Observations on Fixed-Bed Dispersion Models: The Role of the Interstitial Fluid

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Models of the role of the interstitial fluid for dispersion in a fixed bed for very high Reynolds numbers are examined in detail to determine how well they fit the physical requirements of finite speed of signal propagation, no backmixing, conservation and correct asymptotic form. It is concluded that no linear continuous partial differential equation of finite order can satisfy all of these requirements as well as the cell model, although all models for sufficiently large axial space and time variables are very good approximations to the cell model. Some new hyperbolic models are presented and their solutions obtained and compared with the standard solutions. In present day design it appears that no model has an outstanding advantage over any other.

SCOPE

Most design models for fixed-bed reactors, adsorbers and other exchangers are simple lumped constant models in which axial dispersion is neglected. For those with no radial transfer of heat or mass, the model is a two-dimensional one, one for the space variable and one for the time variable. Models such as this cannot be descriptive of the intersticial fluid, as tracer experiments have amply shown. In order to allow for the dispersion which these experiments show, the model equations have been modified by adding a space derivative of second order to make them of the diffusion type. However, experiments have shown that there is no gross backmixing (we neglect molecular diffusion) in the bed even at high Reynolds numbers. Thus, the addition of the second derivative term to allow for dispersion creates a difficulty, since such a model does predict backmixing. It produces another difficulty which is well known. Such equations being of the parabolic type have inherent in their solutions the prediction of an infinite speed of propagation (in attenuated form) for any signal. This is clearly not physically acceptable, although to the authors' knowledge no experiments have ever been performed. Thus, a model to be physically acceptable must satisfy four requirements: it must be a conservation system, it must not allow backmixing, it must predict a finite speed of signal propagation and it must produce the correct asymptotic (steady state form). It is known, of course, that the cell model with appropriate time delays will satisfy all four of these requirements. With this as a starting point, the problem addressed is whether it is possible to develop a second-order continuous model which will do the same. Such a model by necessity must be of the hyperbolic type, and so the problem is to determine how the coefficients of the derivatives in the describing equation should be defined and how the initial conditions and boundary condition at the bed entrance should be formulated. It is shown that there is no second-order model which can satisfy all four requirements, and probably no model of any finite order which will do so.

CONCLUSIONS AND SIGNIFICANCE

The well-known standard dispersion model for flow of interstitial fluid in a fixed bed predicts backmixing and infinite speed of signal propagation. Hiby's (1963) experiments clearly show that there is virtually no backmixing of fluid in fixed beds, and it is obvious that the signal speed will be equal to that of the fastest eddy. In this paper, the flow of interstitial fluid is described by a second-order hyperbolic, linear partial differential equation, in order to meet the requirement of finite signal speed. If this description has to satisfy the requirement of no backmixing, both second-order waves must move only downstream. However, this demands that two boundary conditions be specified at the inlet. The second boundary condition at the inlet is not at all obvious from the physics of the problem. It appears that the requirement of no backmixing is impossible to meet with models described by second-order linear partial differential equations. It is pointed out that unless appropriate boundary conditions are used at the inlet, the model for fluid flow will not be a conservation system. For the hyperbolic model considered here, the results corresponding to the exact inlet boundary condition are obtained and compared with cell model results. Some of the parameters appearing in the model are calculated by requiring that the continuous model reproduce the results of the cell model with least deviation. It is also found that the analysis of the solutions for concentration profiles in fixed beds affords no great discrimination between the various models of the interstitial fluid. It probably means that the standard dispersion model is satisfactory for present design purposes. However, one should recognize that the fine structure of the standard dispersion model is substantially deficient in details.

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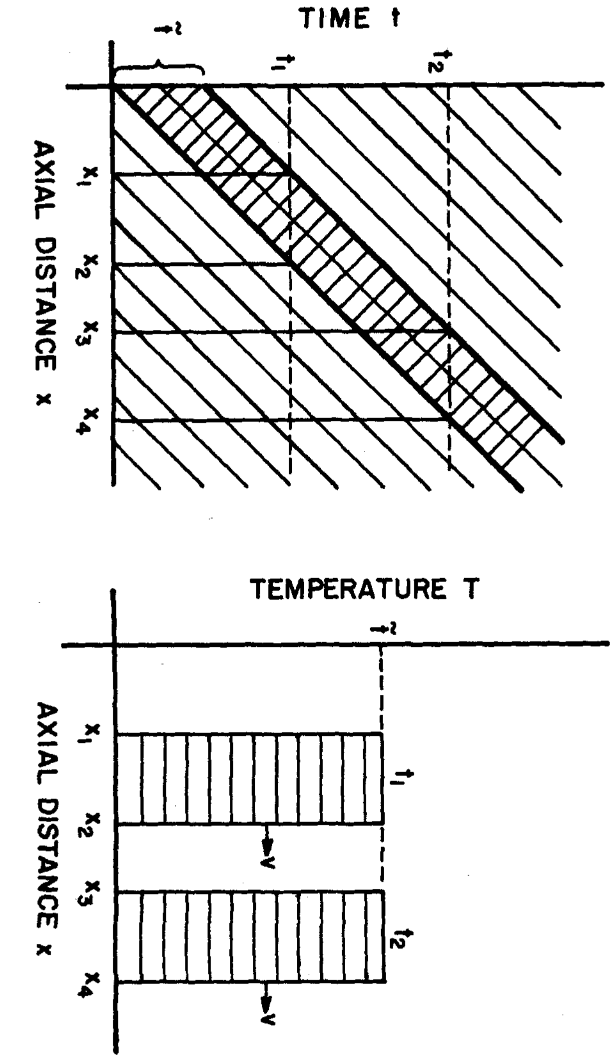

Figure 1. Wave character in the equilibrium heat transfer model for a packed bed.

