Subscripts

1 = light component
2 = solvent
i = general pure component

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Laminar Dispersion in Capillaries: Part V.

Experiments on Combined Natural and Forced Convection in Vertical Tubes

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An approximate theory for transient dispersion with combined free and forced convection in vertical tubes is developed and tested experimentally. It is found that the dispersion coefficient in such systems depends on a parameter α, which is defined by Equation (38). The analytical expression developed for the dispersion coefficient, Equation (33), is found to be a reasonably good approximation in the range $-50 < \alpha < 300$.

Experimental results are reported for a rather wide range of parameters for tubes with both $1\frac{1}{2}$ and 5 mm. diameters.

It is found that the extent of dispersion is enhanced significantly when lighter fluid is on the bottom and displaces a heavier one. On the other hand, the dispersion coefficient is inhibited when the situation is reversed. It is found that these effects are not symmetrical with respect to the absolute magnitude of the parameter α as is illustrated in Figure 12.

Work on dispersion in tubes (*1* to *8, 11* to *13, 15* to *17*) has produced a very clear picture of this phenomenon in cases where natural convection effects are negligible. It remains to study the effects of density differences on dispersion in tubes and eventually to apply this new knowledge to porous media. In a previous report (*1*) it was indicated from the data on horizontal tubes that natural convection can both depress and accentuate the observed dispersion coefficient substantially as compared with that predicted by laminar flow theory which neglects natural convection

effects. One would expect natural convection to play a substantially more important role in dispersion in vertical systems and therefore this process is studied here. In this discussion all flows are isothermal and density differences are caused solely by concentration differences.

The exact mathematical solution of the transient convective diffusion equation with natural convection effects is very difficult to obtain since the velocity profile will be distorted from the normal parabolic shape. Hence the unsteady momentum and diffusion equations must be solved simultaneously in order to describe the dispersion process. However, the analysis of the vertical tube is simpler compared with that of the horizontal tube because of the symmetry with respect to gravity.

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The present work was undertaken to develop a mathematical model for laminar dispersion in vertical tubes with density differences present, and to investigate the parameters of the system; obtain experimental data when the heavier fluid displaces the lighter and vice versa over a wide range of variables involved; and examine the effect of tube diameter, if any, under otherwise identical flow conditions.

ANALYSIS FOR DISPERSION IN VERTICAL TUBES

It can be shown that the dispersion process depends on the parameters

$$N'_{Pe} = a U_m/D \quad N'_{Re} = a U_m \rho_0/\mu$$

$$N'_{Fr} = U_m^2/ag \quad N'_{Gr} = a^3 g \beta (C^+ - C_o^+)/\nu^2$$

Of these, the important parameters for the present system are N'_{Pe}, N'_{Gr}, and N'_{Re}. The Froude group is assumed to have a negligible effect on the system under consideration.

The Froude number is defined as the ratio of the inertial forces to the gravity force in the system and is usually important only when there is a free surface. In a completely submerged system, the effect of gravitational force may be eliminated by the proper choice of the static pressure of the fluid. This reasoning may not apply strictly here, however, since it is not clear that the initial interface between the two miscible solutions of different density does not have some of the properties of a free surface. It seems quite probable that inertial effects are significant in the unstable case of the upward displacement of heavier by lighter fluid.

To develop simple closed-form results which clearly indicate the most important features of the dispersion process in vertical tubes a successive approximation approach will be used. It is assumed that the flow is axially symmetric and laminar and due to the axial symmetry, the dispersion of solute will be independent of the angular coordinate. Also, for slow flows one can neglect inertial terms so that the velocity components can be taken as $u = u(t, r)$, $v = 0$, $w = 0$ as an approximation for the solution envisaged. The equation of continuity is satisfied identically, and the equations of motion and diffusion are

$$\frac{\partial u}{\partial t} = \frac{\nu}{r} \frac{\partial}{\partial r} \left(r \frac{\partial u}{\partial r} \right) - \frac{1}{\rho_o} \frac{\partial p}{\partial x} - \frac{\rho g}{\rho_o} \quad (1a)$$

$$0 = -\frac{\partial p}{\partial r} \quad (1b)$$

$$\frac{\rho}{\rho_o} = 1 - \beta (C^+ - C_o^+) \quad (1c)$$

where subscript o refers to a reference state, say the condition at the inlet, and

$$\beta = -\frac{1}{\rho_o} \frac{\partial \rho}{\partial C^+}\bigg|_{C^+ = C_o^+}$$

and

$$\frac{\partial C}{\partial t} + u \frac{\partial C}{\partial x} = D \left(\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C}{\partial r} \right) + \frac{\partial^2 C}{\partial x^2} \right) \quad (2)$$

The boundary conditions are

$C(0, x, r) = 0$ (initial condition)
$C(t, 0, r) = 1$ (step change at inlet)
$C(t, \infty, r) = 0$ (no material at $x = \infty$ if t finite)   (3)
$\frac{\partial C}{\partial r}(t, x, 0) = \frac{\partial C}{\partial r}(t, x, a) = 0$ (symmetry at center; solid wall at a)
     $u(a) = 0, \quad u(0) = $ finite

