

much additional work is required before a conclusive generalization is in sight.

## NOTATION

$A_s$  = cross-sectional area of agglomerated mass of flow in pipe ( $L^2$ )  
 $C_d$  = drag coefficient (no dimension)  
 $D_p$  = average particle diameter ( $L$ )  
 $D_i$  = pipe diameter ( $L$ )  
 $f$  = friction factor (no dimension)  
 $g$  = gravitational acceleration ( $L\theta^{-2}$ )  
 $G_g$  = gas mass velocity in pipes ( $ML^{-2}\theta^{-1}$ )  
 $G_s$  = solid mass velocity in pipes ( $ML^{-2}\theta^{-1}$ )  
 $k$  = constant for a given particle size  
 $L$  = length of the transport line ( $L$ )  
 $R$  = solids to gas ratio (no dimension)  
 $U_a$  = average linear velocity of gas ( $L\theta^{-1}$ )  
 $U_s$  = average linear velocity of solid particles ( $L\theta^{-1}$ )  
 $V$  = total volume of solid and gas collected in the receiver ( $L^3$ )  
 $W_a$  = weight of gas ( $M$ )  
 $W_s$  = weight of solid ( $M$ )

## Greek Letters

$\Delta P_{aa}$  = pressure drop due to acceleration of gas ( $FL^{-2}$ )  
 $\Delta P_{as}$  = pressure drop due to acceleration of solid particles ( $FL^{-2}$ )  
 $\Delta P_d$  = pressure drop due to body drag ( $FL^{-2}$ )  
 $\Delta P_f$  = pressure drop due to friction ( $FL^{-2}$ )  
 $\Delta P_t$  = total pressure drop ( $FL^{-2}$ )  
 $\Delta U$  = slip velocity between gas and solid particles ( $L\theta^{-1}$ )  
 $\rho_g$  = density of gas ( $ML^{-3}$ )  
 $\rho_{da}$  = dispersed gas density ( $ML^{-3}$ )  
 $\rho_{ds}$  = dispersed solid density ( $ML^{-3}$ )  
 $\rho_s$  = solid particle density ( $ML^{-3}$ )

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# Effective Diffusivity of Packed Bed

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From time to time, A.I.Ch.E. Journal presents translations of certain technical articles written by our Japanese colleagues in their own language. These translations are made by Kenzi Etani, who received his B.S. in chemical engineering in 1953 at the Tokyo Institute of Technology and his M.S. in 1955 at M.I.T. He is associated with Stone & Webster and is an associate member of American Institute of Chemical Engineers. He is also a member of the Society of Chemical Engineers, Japan, and the Japan Oil Chemists' Society. His offer to help break down the language barrier is acknowledged.

Abstracts, notation, literature cited, tables, and figure captions not published here appear in English in the original paper. No figures will be reproduced in these translations.

The following article was published in *Chemical Engineering (Japan)*, **21**, pages 17-25 (1957).

The purpose of this experiment was to obtain the relation between effective diffusivity and fractional voids under nonflowing conditions. The results were compared with electric conductivity.

## APPARATUS AND EXPERIMENT

The apparatus used in this experiment is shown in Figure 1. The diameters of the tubes were 3.50 and 3.05 cm. The height of the packed bed was changed from 5 to 15 cm. All apparatus was kept at the constant temperature of  $40 \pm 0.2^\circ\text{C}$ . in a constant-temperature bath. Air having the same temperature was introduced at the top of the packed bed at the constant velocity of 55 cc./sec. Packing materials used were lead shot, ordinary sand, and crushed calcite. Diffusing materials used were benzene and water.