Operations carried out in fixed beds, that is those in which a fluid, either gaseous or liquid, moves through a stationary matrix of particles, have a long history in chemical engineering. Adsorption, leaching, the pebble heater, the blast furnace, catalytic reactors and more are all prime examples. Unfortunately, analytical description of such processes is almost impossible from a rational point of view since the fluid flow through the interstices of the packing and the transport between this fluid and the solid present formidable difficulties. If the particles are small enough, and if they are assumed to be spheres, the hypothesis that they are in a homogeneous environment is reasonable if not accurate. For larger particles, the ambient conditions, front and back, are certainly not the same. Thus the rational analysis is almost beyond description or computation and, perhaps, even unnecessary. However, one should try to describe the process as well as possible, since the needs of tomorrow may be substantially different from the experience of yesterday. The purpose of this paper is not to examine the complete problem but rather to examine the models for the role of the interstitial fluid at high Reynolds numbers. For very low Reynolds numbers, there is some hope for the analytical description of the interstitial fluid, but from the practical engineering aspect this may not be an important regime. Thus, our interest here is in the asymptotic region of high Reynolds numbers, and we shall examine, in almost a chronological order, the current models and attempt to make explicit the consequences of some rather innocent assumptions.

THE SIMPLE MODEL

Consider the simplest model for heat transfer in a packed bed in which fluid is being heated or cooled, as in a pebble heater, and suppose the simplest assumption of constant parameters is invoked. Then it is well known that the problem can be described in the first instance by

$$\epsilon C_f \rho_f \left(u \frac{\partial T_f}{\partial x} + \frac{\partial T_f}{\partial t} \right) + U_h a_v (T_f - T_s) = 0$$

$$(1 - \epsilon) C_s \rho_s \frac{\partial T_s}{\partial t} = U_h a_v (T_f - T_s)$$

and with

$$T_f = T_{f0},\ x = 0$$
$$T_f = T_s = T_{fi},\ t = 0$$

this problem has a solution which is well known. This is an old and honorable problem whose solution has been rediscovered many times under various guises. Its notable feature for our purpose is that the temperature signal is propagated with the speed u of the interstitial fluid. The functions involved in the solution have been fairly extensively tabulated. We should also observe that the model assumes that there is an average interstitial velocity independent of position so that the fluid moves through the bed with this constant velocity. Obviously such a model does not predict backmixing, but it does produce solutions which have some of the character of dispersion solutions.

If we assume that the particles are very small and/or that the heat transfer coefficient U_h is very large, then the fluid and solid temperatures are essentially the same and as an approximation our model becomes, since now $T_f = T_s = T$

$$u \epsilon \rho_f C_f \frac{\partial T}{\partial x} + [\epsilon C_f \rho_f + (1 - \epsilon) C_s \rho_s] \frac{\partial T}{\partial t} = 0$$

This is a first-order partial differential equation which can be written as

$$\frac{dT}{ds} = 0\ ;\ \frac{dt}{dx} = \frac{\epsilon C_f \rho_f + (1 - \epsilon) \rho_s C_s}{u \epsilon C_f \rho_f} = \frac{1}{v}$$

where s is a parameter along the characteristic direction so that T is a constant along straight lines having slope of dt/dx. In Figure 1 we have shown the solution structure for initial temperature zero and with the inlet temperature T, a constant, for a fixed time interval and zero later. This implies that a square temperature wave T moves through the bed with speed v and length shown by the obvious construction in the figure. If the pulse in Figure 1 becomes an impulse, then the impulse moves through the bed with velocity v which may be substantially less than the fluid interstitial velocity. For heat transfer this is surprising, since such a result is always blurred by various heat transfer mechanism.

Let us consider the injection of a solution into a fixed bed of pure solvent packed with inert insoluble particles. The corresponding model for this case is

$$u \frac{\partial c}{\partial x} + \frac{\partial c}{\partial t} = 0$$

or

$$\frac{dc}{ds} = 0,\ \frac{dt}{dx} = \frac{1}{u}$$

and Figure 1 is valid with a pulse of solute, moving through the bed with velocity u. Now there is ample experimental evidence to show that this is not what happens. An impulse tends to broaden out and for some distance down the bed has all the appearance of a normal distribution curve, flattening out as time increases but with a peak near $x - ut$. Thus, there is a long tail behind the maximum and, more importantly, a part of the signal would move faster than the interstitial velocity. Thus, the original heat transfer model must be deficient in spite of its ubiquity.

THE STANDARD DISPERSION MODEL

It is clear that the normal distribution curve can be generated as the solution of a model for the interstitial fluid of the form

$$D\frac{\partial^2 c}{\partial x^2} - u\frac{\partial c}{\partial x} - \frac{\partial c}{\partial t} = 0 \quad (1)$$

Casting this into the usual dimensionless form

$$\frac{\partial^2 c}{\partial y^2} - Pe\frac{\partial c}{\partial y} - \frac{\partial c}{\partial \theta} = 0$$

to make the Peclet number apparent, where

$$Pe = \frac{ud}{D},\ y = \frac{x}{d},\ \frac{Dt}{d} = \theta$$

d being the particle diameter, and using the generally accepted boundary condition at $x = 0$ with a delta function input

$$D\frac{\partial c}{\partial x} = u[c - \delta(t)],\ x = 0 \quad (2)$$

one can easily obtain the solution for the semi-infinite interval $0 < y < \infty$. The solution can be written in terms of the original variables as

$$c(x,t) = \frac{u}{\sqrt{\pi D t}}\exp\left[-\frac{(x-ut)^2}{4Dt}\right] - \frac{u^2}{2D}\exp\left(\frac{ux}{D}\right)\operatorname{erfc}\left(\frac{x+ut}{2\sqrt{Dt}}\right)$$

Using the asymptotic form for the erfc one obtains for large $x + ut$ in the neighborhood of $x = ut$.