In the formulation given above, a step change is used to describe the inlet condition to the tube. This condition describes the situation when the tube is fed from a large

well-mixed reservoir of uniform concentration. Equation (2) in a coordinate system moving with the mean speed of flow is

$$\frac{\partial C}{\partial t} + (u - U_m) \frac{\partial C}{\partial x_1} = D \left(\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C}{\partial r} \right) + \frac{\partial^2 C}{\partial x_1^2} \right) \quad (4)$$

where

$$x_1 = x - U_m t$$

In order to solve Equation (4) for the concentration distribution, it is necessary to obtain the expression for the velocity distribution under the combined effect of forced and natural convection.

SOLUTION FOR u

As a first approximation, it is assumed that (15, 16)

$$C = C_m + \frac{\partial C_m}{\partial x_1} f(r) \quad (5)$$

where $f(r)$ is yet to be determined.

In solving the momentum equation, to eliminate $\partial p/\partial x$ one can differentiate Equations (1a) and (1b) with respect to r and x, respectively, and then equate the results for the second derivatives of p to get

$$\frac{\partial^2 u}{\partial t \partial r} = \frac{\partial}{\partial r} \frac{\nu}{r} \frac{\partial}{\partial r} r \frac{\partial u}{\partial r} + g\beta \frac{\partial C}{\partial r} \quad (6)$$

By using Equation (5), we get

$$\frac{\partial^2 u}{\partial t \partial r} = \frac{\partial}{\partial r} \frac{\nu}{r} \frac{\partial}{\partial r} r \frac{\partial u}{\partial r} + g\beta \frac{\partial f}{\partial r} \frac{\partial C_m}{\partial x_1} \quad (7)$$

and this result can be integrated to obtain

$$\frac{\partial u}{\partial t} = \frac{\nu}{r} \frac{\partial}{\partial r} r \frac{\partial u}{\partial r} + g\beta \frac{\partial C_m}{\partial x_1} f(r) + h \quad (8)$$

where h is a constant of integration. Since $\partial C_m/\partial x_1$ is a function of t, it is convenient to use the Duhamel theorem which states that

$$u = \frac{\partial}{\partial t} \int_0^t F(t - \lambda, \lambda, r) d\lambda \quad (9)$$

if we define F by

$$\frac{\partial F}{\partial t} = \frac{\nu}{r} \frac{\partial}{\partial r} r \frac{\partial F}{\partial r} + g\beta \frac{\partial C_m}{\partial x_1} (\lambda, x_1) f(r) + h \quad (10)$$

where in Equation (10) the parameter λ is considered to be a constant. Now let

$$F = F_s (\lambda, r) - F_t (\lambda, t, r)$$

where F_s and F_t satisfy

$$\frac{\nu}{r} \frac{d}{dr} r \frac{dF_s}{dr} + g\beta \frac{\partial C_m}{\partial x_1} f(r) + h = 0 \quad (11)$$

$$F_s (a) = \frac{dF_s}{dr} (0) = 0$$

and

$$\frac{\partial F_t}{\partial t} = \frac{\nu}{r} \frac{\partial}{\partial r} r \frac{\partial F_t}{\partial r} \quad (12)$$

$$F_t (0, r) = F_s$$

$$F_t (t, a) = \frac{\partial F_t}{\partial r} (t, 0) = 0 \quad (13)$$

It follows then by integrating Equation (11) and using Equation (9) to obtain u from F, that the part of the ve-

locity that is dominant for large values of time is

$$u_s - u_s(0) = -\frac{a^2}{4\nu} h\, y^2 -$$

$$\frac{g\beta a^2}{\nu} \frac{\partial C_m}{\partial x_1} \int_0^y \frac{1}{y} \left\{ \int_0^y f(y)\,y\,dy \right\} dy \quad (14)$$

where $u_s(0)$ is the velocity at the center of the tube and $f(y)$ is determined in the following manner. If one substitutes Equation (5) into Equation (4) and, following Taylor, neglects terms involving higher order derivatives of C_m with respect to x_1, then one gets

$$f(y) - f(0) = \frac{a^2}{D} \int_0^y \frac{1}{y} \left\{ \int_0^y y\,(u - U_m)\,dy \right\} dy \quad (15)$$

Now, to a first approximation, using

$$u = 2U_m(1 - y^2)$$

one gets

$$f(y) - f(0) = \frac{a^2 U_m}{4D} \left(y^2 - \frac{y^4}{2} \right) \quad (16)$$

$f(0)$ is determined from the condition. Since $C_m = 2\int_0^1 yC\,dy$, we see that multiplying Equation (5) by y and integrating from 0 to 1, Equation (17) follows.

