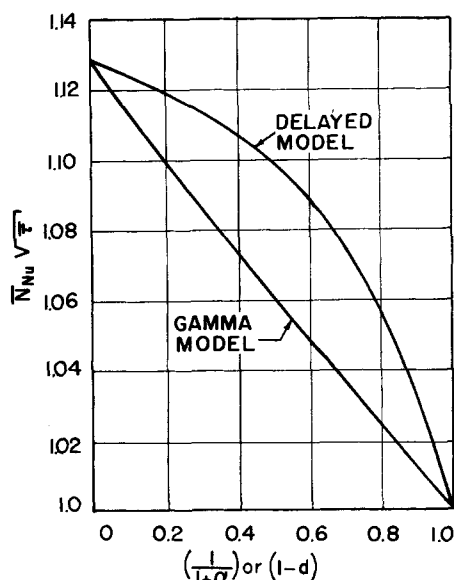


Fig. 5. Average Nusselt number from gamma and delayed exponential distributions.

This may be inverted to

$$N_{Nu}(a) = Me^{M^2a} \operatorname{erfc}(M\sqrt{a}) \quad (23)$$

Note that as  $M$  approaches infinity, Equations (22) and (23) approach the results for Mickley's model.

Ziegler et al. (7) have proposed a model involving unsteady state accumulation of energy in the surface layer of particles. To put their model on a comparable basis with the models described above, it is necessary to consider this mechanism to be in series with a mechanism whereby the surface particles also lose heat by conduction to the quiescent solids behind them. For this model, the resulting Laplace transform of the average Nusselt number is given by

$$N_{Nu}(p) = \frac{\frac{\pi\mu_o\lambda}{4\gamma}(p + \gamma\sqrt{p})}{p(p + \gamma\sqrt{p} + \lambda)} \quad (24)$$

where  $\mu_o$  is the density of packing of particles at the surface, expressed in particles per unit area, and  $\lambda$  and  $\gamma$  are constants involving bed and gas density and conductivity. Note that inversion of Equation (24) is not simple, but will not be required in order to calculate average coefficients, because Equation (11) will be used.

The three models described were compared on Mickley's data for heat transfer to M-11 microspheres, at a velocity of 1 ft./sec. over the minimum fluidizing velocity. At these conditions Mickley found that bubbles occupied the heater surface approximately 48% of the time; hence the coefficients computed from Equation (9) will be multiplied by 0.52 to account for this factor. Furthermore, Mickley reports two types of average residence times. The first is based upon the observed frequency of recurrence of a temperature pattern at the heating surface, described by an abrupt decrease in the heater temperature. The second type of average residence time results from the frequency of recurrence of crests or troughs in the temperature-time curves. It is assumed that the former frequency, designated  $\omega_d$  is indicative of replenishment of the entire surface with fresh particles from the fluidized bed, while the latter, designated as  $\omega_o$ , is associated with the frequency of stirring of the particles near but not at the heat transfer surface. Mickley prefers to use  $\omega_d$  in his calculation of average transport coefficient, while Baskakov prefers to use  $\omega_o$ . Since some compromise is required, we shall here calculate the average Nusselt number from both possible average residence times.

The observed value of Nusselt number is given by Mickley as 0.54. The values computed from the various models, with a gamma distribution for the residence time, with different values of the shape parameter  $\alpha$ , and with

both average residence times are reported in Table 1. It may be seen that none of the models is greatly dependent upon the shape parameter  $\alpha$ . Note that for  $\alpha = \infty$ , the results for the model of Ziegler et al. were not computed or approached because successive differentiations of Equation (24) become increasingly difficult. However, the results for  $\alpha$  zero and unity give no reason to suspect increased sensitivity of this model to  $\alpha$ . Also note that regardless of model, average residence time, or shape parameter  $\alpha$ , none of the proposed calculations is more than 35% inaccurate in its prediction of the average Nusselt number. In particular, knowledge of the average residence time alone is sufficient to estimate the  $\bar{N}_{Nu}$  predicted by each of the models, independently of  $\alpha$ , which describes only the shape of the distribution.

## ACKNOWLEDGMENT

Work performed under the auspices of the U. S. Atomic Energy Commission through Contract No. W-31-109-eng-38.

## NOTATION

$a$	= dimensionless age
$c_n$	= Laguerre coefficient
$d$	= dead time as fraction of average residence time
$F$	= cumulative distribution resulting from residence time density function
$f$	= residence time density function
$f_d$	= delayed residence time density function
$L_n$	= Laguerre function
$M$	= Nusselt number for contact resistance
$N_{Nu}$	= convective Nusselt number
$\bar{N}_{Nu}$	= Nusselt number averaged over age distribution
$p$	= Laplace transform variable
$s$	= $(\alpha + 1)/\bar{\tau}$
$t_b$	= bulk temperature of fluidized bed
$t_w$	= temperature of heating surface
$u(\tau)$	= unit step function = $\begin{cases} 0 & \tau < 0 \\ 1 & \tau > 0 \end{cases}$

## Greek Letters

$\alpha$	= shape parameter in gamma distribution
$\beta$	= constant of proportionality
$\Gamma$	= gamma function
$\gamma$	= constant
$\lambda$	= constant
$\mu_o$	= density of particle packing at surface, particles per unit area
$\tau$	= dimensionless residence time
$\bar{\tau}$	= average dimensionless residence time
$\phi$	= age density function
$\phi_d$	= delayed age density function
$\omega_d$	= frequency of total replenishment of heater surface
$\omega_o$	= frequency of partial replenishment of heater surface

## LITERATURE CITED

1. Baskakov, A. P., *Intern. Chem. Eng.*, **4**, 320 (1964).
2. Danckwerts, P. V., *Ind. Eng. Chem.*, **43**, 1460 (1951).
3. Higbie, R., *Trans. Am. Inst. Chem. Engrs.*, **31**, 65 (1935).
4. Mickley, H. S., D. F. Fairbanks, and R. D. Hawthorn, *Chem. Eng. Progr. Symposium Ser. No. 32*, **57**, 51 (1961).
5. Perlmutter, D. D., *Chem. Eng. Sci.*, **16**, 287 (1961).
6. Wasmund, B., and J. W. Smith, *Can. J. Chem. Eng.*, **43**, 246 (1965).
7. Ziegler, E. N., L. B. Koppel, and W. T. Brazelton, *Ind. Eng. Chem. Fundamentals*, **3**, 324 (1964).
8. Zwieterling, T. N., *Chem. Eng. Sci.*, **11**, 1 (1959).

Manuscript received November 5, 1965; revision received March 7, 1966; paper accepted March 8, 1966.

TABLE 1. AVERAGE NUSSULT NUMBERS FOR FLUIDIZED-BED HEAT TRANSFER WITH DIFFERENT MODELS, AVERAGE RESIDENCE TIMES, AND AGE DISTRIBUTIONS

(Experimental value = 0.54)

Distribution parameter, $\alpha$	Mickley, $\omega_d$	Mickley, $\omega_o$	Baskakov, $\omega_d$	Baskakov, $\omega_o$	Ziegler, $\omega_d$	Ziegler, $\omega_o$
0	0.47	0.61	0.38	0.46	0.47	0.59
1	0.50	0.65	0.40	0.49	0.50	0.63
$\infty$	0.53	0.69	0.43*	0.53*	—	—

\* Calculated by direct integration of Equation (23) with  $\bar{\tau}\phi(a;\bar{\tau}) = u(a) - u(a - \bar{\tau})$ .