$$c(x,t) = \frac{u}{\sqrt{4\pi D t}}\exp\left[-\frac{(x-ut)^2}{4Dt}\right]$$

a normal distribution of concentration as a function of x with mean ut and variance $2Dt$. For future reference, we note that the model indeed is conservation system for on integration over the whole tube

$$\int_0^\infty\left(D\frac{\partial^2 c}{\partial x^2} - u\frac{\partial c}{\partial x} - \frac{\partial c}{\partial t}\right)dx = 0$$

or

$$\left(-D\frac{\partial c}{\partial x} + uc\right)_{x=0} - \frac{d}{dt}\int_0^\infty c(x,t)dx = 0$$

assuming $c = \partial c/\partial x = 0$ as $x \to \infty$. This may be written using the boundary conditions given above as

$$\frac{dc_\Sigma}{dt} = \left(-D\frac{\partial c}{\partial x} + uc\right)\Big|_{x=0} = u\delta(t)$$

or

$$c_\Sigma(t) = \int_0^\infty c(x,t)dx = u$$

which, since $u\delta(t)$ was the amount introduced, implies that all of the molecules introduced remain in the bed, and hence the model with the appropriate boundary condition is a conservation system. (Observe that with any other boundary condition, the model is not conservative.) In order to show that the model had validity. McHenry and Wilhelm (1957) showed that the axial Peclet number Pe was a constant and equal to a little more than 2 for Reynolds numbers from 10 to 400. This model is generally referred to as the dispersion model and has been widely used in a variety of fixed-bed operations. It should be noted that the model is of Fickian form and could be derived by assuming that the flux of molecules in the packed bed is given by

$$q = uc - D\frac{\partial c}{\partial x}$$

On continued reflection, however, it is difficult to sustain the idea that mixing in a packed bed is determined by concentration gradients, since we are not concerned with molecular diffusion, turbulent diffusion or Taylor dispersion, but rather with mechanical mixing caused by fluid flow.

Although it has generally been ignored except by Professor E. Wicke, who has brought it to the attention of the authors repeatedly, the experiments of Hiby (1963) show that such a model has at least one serious deficiency. In Hiby's experiment, N/3000 sulfuric acid containing phenolphthalien flows through a randomly packed bed of 9 mm spheres. A continuous point source of one normal alkali also containing the indicator was injected. The violet color produced should be visible when only 1/3000$^{\text{th}}$ part of the alkali has been introduced. The solid line in Figure 2 shows where the violet color should be visible, calculated from a dispersion model including the radial term

$$D_a\frac{\partial^2 c}{\partial x^2} + D_r\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial c}{\partial r}\right) - u\frac{\partial c}{\partial x} = 0$$

with Re = 300 and the appropriate values of the radial and axial Peclet numbers. Thus, the dispersion model predicts backmixing, which is obvious from the form of the solution, but which is not confirmed experimentally. We should note here, since we are criticizing the model, that it also predicts a speed of signal propagation which is infinite both fore and aft, and for large $x + ut$ the solution is symmetric with respect to $x = ut$.

THE CELL MODEL

While the cell model was not introduced to circumvent the deficiencies of the dispersion model, it does seem to be some-

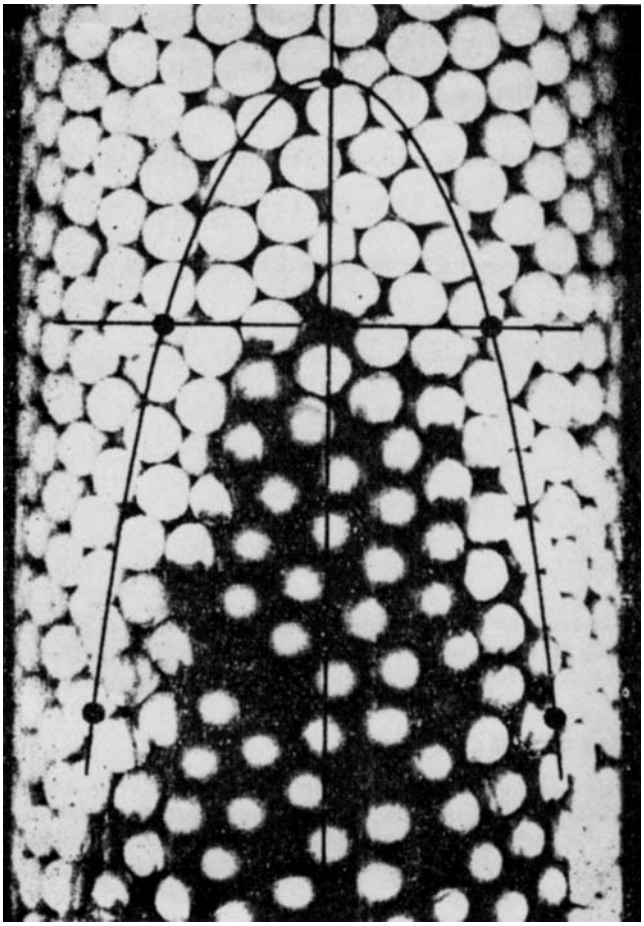

Figure 2. The Hiby experiment. Tracer injected continuously at a point in a packed bed.

what more rational in this regard. The cell model was originally introduced when it appeared that the solution of large numbers of algebraic equations for the steady state or ordinary differential equations for the unsteady state was simpler than the solution of the partial differential equations required of dispersion models. In the cell model it is assumed that the interstitial velocity in the bed varies as the fluid moves axially, since, as has been pointed out, the fractional free area normal to the flow can vary substantially. We can therefore assume that regions of low velocity are cells connected by tubes in which fluid moves at a high velocity. We can thus write the cell model in standard one-dimensional form as

$$qc_{n-1} - qc_n = v\frac{dc_n}{dt},\ n \geq 2$$

$$q\delta(t) - qc_1 = v\frac{dc_1}{dt}$$

$$c_n(0) = 0,\ n \geq 1$$

The solution of this system is

$$c_n(t) = \left(\frac{q}{v}\right)^n \frac{t^{n-1}}{(n-1)!}\exp\left(-\frac{qt}{v}\right),\ n \geq 1$$

$$= \frac{1}{\theta^n}\frac{t^{n-1}}{(n-1)!}\exp\left(-\frac{t}{\theta}\right),\ n \geq 1$$

a Poisson distribution function. Such a solution, as is well known, approaches a normal distribution, and direct calculation shows that with the appropriate values for the parameters, the cell model solution and the standard dispersion model solution are almost superimposable. Comparisons of the moments of the distributions, that is, the zeroth, first and second for a rhombohedral blocked passage bed, gives a Pecklet number of 2.46 and slightly more than 2 for a randomly packed bed. Thus, the cell model and the dispersion model describe the observed phenomenon of longitudinal dispersion adequately, but the cell model does not produce backmixing. However, it does, as written above, predict an infinite speed of signal propagation, since for any $t > 0$, there is a nonzero concentration in the n^{th} cell for any n.