$$\int_0^1 f(y)\,y\,dy = 0 \quad (17)$$

and one obtains

$$f(0) = -\frac{a^2 U_m}{12D}$$

so that

$$f(y) = \frac{a^2 U_m}{4D} \left(y^2 - \frac{y^4}{2} - \frac{1}{3} \right) \quad (18)$$

By substituting Equation (18) in Equation (14) one gets

$$u_s - u_s(0) = -\frac{a^2}{4\nu} hy^2 - \frac{g\beta U_m a^4}{4\nu D} \frac{\partial C_m}{\partial x_1} \left(\frac{y^4}{16} - \frac{y^6}{72} - \frac{y^2}{12} \right) \quad (19)$$

Since

$$U_m = 2\int_0^1 u_s\,y\,dy \quad (20)$$

by substituting for u_s from Equation (19) and integrating, it follows that

$$u_s(0) = U_m \left(1 - \frac{7}{1152}\alpha \right) + \frac{a^2}{8\nu} h \quad (21)$$

where

$$\alpha = \frac{g\beta a^4 \partial C_m/\partial x_1}{\nu D} \quad (22)$$

Use of $u_s(1) = 0$ in Equation (19) yields

$$u_s(0) = \left(\frac{a^2}{4\nu} h - \frac{5}{576} U_m \alpha \right) \quad (23)$$

and one can find h from Equations (21) and (23) to be given by

$$\frac{a^2}{8\nu} h = U_m \left(1 + \frac{3}{1152}\alpha \right) \quad (24)$$

and then it follows that

$$u_s(0) = 2U_m \left(1 - \frac{\alpha}{576} \right) \quad (25)$$

Finally, by substituting Equation (25) in Equation (19), one gets

$$u_s = 2U_m(1 - y^2) - \frac{\alpha U_m}{1152}(4 - 18y^2 + 18y^4 - 4y^6) \quad (26)$$

DETERMINATION OF THE DISPERSION COEFFICIENT

With the velocity distribution known to a first approximation, it remains to find the concentration distribution. For this purpose let

$$C = C_m + \frac{\partial C_m}{\partial x_1} g(r) \quad (27)$$

where $g(r)$ is yet an undetermined function. $g(r)$ represents a second approximation, whereas $f(r)$ is a first approximation in the concentration distribution expression. Equation (2), after multiplying by r and integrating with respect to r on $(0, a)$, yields

$$\frac{\partial C_m}{\partial t} = \left[D - \frac{2}{a^2} \int_0^a ug(r)\,r\,dr \right] \frac{\partial^2 C_m}{\partial x_1^2} \quad (28)$$

and from inspection of Equation (28) the dispersion coefficient is clearly

$$K = D - \frac{2}{a^2} \int_0^a ug(r)\,r\,dr \quad (29)$$

The function $g(r)$ is obtained in the same manner as $f(r)$ in Equation (15), and it is found that

$$g(y) - g(0) = \frac{a^2}{D} \int_0^y \frac{1}{y} \left\{ \int_0^y (u_s - U_m)\,y\,dy \right\} dy \quad (30)$$

where $g(0)$ is determined from $\int_0^1 g(y)\,y\,dy = 0$ to be

$$g(0) = -\frac{a^2 U_m}{12D} + \frac{\alpha U_m a^2}{1152D} \left(\frac{57}{240} \right) \quad (31)$$

so that

$$g(y) = \frac{a^2 U_m}{4D} \left(y^2 - \frac{y^4}{2} - \frac{1}{3} \right) -$$

$$\frac{\alpha U_m a^2}{1152D} \left(-\frac{57}{240} + y^2 - \frac{9}{8}y^4 + \frac{1}{2}y^6 - \frac{1}{16}y^8 \right) \quad (32)$$

If Equations (26) and (32) are substituted into Equation (29), the dispersion coefficient, K_{nat}, which includes the effects of buoyancy forces, is given to a first approximation by

$$\frac{K_{nat}}{D} = 1 + \frac{N_{Pe}^2}{192} \left(1 - \frac{11}{2880}\alpha + 4.16 \times 10^{-6} \alpha^2 \right) \quad (33)$$

where

$$\alpha > 0 \text{ for heavy fluid on bottom}$$
$$\alpha < 0 \text{ for heavy fluid on top}$$

In order to calculate α, $\partial C_m/\partial x_1$ has to be evaluated. For small buoyancy effects, $\partial C_m/\partial x_1$ calculated from Taylor's solution (15) should give satisfactory results. Since

$$C_m = \frac{1}{2} \text{erfc} \frac{x_1}{2\sqrt{Kt}} \quad (34)$$

it follows that

$$\frac{\partial C_m}{\partial x_1} = -\frac{1}{2} \left(\frac{2}{\sqrt{\pi}} \right) \left(e^{-(x-U_m t)^2/4Kt} \right) \left(\frac{1}{2\sqrt{Kt}} \right) \quad (35)$$