After the equipment had been immersed for 30 to 60 min. in the constant-temperature bath, air was introduced. Prior to the introduction of air, water or benzene diffuses upward and reaches the saturated state. This diffusion is expressed by the following basic equation:

$$\frac{\partial p}{\partial \theta} = \frac{De}{\epsilon} \frac{\partial^2 p}{\partial x^2} \quad (1)$$

$$\left. \begin{aligned} p &= p_s & x &= 0 & \theta &= \theta \\ \partial p / \partial x &= 0 & x &= X & \theta &= \theta \\ p &= p_s & x &= X & \theta &= \infty \\ p &= 0 & x &= X & \theta &= 0 \end{aligned} \right\} \quad (2)$$

If one assumes that packing material is located above the liquid, the relationship

between the concentration gradient at the liquid surface and time would be

$$\left[ \frac{\partial(p/p_s)}{\partial x} \right]_{x=0} = -\frac{2}{x} [e^{-(\pi/2)^2 \tau} + e^{-9(\pi/2)^2 \tau} + e^{-25(\pi/2)^2 \tau} + \dots] \quad (3)$$

where

$$\tau = De\theta/\epsilon X^2$$

Figure 2 illustrates the calculated results when  $De = 0.03$  sq. cm./sec.,  $X = 8, 12$ , and  $16$  cm., and  $\epsilon = 0.5$ . It is shown also that 5, 11.5, and 20 min. are required to reach a saturated state at a liquid surface for three different bed depths.

The time necessary for reaching the saturated state after the introduction of air would be solved under the following conditions:

$$\left. \begin{aligned} p &= p_s & x &= x & \theta &= 0 \\ p &= p_s & x &= 0 & \theta &= \theta \\ p &= p_s & x &= X & \theta &= 0 \\ p &= 0 & x &= X & \theta &< 0 \end{aligned} \right\} \quad (4)$$

This solution was obtained by Lee and others (8) and is given as follows:

$$\left[ \frac{\partial(p/p_s)}{\partial x} \right]_{x=X, \theta=\theta} = 1 - 2e^{-\pi^2 \tau} \quad (5)$$

$$\left[ \frac{\partial(p/p_s)}{\partial x} \right]_{x=X, \theta=\infty} = 1 - 2e^{-\pi^2 \tau} + 2e^{-4\pi^2 \tau} - 2e^{-9\pi^2 \tau} + 2e^{-16\pi^2 \tau} - 2e^{-25\pi^2 \tau}$$

The conclusions are shown in Figure 3. When air is introduced, the saturated state should be reached in a shorter time. To eliminate errors in vaporization of diffusing material during weighing and during saturation in the apparatus before the experiment is started, the following procedure was used.

Benzene or water was weighed after air had been introduced for 30 to 40 min. and again after 5 or 6 hr. The rate of diffusion was determined from the weight at the shorter period minus the weight at the longer period.

#### EXPERIMENTAL RESULTS

Diffusion takes place through the air, screen, packed bed, and turbulent air layer (Figure 4). If one calls the effective diffusivities of these layers  $D_f$ ,  $D_s$ ,  $D_e$ , and  $D_t$ , and the partial pressure of the air above the liquid in the container and at the top of the packed bed  $P_{a0}$  and  $P_{a1}$ , the following equation is derived:

$$N = \frac{\pi}{RT} \ln \frac{P_{a1}}{P_{a0}} \left/ \left[ \frac{\Delta X_a}{D_f} + \frac{\Delta X_s}{D_s} + \frac{\Delta X_e}{D_e} + \frac{\Delta X_t}{D_t} \right] \right. \quad (6)$$

With the total length  $\Delta X_{ap}$  and apparent diffusivity  $D_{ap}$  based on partial pressures  $P_{a0}$  and  $P_{a1}$  considered,

$$N = \frac{D_{ap}\pi}{RT \Delta X_{ap}} \ln \frac{P_{a1}}{P_{a0}} \quad (7)$$

From Equation (6) and (7)

$$\Delta X_{ap}/D_{ap} = \Delta X_a/D_f + \Delta X_s/D_s + \Delta X_e/D_e + \Delta X_t/D_t \quad (8)$$

$$1/D_{ap} = [\Delta X_a/D_f + \Delta X_s/D_s + \Delta X_t/D_t - (\Delta X_a + \Delta X_s + \Delta X_e)/D_e] 1/\Delta X_{ap} + 1/D_e \quad (9)$$

$D_{ap}$  was obtained experimentally. Since the depth of bed was changed and other conditions were kept constant,  $1/D_e$  was obtained at  $\Delta X_{ap}$  equals 0 in the plot of  $1/D_{ap}$  and  $1/\Delta X_{ap}$ .