As a diversion, we will write the cell model in the form

$$c(n-1,t) - c(n,t) = \theta\frac{dc}{dt}(n,t)$$

which, on expanding the first term in a Taylor series, gives, since $\Delta n = 1$

$$-\frac{\partial c}{\partial n} + \frac{1}{2}\frac{\partial^2 c}{\partial n^2} = \theta\frac{\partial c}{\partial t}(n,t)$$

Now

$$dx = \gamma dn,\ q = A\epsilon u,\ A\epsilon\gamma d = v,\ \theta = \frac{v}{q}$$

and therefore

$$-u\frac{\partial c}{\partial x} + \frac{\gamma du}{2}\frac{\partial^2 c}{\partial x^2} = \frac{\partial c}{\partial t}$$

and hence the Peclet number is indeed $2/\gamma$, since $D = u\gamma d/2$. While this may not seem surprising, we will show that the obvious mathematical extension is invalid.

MORE ON THE CELL MODEL

The cell model in the form in which it is presented above hides an essential ingredient; that is, the fluid does not pass from one cell to the next instantaneously. The cells are connected by passages of varying lengths, depending upon how the bed is packed, and the time of transit Δt from one cell to the next is therefore finite and nonzero (the definition of Δt as an operational parameter is not obvious). Thus we should probably write the cell model as

$$qc(n-1,\ t-\Delta t) - q\,c(n,t) = v_s\frac{\partial c}{\partial t}c(n,t)$$

Before we proceed it is convenient to change the variables. Let

$$v_s + v_p = v$$

$$v/q = \theta$$

If τ is defined as the fraction of the total residence time which the fluid spends in the plug flow section of a cell, then $\theta\tau$ is the residence time in the plug flow section and $\theta(1-\tau)$ that for the remainder, or

$$\theta\tau = v_p/q;\ \theta(1-\tau) = v_s/q$$

Thus, the cell model is

$$c(n-1,z-\tau) - c(n,z) = (1-\tau)\frac{\partial c}{\partial z}(n,z) \quad (3)$$

where $z = t/\theta$ is the dimensionless time. The solution of Equation (3) with the conditions

$$c(n,0) = 0$$

$$c(0,z) = \delta(z) \quad (4)$$

is given by

$$c(n,z) = \begin{cases} 0 & n > \frac{z}{\tau} \\ \frac{1}{(1-\tau)^n}\frac{(z-n\tau)^{n-1}}{(n-1)!}\exp\left(-\frac{(z-n\tau)}{(1-\tau)}\right), & n \leq \frac{z}{\tau} \end{cases}$$

As one expects, there is a finite speed of signal propagation, which depends upon the parameter τ. Thus, this solution has all the desirable properties of a proper model for the interstitial flow. Note that this model predicts that the signal enters the stirred-tank section of the first cell only at $z = \tau$; that is, plug flow section precedes the well-stirred region in every cell. If, however, one replaces (4) by

$$\delta(z) - c(1,z) = (1-\tau)\frac{dc}{dz}(1,z)$$

the solution corresponds to a situation in which the well-stirred region precedes the plug flow section in every cell.

CONTINUOUS MODEL

If one expands the first term in Equation (3) in a Taylor series, one obtains, after dropping terms of order higher than the second (with $\Delta n = 1$)

$$-\frac{\partial c}{\partial n} - \tau\frac{\partial c}{\partial z} + \frac{1}{2}\left[\frac{\partial^2 c}{\partial n^2} + 2\frac{\partial^2 c}{\partial n\partial z}\tau + \tau^2\frac{\partial^2 c}{\partial z^2}\right] = (1-\tau)\frac{\partial c}{\partial z}$$

or

$$-\frac{\partial c}{\partial y} - \frac{\partial c}{\partial z} + \frac{1}{2}\left[\frac{\partial^2 c}{\partial y^2} + 2\tau\frac{\partial^2 c}{\partial y\partial z} + \tau^2\frac{\partial^2 c}{\partial z^2}\right] = 0$$

where the continuous variable y will be used in place of n. In order to obtain a solution to this equation for the semi-infinite domain $0 < y < \infty$, $z < 0$, one needs to specify, in addition to some boundary conditions at $y = 0$, the values of c and $\partial c/\partial z$ at $z = 0$. This equation is a second-order partial differntial equation of parabolic type, and it is well known that such a system is not well posed mathematically, since the solution will not be a continuous function of the initial conditions. Note that the equation is of the form

$$\alpha\frac{\partial c}{\partial y} + \beta\frac{\partial c}{\partial z} + a\frac{\partial^2 c}{\partial y^2} + 2b\frac{\partial^2 c}{\partial y\partial z} + \tilde{a}\frac{\partial^2 c}{\partial z^2} = 0$$

If $b^2 - a\tilde{a} = 0$, the system is parabolic, and if $b^2 - a\tilde{a} > 0$, it is hyperbolic. It is clear that other expansions than that above are possible, for the cell model can be written in other ways; for example

$$c(n-1,z) - c(n,z+\tau) = (1-\tau)\frac{\partial c}{\partial z}(n,z+\tau)$$

$$c(n,z-\tau) - c(n+1,z) = (1-\tau)\frac{\partial c}{\partial z}(n+1,z)$$

$$c(n,z) - c(n+1,z+\tau) = (1-\tau)\frac{\partial c}{\partial z}(n+1,z+\tau)$$

All of these certainly give the same solution, but their approximations by expanding the appropriate terms in Taylor series, keeping only terms up to and including order two, will be different. In fact, one can generalize all of these four into a single cell model

$$c(n-\lambda, z-\mu\tau) - c[n+1-\lambda, z+(1-\mu)\tau]$$
$$= (1-\tau)\frac{\partial c}{\partial z}[n+1-\lambda, z+(1-\mu)\tau]$$

where we assume that $0 \le \lambda \le 1$, $0 \le \mu \le 1$. It is apparent that for appropriate choices of λ and μ, the four cell models above will be generated. What choices of these parameters would produce the best representation by a continuous model is an open question.