From Equation (35), using the dimensionless quantities

$$\tau = \frac{tD}{a^2}, \quad X = \frac{xD}{2a^2 U_m} \tag{36}$$

and Aris' expression (4) for the dispersion coefficient

$$\frac{K}{D} = \left(1 + \frac{N_{Pe}^2}{192}\right)$$

one obtains the expression

$$\frac{\partial C_m}{\partial x_1} = -\frac{1}{d\sqrt{\pi}} \frac{\exp - \left[\dfrac{N_{Pe}^2 (X - \tau/2)^2}{4\tau(1 + N_{Pe}^2/192)}\right]}{\tau^{1/2}\left(1 + \dfrac{N_{Pe}^2}{192}\right)^{1/2}} \tag{37}$$

If β is a constant then α can be written in the form

$$\alpha = -\frac{1}{16} \frac{\dfrac{g}{\rho}\dfrac{\Delta\rho}{\Delta C_m}\dfrac{\partial C_m}{\partial x_1} d^4}{\nu D}$$

$$= -\frac{1}{16}\left(\frac{g(\Delta\rho/\rho)d^3}{\nu^2}\right)\left(\frac{\nu}{D}\right)\left(\frac{d}{\Delta C_m}\right)\frac{\partial C_m}{\partial x_1}$$

$$= -\frac{1}{16}(N_{Gr} N_{Sc})\left(\frac{d}{\Delta C_m}\right)\frac{\partial C_m}{\partial x_1} \tag{38}$$

and

$$N_{Gr} > 0 \text{ for heavy fluid on bottom}$$
$$< 0 \text{ for heavy fluid on top}$$

It is obvious that the value of α obtained from Equations (37) and (38) is not strictly constant. However, for the purpose of obtaining the simplest result which still describes most of the important aspects of the effect of natural convection on the dispersion process, α can be written as

$$\alpha = \frac{g\left(-\dfrac{1}{\rho}\dfrac{\Delta\rho}{\Delta C_m}\right)\left(\dfrac{\Delta C_m}{\Delta x_1}\right)d^4}{16\nu D}$$

$$\alpha = \frac{(N_{Gr} N_{Sc})}{16\left(\dfrac{\Delta x_1}{d}\right)} \tag{39}$$

where Δx_1 = mixing length over which most of the concentration change takes place. If to a first approximation, the Taylor-Aris solution is used for calculating this mixing length for a 90 to 10% concentration change, the result is

$$\frac{\Delta x_1}{d} = 1.81\sqrt{\tau}\left(1 + \frac{N_{Pe}^2}{192}\right)^{1/2} \tag{40}$$

In principle, then we should be able to predict the effects of natural convection due to concentration-density differences by use of Equations (33), (39), and (40).

It is noted that the dispersion coefficient calculated from Equation (33) for positive values of α increases for values of $\alpha > 460$, which is contrary to what is to be expected. In view of the many simplifications made, it is apparent that the above analysis would be valid only for small values of α where the buoyancy effects are relatively small.

The conventional method of improving the analysis by making additional iterations to improve $f(r)$ in Equation 5 by use of Equation (15) is probably not warranted unless a more refined model is used. Such refinements could include viscosity variations with concentration and inertial

effects which will yield x variation of the velocity profile. The inclusion of inertial effects increases the complexity of the analytical problem very substantially. However, a first estimate of the effect of viscosity variations can be obtained relatively easily.

Viscosity variation with concentration could have been considered in the present study by defining a function ϕ such that

$$u_s = \int_0^y \left[\frac{d\phi/dy}{\mu/\mu_0}\right] dy$$

Then the expression for ϕ is identical to that given by Equation (26) and the new u_s is given by

$$u_s = -\int_0^y \frac{4U_m y + \dfrac{\alpha U_m}{1152}(-36y + 72y^3 - 24y^5)}{\mu/\mu_0} dy$$

To a first approximation, one finds

$$\frac{\mu}{\mu_0} = 1 - \left[\frac{\alpha N_{Re}\left(-\dfrac{1}{\mu_0}\dfrac{\partial\mu}{\partial C}\right)_{C=1}}{N_{Gr}}\right]\left(y^2 - \frac{y^4}{2} - \frac{1}{3}\right)$$

The viscosity distribution calculated by this formula, in conjunction with Table 1, shows that the variation for a given N_{Gr} is greater for runs at higher N_{Re} and smaller values of τ. For example, roughly a 5% variation occurs in run 4 with $N_{Gr} = 128$ and it is about $2\frac{1}{2}\%$ for run 5. In contrast, the variation is on the order of 0.3% for run 14. The viscosity effect is carried through to alter $g(y)$ and K as given in Equation (29), but in view of the small percentage variation involved it seems safe to conclude that viscosity effects are not a dominant factor in the present experiments.

As long as the natural convection effects are large (and in over 90% of our experiments the effects are greater than 20%), the viscosity contribution can safely be neglected. In the majority of the runs the natural convection effects are pronounced and affect the dispersion coefficient by factors of 2 to 10. When the natural convection effects are small the viscosity corrections to the velocity distribution should probably be carried through for improved accuracy.