Liquid-level change during experimentation was so small that the effect was neglected. The same fractional void was maintained throughout the experiments.

No effect due to the shape of packing materials was found. Crushed calcite and sand were previously screened into three groups: 6 to 8, 8 to 10, and 10 to 14 mesh.

Figures 5, 6, and 7 give the results.

#### EXPERIMENTS ON ELECTRIC CONDUCTIVITY

In the study of thermal diffusion the temperature gradient can be estimated from the electric-voltage gradient. It was assumed that diffusion through packing materials was similar to the case of electric conduction. Thus effective diffusivity obtained in this experiment was compared with electric conductivity, a bridge being employed to determine electric resistance. Electric conductivity of the solution in the packing material voids was determined. When shot lead was used as a packing material, electric resistance was so small that it could not be measured. No effect due to height was found. (See Figure 8.) The concentration of this solution was so high that surface electric conductivity on  $\zeta$  potential was neglected.

#### EXPERIMENTAL RESULTS

Wyllie studied the relationship between voids and electric conductivity in the packing bed, for example glass beads, sand (16) and ion exchange resin (11).

Electricity was conducted three ways: from packing material to solution, through packing material, and through solution. In the case of a nonconductor, the fractional area of fluid  $c$  equals the ratio of specific electric conductivity of the packed bed to that of the fluid itself; that is,  $k_e/k_f$ . The relationship between  $k_e/k_f$  and fractional void  $\epsilon$  is shown in Figure 10. Wyllie (16) mentioned that the following equations can be used: Archie's equation (1)

$$F = \epsilon^{-1.3} \quad (10)$$

Slawinski's equation (13)

$$F = (1.3219 - 0.3219\epsilon)^{2/\epsilon} \quad (11)$$

where  $F$  equals  $1/c$ . When the fractional void is small, Equation (10) can be applied, and when it is large, Equation (11) can be used. From Equations (10) and (11)

$$c = \epsilon^{1.3} \quad (12)$$

$$c = \epsilon/(1.3219 - 0.3219\epsilon)^2 \quad (13)$$

Equations (12) and (13) are shown in Figure 10.

In the case of diffusion  $c$  is considered to equal the ratio of effective diffusivity  $D_e$  to diffusivity  $D_f$  without packing. The ratio  $D_e/D_f$  vs. fractional void  $\epsilon$  is shown in Figure 11, in which data obtained by Piret and others (9) are also plotted. These experimental results show that the relationship between effective diffusivity and the fractional void in the packed bed will coincide with that of electric conductivity and fractional voids.

## COMMUNICATIONS TO THE EDITOR

### Physical Interpretation of the Relaxation Method in Heat Conduction

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The use of the relaxation method for numerical solutions of engineering problems was pioneered by Southwell (1). It was introduced to the field of steady state heat conduction by Emmons (2)

and further developed by Dusenberre (3). (See also references 4 to 7.) The essential requirement that the relaxation procedure must converge to give an exact solution of the difference equations was

demonstrated by Southwell (1) and more rigorously by Temple (10), both using the terminology of elastic-stress problems.

When the convergence proof is stated in thermal terms, it becomes apparent that this method is related to Prigogine's theorem on the minimum production of entropy (8). The one-dimensional problem of a slab with constant surface temperatures, which is divided into  $N$  layers for computational purposes, will be considered. For this system the theorem reduces to the assertion that for fixed values of temperatures  $t_0$  and  $t_N$  all other