If one expands all of the terms into their Taylor series representations, dropping terms of order higher than the second, one obtains

$$\frac{\partial c}{\partial z} + \frac{\partial c}{\partial y} + \eta\left[\frac{\partial}{\partial z} + a_1\frac{\partial}{\partial y}\right]\left[\frac{\partial c}{\partial z} + a_2\frac{\partial c}{\partial y}\right] = 0 \quad (5)$$

where

$$\eta = \tau[1-\mu-\tau/2] \quad (6a)$$
$$\eta(a_1+a_2) = 1-\lambda-\mu\tau \quad (6b)$$
$$\eta a_1 a_2 = (1-2\lambda)/2 \quad (6c)$$

We note that this equation, following Whitham (1974), predicts the existence of three interacting waves, one with unit velocity, and the other two with velocities a_1 and a_2. Depending upon the signs of a_1 and a_2, the corresponding waves will move upstream or downstream. There are some restrictions on the problem necessitated by the physics and the knowledge obtained from simple dispersion experiments and also some from mathematical requirements.

With a fixed-bed model, the only boundary condition which makes sense at $y = 0$ is the specification of the flux or, as an approximation, the specification of the concentration. This would mean for Equation (5) that $\eta a_1 a_2 < 0$, since otherwide two conditions would have to be specified at $y = 0$, owing to the fact that if $\eta a_1 a_2 > 0$, all waves would point downstream. Also, as Whitham points out, for physical stability of the system, it is necessary that $\eta > 0$ and $a_1 > 1 > a_2$. Thus, one wave points downstream ($a_1 > 0$) and the other second-order wave points upstream ($a_2 < 0$). These conditions imply that

$$0 \le \mu \le 1 - \tau/2$$

$$\frac{1}{2} \le \lambda \le 1 \quad (6d)$$

One can show that the dimensionless speed of propagation downstream in the cell model is $1/\tau$, and it follows that

$$a_1 = \frac{1}{\tau};\ 0 < \tau < 1 \quad (6e)$$

Now whether or not these observations are correct is not clear. We are accustomed to a certain form for physical equations, and when these come into question, we are not prepared for the consequences. For example, it is not at all obvious that a fixed-bed model for the interstitial fluid should be of the form

$$\frac{\partial c}{\partial y} + \frac{\partial c}{\partial t} + \eta a_1 a_2 \frac{\partial^2 c}{\partial y^2} = 0 \quad (7)$$

with $\eta a_1 a_2 < 0$. With the appropriate boundary conditions it gives a well-posed mathematical problem, is a conservation system and presumably fits the experimental facts reasonably well, although a rigorous test has never been concluded. It certainly does not fit the no backmixing or finite signal speed criteria even approximately. Thus one is left with the uneasy feeling that there is need for a rational explanation of the problem. However that may be, we will proceed and attempt to devise a second-order model which will give the best fit to the cell model.

SOLUTIONS TO CONTINUOUS MODELS

Unfortunately, the formal solutions developed for the hyperbolic Equation (5) are not simple in structure. The details of finding the solutions will not be presented except to say that some rather tedious and ingenious manipulations for finding the inverse Laplace transforms are required. We will present solutions for four different boundary conditions with the initial condition of the interstitial fluid described by

$$c(y,0) = 0$$
$$\frac{\partial c}{\partial z}(y,0) = 0$$

The differential equation to be solved in Equation (5), where the parameters η, a_1, and a_2 are dimensionless.

Case I at $y = 0$, $c(0,z) = \delta(z)$

The solution can be shown to be

$$c(y,z) = \begin{cases} \exp\left\{\dfrac{(2-a_1-a_2)y}{\eta(a_1-a_2)^2} - \alpha_2 z\right\} G_1\left(z - \dfrac{y}{a_1}\right); & z > y/a \\ 0 & ; z < \dfrac{y}{a_1} \end{cases}$$

where

$$G_1(\xi) = \frac{\alpha_1\alpha_3 y}{2}[\xi^2 + \alpha_3 y\xi]^{-1/2} I_1[\alpha_1(\xi^2+\alpha_3 y\xi)^{1/2}] + \delta(\xi)$$

$$\alpha_1 = \frac{2[a_1a_2(a_1-1)(a_2-1)]^{1/2}}{\eta(a_1-a_2)^2};\quad \alpha_2 = \frac{a_1+a_2-2a_1a_2}{\eta(a_1-a_2)^2}$$
$$\alpha_3 = \frac{a_2-a_1}{a_1a_2}$$

Case II: at $y = 0$, $\delta(z) = c(0,z) + \eta a_1 a_2\, \partial c(0,z)/\partial y$