EXPERIMENTAL WORK

Optical Method for Measuring Concentration

The equipment used for measuring concentration of solute along the tube was the same as that used for horizontal tube experiments, the details of which are given elsewhere (11, 13). The main objective of the experimental work was to measure dispersion when resident and invading fluids have different densities. Clearly, two basically different physical situations can exist wherein the heavy fluid can displace the lighter or vice versa. The displacing fluid always enters from the bottom of the tube.

Selection of Solute

One problem which was investigated rather thoroughly was that of finding a colored solution in which the density could be varied independently of the diffusivity and the optical characteristics, and it was found that the best answer was to use two solutes, one colored and the other colorless. The two solutes should have the same diffusivity and the concentration of the colorless one should not affect the absorptivity of the colored one. The solutes should also have reasonable solubilities since it is desirable to vary the density widely.

Pontamine blue dye, which was used for all our previous experiments, was unsuitable since its diffusivity decreases markedly in the presence of salts (18), as indicated in Figure 13b.

Solutions of $KMnO_4$, $K_2Cr_2O_7$ and water-soluble compounds of Cu, Fe, Ni, and Co were tried. Absorbance data were obtained for various concentrations of these compounds in a 1.5-mm. I.D. capillary tube. Suitable Kodak-Wratten filters were used in each

case in order to get the transmitted light in the wavelength band corresponding to the maximum absorption of light. These are as follows:

Compounds	λ_{max}, $m\mu$, (9, 10, 14)	Kodak-Wratten filter No.
$CuSO_4 \cdot 5H_2O$	800	53 + 25
$FeCl_3 \cdot 6H_2O$	360	48 + 49B
$NiSO_4 \cdot 6H_2O$	650	70
$CoCl_2 \cdot 6H_2O$	510	74
$KMnO_4$	530	74
$K_2Cr_2O_7$	460	48 + 49B

In addition, some neutral density filters were also used to obtain the light intensity in the desired range of the instrument.

It was observed from these data (see Figure 13a) that the absorbance of solutions of the metals Cu, Fe, Ni, and Co does not show appreciable change over a wide concentration range and thus they were found to be unsuitable. The optimum concentrations for $KMnO_4$ and $K_2Cr_2O_7$ solutions were found to be 1.0 and 6 g./liter, respectively. An advantage of using $K_2Cr_2O_7$ solution is that high concentrations of the solute can be used, thereby giving higher values of the density-difference $\Delta \rho/\rho$ between the displacing and displaced fluids. A still higher value of $\Delta \rho/\rho$ could be obtained by addition of K_2SO_4 of the required amount in the 6 g./liter solution of $K_2Cr_2O_7$.

The absorbance of $K_2Cr_2O_7$ solution of a given concentration is not affected by the amount of K_2SO_4 added (see Figure 13b.) Also, the reported values of the diffusion coefficients of $K_2Cr_2O_7$ and K_2SO_4 in distilled water are nearly the same and their variations over the concentration range are very small. The reported values of D (International Critical Tables, 5, 69) for both of these compounds are indicated below:

$K_2Cr_2O_7$		K_2SO_4	
Conc., g./liter	$D \times 10^5$ (at $18°$C.), sq. cm./sec.	Conc., g./liter	$D \times 10^5$ (at $20°$C.), sq. cm./sec.
2	1.24	0.9	1.20
5	1.17	9	1.10
15	1.17	45	1.00

Therefore it was decided to use $K_2Cr_2O_7$ (6 g./liter) in distilled water to displace or be displaced by K_2SO_4 solutions of various concentrations depending upon the desired value of $\Delta \rho/\rho$.

Measurement of the Diffusion Coefficient of $K_2Cr_2O_7$ in K_2SO_4 Solutions

It was thought advisable to determine our own values of D for $K_2Cr_2O_7$ in K_2SO_4 solutions. One of the methods for finding the diffusion coefficient is to use Taylor's diffusion equation, when two fluids of equal density and viscosity displace each other under the restrictions found previously from our numerical and experimental work on horizontal tubes (2, 13), that the dimensionless time of flow $\tau(= tD/a^2)$ be greater than 1 and Peclet number be greater than 100. Taylor's equation is written as

$$C_m = \frac{1}{2} \, erfc \, \frac{(x - Ut)}{\sqrt{4kt}}, \quad k = \frac{a^2 U^2}{48D}$$

The mixing length (L') for 10 to 90% concentration change is found from the above equation as $k = L'^2/(13.1t)$. Hence, for a given Peclet number and time of flow, values of D can be calculated by measuring this mixing length. For Peclet numbers less than 100, Aris' modification of the dispersion coefficient $K = k + D$ should be used for finding the value of D.

Displacement experiments were carried out in 1.5-mm. I.D. horizontal tubes. A maximum of three tubes was used in series. In these experiments, $K_2Cr_2O_7$ solution (6 g./liter) displaced K_2SO_4 solution (5.3 g./liter). The experimental procedure for these runs was exactly the same as that used in the previous work on horizontal tubes. The density difference $\Delta \rho/\rho$ for the two solutions was measured to be 1.4×10^{-4}.