The solution can be shown to be

$$c(y,z) = e^{-\alpha_2 z}[D_2(y,z) - D_1(y,z)]$$

where $D_1(y,z)$ 0 for $z \le y/a_1$, while for $z > y/a_1$

$$D_1(y,z) = \exp\left\{f_1 y + p_1\frac{p_2(a_1+a_2)}{2\eta a_1a_2}\left(z-\frac{y}{a_1}\right)\right\}$$
$$\left[\frac{\alpha_3^2 p_2}{4\eta^2 f_3}\sinh\left\{f_3\left(z-\frac{y}{a_1}\right)\right\} + p_1\frac{a_1+a_2}{2\eta a_1a_2}\cosh\left\{f_3\left(z-\frac{y}{a_1}\right)\right\}\right.$$

$$+\frac{\alpha_1\alpha_3 y}{2\eta^2 a_1a_2}e^{f_1 y}\int_{\frac{y}{a_1}}^{z}\left[\left(x-\frac{y}{a_1}\right)^2 + \alpha_3 y\left(x-\frac{y}{a_1}\right)\right]^{-1/2}$$

$$I_1\left[\alpha_1\left\{\left(x-\frac{y}{a_1}\right)^2 + \alpha_3 y\left(x-\frac{y}{a_1}\right)\right\}^{1/2}\right]$$
$$\exp\left[\frac{p_2(a_1+a_2)p_1}{2\eta a_1a_2}(z-x)\right]$$

$$\left[\frac{p_2\alpha_3^2 a_1 a_2}{4f_3}\sinh[f_3(z-x)] + p_1\frac{\eta(a_1+a_2)}{2}\cosh[f_3(z-x)]\right]dx$$

$$D_2(y,z) = 0 \quad \text{for} \quad z \le y/a_1, \text{ while} \quad \text{for} \quad z > y/a_1$$

$$D_2(y,z)\frac{\alpha_3}{2\eta}\exp\{f_1 y\}$$

$$I_0\left[\alpha_1\left\{\left(z-\frac{y}{a_1}\right)^2 + \alpha_3 y\left(z-\frac{y}{a_1}\right)\right\}^{1/2}\right]$$

$$+\frac{\alpha_3}{2\eta}\int_{\frac{y}{a_1}}^{z}\exp\left\{f_1 y + p_1 p_2\frac{(a_1+a_2)}{2\eta a_1 a_2}(z-x)\right\}$$

$$I_0\left[\alpha_1\left\{\left(x-\frac{y}{a_1}\right)^2 + \alpha_3 y\left(x-\frac{y}{a_1}\right)\right\}^{1/2}\right]$$

$$\left[\left\{\frac{a_1^2+a_2^2}{2\eta^2 a_1^2 a_2^2}p_2^2 - \frac{(a_1+a_2)^2}{4a_1 a_2}\alpha_1^2\right\}\frac{\sinh\{f_3(z-x)\}}{f_3} + \frac{p_1 p_2(a_1+a_2)}{\eta a_1 a_2}\cosh\{f_3(z-x\}\right]dx$$

where

$$f_1 = \frac{2-a_1-a_2}{\eta(a_1-a_2)^2}; \quad f_2 = 1 + \frac{a_1 a_2(2-a_1-a_2)}{(a_1-a_2)^2}$$

$$f_3 = \frac{\alpha_3}{2\eta}[f_2^2 - \eta^2 a_1 a_2 \alpha_1^2]^{1/2}$$

$$p_1 = +1 \quad \text{and} \quad p_2 = f_2$$

Case III: at $y = 0$, $\delta(z) = c(0,z)$

$$+\ \eta(a_1 a_2)\frac{\partial c}{\partial z}(0,z) + \eta(\alpha_1 + a_2)\frac{\partial c}{\partial z}(0,z)$$

The solution may be shown to be the same as in case II with the following redefinitions:

$$p_1 = -1 \quad \text{and} \quad p_2 = f_2 - \eta\alpha_2(a_1 + a_2)$$

Case IV: at $y = 0$

$$\delta(z) + \eta\delta'(z) = c(0,z)$$

$$+\ \eta a_1 a_2\frac{\partial c}{\partial y}(0,z) + \eta(a_1 + a_2)\frac{\partial c}{\partial z}(0,z) \tag{8}$$

In this case, it may be shown that if c_3 is the solution of case III, then the solution now is

$$c(y,z) = c_3(y,z) + \eta\frac{\partial c}{\partial z}c_3(y,z)$$

DISCUSSION OF SOLUTIONS

Before we proceed, it is important to discuss the motivation to analyze the solutions corresponding to the four different boundary conditions mentioned above. Equation (5) can be equivalently represented as

$$\frac{\partial c}{\partial z} + \frac{\partial q}{\partial y} = 0$$

where q, the flux, is given by the constitutive equation

$$\eta\frac{\partial q}{\partial z} + q = c + \eta a_1 a_2\frac{\partial c}{\partial y} + \eta(a_1 + a_2)\frac{\partial c}{\partial z} \tag{9}$$

The appropriate condition at $y = 0$ corresponding to an impulse input would be $q(0^-,z) = \delta(z)$. Then by continuity of flux, $q(0^+,z) = \delta(z)$. Introducing this into Equation (9) one obtains the B.C. in case IV. Thus, the B.C. of case IV is the appropriate one to use in order to make the model a conservation system.

As we mentioned earlier, the widely used standard dispersion model [Equation (1)] demands that the appropriate B.C. for an impulse input be Equation (2), in order that the model should be a conservation system. However, quite often, when one is interested in the solution for large values of time, the flux B.C. [Equation (2)] is replaced by the concentration B.C. (where the concentration at the inlet is specified as an impulse). Clearly this violates the conservation condition. However, the error made can be shown to approach zero rather quickly, indicating good agreement between the solutions of the standard dispersion model with the flux B.C. and the concentration B.C. for large times, thus justifying the use of the concentration B.C. and its somewhat easier solution. Fan and Ahn (1962) have studied this for the simple dispersion model, and two recent papers (Kreft and Zuber, 1978, and Choi and Perlmutter, 1976) bear witness to the perennial charms of these boundary conditions. By the same token, one would like to analyze the relative importance of various terms in Equation (8). Among the various combinations possible, we felt that the four cases above were the interesting ones.