The data from these runs were plotted on probability paper, as concentration vs. distance in centimeters from the tube inlet. Most of the data points were found to lie on a straight line, and met the symmetry criterion of having 50% concentration at the point $x/(2Ut) = 0.5$, where x is the distance from the inlet. The distances, corresponding to the concentration change from 10 to 90%, were measured from these graphs and then the values of the dispersion coefficient k were calculated with Taylor's equation.

It was also desirable to know the variation of D in a lower concentration range. Hence, some experiments were conducted in which $K_2Cr_2O_7$ solution (3 g./liter) displaced K_2SO_4 solution (2.65 g./liter). The value of $\Delta \rho/\rho$ for these two solutions was

measured to be 1.07×10^{-4}. Within the experimental accuracy, values of D_{obs} were about the same as those in the former case. An average value of D for all these runs was thus taken to be 1.08×10^{-5} sq. cm./sec. From this the average Schmidt number was found to be 893.

Experiments in Vertical Tubes

Displacement experiments were carried out in 1.5- and 5-mm. I.D. capillary tubes; the length of each tube was about 85 cm. They were connected to the syringe through a bent capillary piece, one end of which was connected to the syringe tip and the other end was joined to a glass joint about 2 cm. long which had ground ends, and an inside diameter equal to the outside diameter of the capillary tube. The straight dispersion tube also had a ground end which fitted this joint. The injection system used in the displacement experiments was the same as that used in the horizontal tube experiments.

Calibration curves for Pontamine dye and $K_2Cr_2O_7$ solution were prepared separately. Recalibrations were made whenever changes in the light source were made. One-hundred percent concentration for Pontamine dye and $K_2Cr_2O_7$ corresponds to 0.2 and 6 g./liter, respectively, in 1.5-mm. tubes. For 5-mm. tubes, 100% concentration of $K_2Cr_2O_7$ is 2 g./liter.

In the preparation for an individual run, the tube was filled with the fluid to be displaced and the top end was sealed to prevent drainage. The tube was then held vertically in the angle section of the optical system by clamps and joined to the syringe connection while care was taken to avoid air entrapment at that section. After the entrance end of the tube was connected to the syringe connection, the seal at the other end of the tube was then broken. The two fluids, that is, in the syringe connection and in the capillary tube, formed a reasonably sharp interface of only a few millimeters width, thereby providing a step change condition for the inlet of solute. The displacement was carried out for a given length of time and after the flow was terminated, the discharge end of the capillary tube was sealed again. The tube was detached from the syringe connection and concentration of solute was measured along its length by sliding the tube in the vertical angle section.

EXPERIMENTAL RESULTS

It was desired to make some runs at first with two solutions of equal densities in order to compare the results with those of the horizontal tubes. Plots of concentration
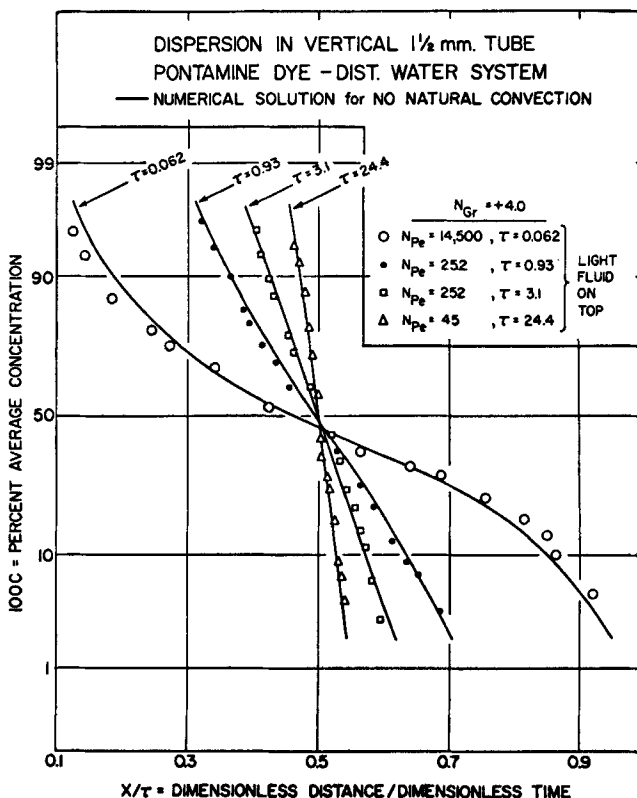