The parameter τ is not arbitrarily adjustable. It would be determined by the size and shape of the particles in the fixed bed and how these particles are packed. Hence we will leave it as a free parameter and demand that for any value of τ, $0 < \tau < 1$, the continuous model must fit the cell model. Now, Equation (6) provides four equality constraints on the values which the five variables η, a_1, a_2, λ and μ may assume; that is, there is only one freely adjustable parameter. From Equation (6), it can be shown that

$$\mu = \lambda$$

$$\frac{1}{2} < \mu < 1 - \frac{\tau}{2}$$

$$0 < \tau < 1$$

In particular, when $\mu = 1/(1+\tau)$, one obtains $a_2 = -a_1 = -1/\tau$. Though this model cannot satisfy the no backmixing condition, we may impose a condition that $a_1 + a_2 \ge 0$; that is

$$\frac{1}{2} < \mu \le \frac{1}{1+\tau}$$

We can now proceed to determine the optimal choice of μ by comparing the results of the continuous model with the cell model. Since, among the four cases above, only case IV satisfies the conservation condition exactly, we will use this case to determine the optimal choice of μ.

The average concentration of the tracer in the n[th] cell predicted by the cell model would be

$$\bar{c}_1(n,z) = \int_{z-\tau}^{z} c(n-1,z')dz' + (1-\tau)\,c(n,z)$$

where c is given by the solution of Equations (3) and (4). The average concentration of tracer in the region $n-1 < y < n$

TABLE 1. ($\tau = 0.05$, $p = 2$)

μ	$E(0.5;\ \mu)$	$E(20;\ \mu^2$
0.51	0.2259 D-02	—
0.53	0.2366 D-02	—
0.55	0.2458 D-02	—
0.57	0.2644 D-02	—
0.90	0.1228 D-01	0.5839 D-02
0.92	0.1290 D-01	0.4421 D-02
0.94	0.1348 D-01	0.6045 D-03
$1/(1+\tau) = 0.9524$	0.1381 D-01	0.1217 D-03
0.96	0.1400 D-01	0.8788 D-03

predicted by the continuous model would be

$$\bar{c}_2(n,z) = \int_{n-1}^{n} c(y,z)dy$$

where c is given by the solution in case IV. This suggests that we can form an expression for the error of the form

$$E(z;\mu) = \sum_{n=1}^{z/\tau} |\bar{c}_1(n,z) - \bar{c}_2(n,z)|^p$$

where $p > 0$ is an arbitrary exponent. We tried $p = 1$ and $p = 2$, and both cases resulted in the same conclusion. The results presented are for $p = 2$. It is not at all obvious at what value of z one must attempt to minimize the error. We performed the minimization at different values of z. Typical computations (E vs. μ) for a small value of z and for a large value of z are presented in Table 1. After extensive comparisons for different values of z and τ, we conclude the following.

1. For small values of the time, the best fit is obtained when $\mu = 0.5^+$; that is, $a_2 = 0^-$.

2. As the time increases, the optimum shifts, and for large values of time the best fit is obtained when $\mu = 1/(1 + \tau)$; that is, $a_2 = -a_1 = -1/\tau$.

Since $z = 20$ (see Table 1) is approximately equal to the time required for the bulk fluid to travel a distance of twenty particle diameters, it is clear that μ reaches its asymptotic value rapidly. In other words, one may state that initially optimal μ is a function of time, and this optimal value rapidly approaches an asymptotic value of $1/(1 + \tau)$. This asymptotic value accords with the observations of McHenry and Wilhelm that were reported earlier for the apparent Peclet number in Equation (7) is then

$$\frac{-1}{\eta a_1 a_2} = 2\frac{1+\tau}{1-\tau}$$

which (for small τ) is slightly greater than 2.

When $\mu = 1/(1 + \tau)$, it can be easily shown that $a_1 + a_2 = 0$, and, therefore, cases II and III yield the same solution. Figure 3 is a typical comparison of the various cases of continuous models and the cell model, when z is large enough for μ to reach its asymptotic value. As mentioned earlier, case I (the simplest B.C.) yields a very compact solution compared to the unwieldy solution in case IV (exact B.C.). It can be seen from Figure 3 that the maximum difference between the results of the cell model and case IV of the continuous model is less than approximately 5% of the peak value. When $z = 40$, this difference becomes almost negligible. It can be seen from Figure 3 that the maximum difference between the results of cases I and IV of the continuous model is approximately 5% of the peak value. When $z = 40$, this difference is about 2%. It can be seen from Figure 3 that when $z = 20$, there is almost no difference between the results of cases II, III and IV. It can be seen from Figure 3 that the maximum difference between the results of the cell model and case I of the continuous model is about 10% of the peak value when $z = 20$. This difference decreases to about 2% when $z = 40$.

Hence, one can conclude that for sufficiently large values of time, the approximate B.C. in case I may be used instead of the exact one (case IV) for computational ease. It must be kept in mind that for small values of z, the error made by using the approximate forms of the boundary condition is significant. However, in the context of fixed beds, such small values of z are probably not of critical importance.

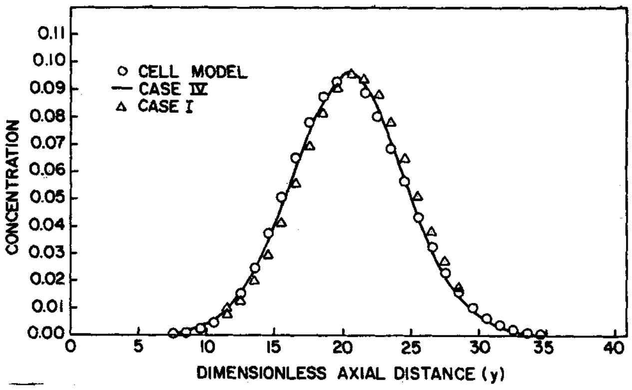

Figure 3. Comparison of the continuous models and the cell model. $z = 20$, $\tau = 0.05$. Results corresponding to cases II and III virtually overlap with that of case IV.