Fig. 1. Comparison of experimental data with theory for dispersion in vertical tubes when natural convection is essentially negligible. The agreement is considered to be excellent.
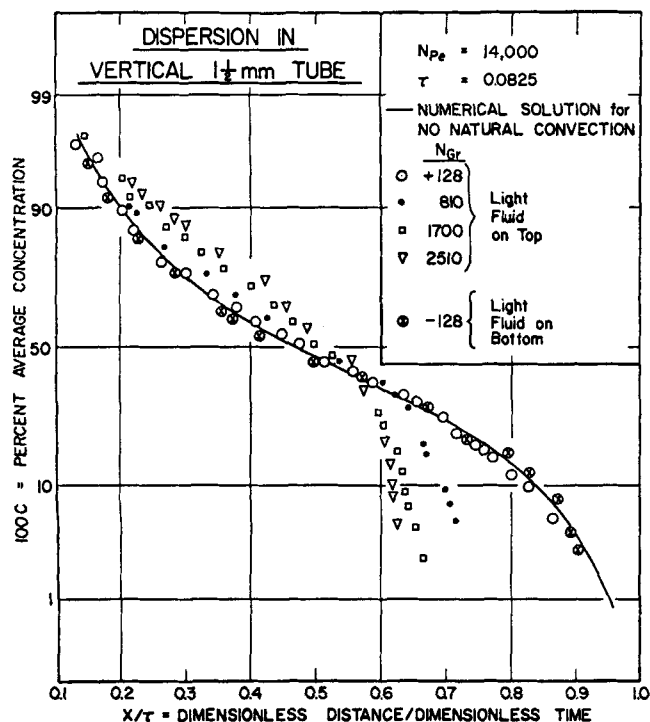
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Fig. 2. Effect of Grashof number on dispersion in vertical tubes at small values of τ where molecular diffusion play a minor role. Note that natural convection inhibits axial dispersion most profoundly in the region nearest to the forward end of the paraboloid.
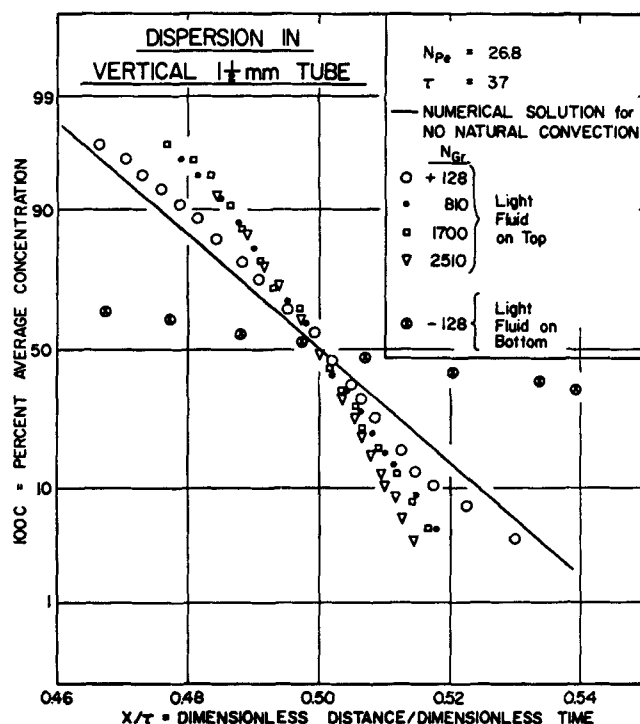

Fig. 3. Effect of Grashof number on dispersion in vertical tubes after long times of flow when molecular diffusion has played an important role.

vs. X/τ on probability coordinates in Figure 1 correspond to the case in which Pontamine dye (0.2 g./liter) displaced distilled water. Some runs were also made with K_2SO_4

(5.3 g./liter) displacing $K_2Cr_2O_7$ (6 g./liter) in the capillary tube. These results satisfied us that there were no experimental difficulties in obtaining identical dispersion results

TABLE 1. COMPARISON OF EXPERIMENTAL DATA WITH THE MATHEMATICAL ANALYSIS

(Vertical 1.5-mm. tube)

No.	τ	N_{Pe}	$\Delta x_1/d$	$\Delta (X/\tau)_{num}$ for 90 to 10% conc. change	$N_{Gr} = +128$			$N_{Sc} = 893$	$N_{Gr} = -128$			$N_{Sc} = 893$
					α	$\Delta (X/\tau)_{obs}$	K_{nat}/K_{num}	K_{obs}/K_{num}	α	$\Delta (X/\tau)_{obs}$	K_{nat}/K_{num}	K_{obs}/K_{num}
1	0.0825	14,000	388	0.635	18.4	0.63	0.93	0.98	18.4	0.645	1.07	1.03
2	0.241	5,050	294	0.49	24.3	0.465	0.91	0.90	24.3	0.555	1.10	1.28
3	0.0825	1,680	43.5	0.635	165	0.475	0.48	0.56	165	(?)	1.74	
4	0.241	1,680	98	0.49	73	0.41	0.74	0.70	73	0.595	1.30	1.48
5	0.93	1,680	211	0.27	34	0.241	0.87	0.80	34	0.285	1.13	1.11
6	0.93	234	29.5	0.27	242	0.15	0.31	0.31	242	(?)	2.17	
7	2.8	234	51.2	0.154	140	0.0945	0.55	0.38	140	0.2875	1.61	3.48
8	6.2	234	76.2	0.102	94	0.082	0.67	0.65	94	0.468	1.40	9.3
9	2.8	89	19.7	0.158	363	0.087	0.165	0.31	363	0.75	2.89	22.6
10	6.2	89	29.4	0.108	243	0.066	0.33	0.37	243	0.37	2.14	11.8
11	12.4	89	41.5	0.076	172	0.047	0.48	0.38	172	0.20	1.77	6.92
12	6.2	26.8	9.8	0.12	728	0.107	0.58	0.79	728	(?)	4.92	
13	12.4	26.8	13.9	0.086	515	0.067	0.33	0.61	515	0.585	3.43	46.4
14	37	26.8	24	0.049	298	0.0375	0.39	0.59	298	0.186	2.19	14.4