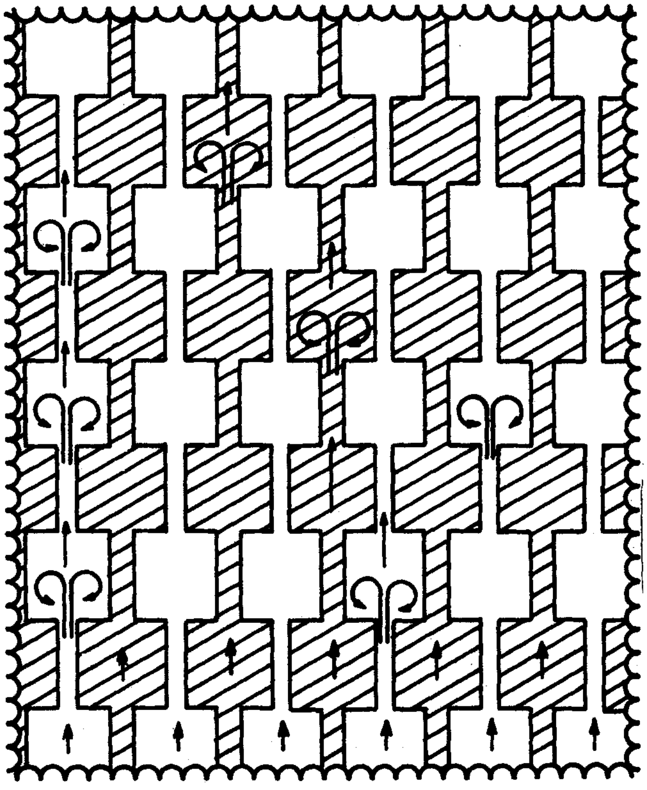

Figure 4. The idealized packed bed with packing on a set of measure zero.

CONCLUSIONS

The question is what does all this mean. It probably means that the model used for a fixed bed with dispersion is satisfactory for present design purposes, since none of the models shows any great difference from any other model. But one should recognize that the fine structure of the solutions is substantially different, and, if indeed one had to know the concentration of a particular species accurately at some pont, one should use the model which is physically the most acceptable. For example, given the structure shown in Figure 4 or its three-dimensional analogue, it would be desirable to describe it for high Reynolds numbers with some accuracy to be sure we know how to do it approximately. One of the essential conclusions of this paper is that there is no continuous model of the second order which can satisfy all of the requirements. There is probably no linear continuous model of any finite order containing only partial derivations which will satisfy the no backmixing and finite speed of propagation requirements.

ACKNOWLEDGMENT

A Laplace transform solution of Equation (5) corresponding to one of those given here was obtained by M. Cardon in a preliminary study of "Models for Axial Dispersion in Packed Bed Reactors" (M.S. thesis, University of Minnesota, 1978).

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NOTATION

a_1, a_2 = dimensionless velocities of the dispersion waves
a_v = area available for heat transfer between gas and solid phases in a packed bed
c = concentration (normalized with respect to unit concentration)
C_f, c_s = heat capacities of fluid and solid, respectively
d = particle diameter
D = dispersion coefficient
D_a, D_r = axial and radial dispersion coefficients, respectively
P_e = Peclet number = ud/D
t = time variable
T = temperature
u = interstitial velocity
U_h = heat transfer coefficient between gas and solid
v = volume of a cell (in cell model)
x = distance coordinate
y = dimensionless distance coordinate
z = dimensionless time coordinate

Greek Letters

ϵ = voidage of the packed bed
η, λ, μ = see Equations (5) and (6)
γ = length of a cell
ρ_f, ρ_s = densities of fluid and solid, respectively
θ = residence time in a cell
τ = fraction of the total residence time which the fluid spends in the plug flow section of a cell

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Conjugate Unsteady Heat Transfer From a Droplet in Creeping Flow

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The phenomenon of unsteady heat transfer from a spherical droplet or solid particle moving slowly in a different fluid is analyzed, for the case in which the thermal resistances of the dispersed and continuous phases are comparable. The effect of the Peclet number (over the range $0 \leq Pe \leq 1{,}000$) and of the internal circulation of the droplet on the development of temperature fields is investigated. The energy equations for the interior and exterior of the droplet are solved by the finite-difference method of alternating directions. The results are compared with a number of previously published approximate models.

SCOPE

The problem of unsteady heat transfer from droplets moving in another immiscible fluid is important for a number of engineering applications. The majority of theoretical studies on this problem have been carried out on the assumption that most of the thermal resistance is concentrated either in the dispersed phase or in the continuous medium. But in many cases (for example, in direct-contact heat exchangers), the physical properties of the two phases are similar, and their thermal resistances are comparable in magnitude. The available solutions pertain to limiting cases of either very low ($Pe = 0$) or very high ($Pe \rightarrow \infty$) Peclet numbers. For $Pe = 0$, an analytic solution was obtained by Cooper (1977). For high Peclet numbers, two competing approximate models have been published. The first of these assumes that a thin thermal boundary layer exists at both sides of the droplet surface (Levich et al. 1965, Chao 1969). In the second model (Elzinga and Banchero 1959, Brounshtein et al. 1970) the ideas of Kronig and Brink (1950) on a highly developed circulation within the droplet are applied, but the boundary conditions at the droplet surface make allowance for the heat transfer resistance in the continuous phase.

There is no satisfactory solution for the case of comparable phase resistances at intermediate values of Pe. Nevertheless, this region is important for the analysis of heat transfer from small droplets moving at $Re \lesssim 1$, since the Prandtl numbers for liquids are of the order of 10-1,000, and hence, $Pe = Re \cdot Pr \lesssim 1{,}000$.

The purpose of the present article is the study of the physics of interphase transport on the basis of a correct numerical solution of complete equations for energy transport. In particular, the effect of the Peclet number (over the range $0 \leq Pe \leq 1{,}000$) and of the internal circulation on the development of temperature fields with time in both phases is investigated. The limits of applicability of existing approximate solutions of the given problem were also determined.

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