(?) $(X/\tau)_{obs}$ could not be measured for 90 to 10% concentration change.

No.	$N_{Gr} = 810$			$N_{Sc} = 898$	$N_{Gr} = 1700$			$N_{Sc} = 904$	$N_{Gr} = 2510$			$N_{Sc} = 910$
	α	$\Delta (X/\tau)_{obs}$	K_{nat}/K_{num}	K_{obs}/K_{num}	α	$\Delta (X/\tau)_{obs}$	K_{nat}/K_{num}	K_{obs}/K_{num}	α	$\Delta (X/\tau)_{obs}$	K_{nat}/K_{num}	K_{obs}/K_{num}
1	117	0.475	0.61	0.56	248	0.385	0.31	0.37	368	0.3275	0.16	0.265
2	155	0.3025	0.51	0.38	326	0.2275	0.195	0.216	486	0.180	0.056	0.135
3	1,045*	0.206		0.105	1,520*	0.210		0.11	2,270*	0.140		0.048
4	465	0.176	0.12	0.13	890*	0.118		0.059	1,320*	0.101		0.042
5	216	0.152	0.37	0.32	455	0.093	0.123	0.12	678	0.086	0.32	0.102
6	1,550*	0.068		0.064	3,260*	0.064		0.056	4,850*	0.0505		0.035
7	890*	0.0375		0.059	1,880*	0.0425		0.076	2,790*	0.0245		0.025
8	598	0.0283	0.213	0.077	1,260*	0.0202		0.039	1,870*	0.017		0.028
9	2,310*	0.0545		0.119	4,880*	0.0605		0.147	7,250*	0.055		0.12
10	1,550*	0.037		0.118	3,270*	0.035		0.105	4,860*	0.0305		0.08
11	1,100*	0.022		0.084	2,310*	0.0172		0.051	3,440*	0.0148		0.038
12	4,640*	0.084		0.49	9,780*	0.086		0.51	14,550*	0.085		0.50
13	3,280*	0.0525		0.37	6,900*	0.048		0.31	10,300*	0.0465		0.29
14	1,900*	0.0268		0.30	4,000*	0.026		0.28	5,950*	0.0248		0.25

*Mathematical analysis does not apply for high values of α.

TABLE 2. COMPARISON OF EXPERIMENTAL DATA WITH THE MATHEMATICAL ANALYSIS

(Vertical 5-mm. tube)

No.	τ	N_{Pe}	$\Delta x_1/d$	$\Delta(X/\tau)_{num}$ for 90 to 10% conc. change	$N_{Gr} = 148$ α	$\Delta(X/\tau)_{obs}$	K_{nat}/K_{num}	$N_{Sc} = 842$ K_{obs}/K_{num}	$N_{Gr} = 2540$ α	$\Delta(X/\tau)_{obs}$	K_{nat}/K_{num}	$N_{Sc} = 856$ K_{obs}/K_{num}
1	0.0825	4,500	118	0.635	66	0.595	0.93	0.88	1,150*	0.204		0.103
2	0.0825	1,350	36	0.635	218	0.425	0.36	0.45	2,680*	0.096		0.023
3	0.241	1,350	80	0.49	98	0.37	0.66	0.57	1,560*	0.070		0.0205
4	0.0825	720	19	0.635	414	0.41	0.133	0.42	5,040*	0.084		0.0175
5	0.241	225	13.2	0.49	590	0.31	0.193	0.40	9,400*	0.083		0.029
6	0.93	225	28.5	0.27	274	0.144	0.265	0.285	4,790*	0.031		0.013
7	2.8	90	20	0.158	392	0.084	0.16	0.28	6,840*	0.0305		0.037
8	0.93	45	6.3	0.28	1,270*	0.166		0.35	22,100*	0.135		0.23
9	6.2	27	9.9	0.12	788	0.0605	0.66	0.255	13,700*	0.0545		0.206
10	12.4	27	14	0.086	556	0.053	0.34	0.38	9,700*	0.0435		0.26
11	6.2	9	5.4	0.195	1,460*	0.158		0.66	25,200*	0.182		0.87

*Mathematical analysis does not apply for high values of α.

No.	τ	N_{Pe}	$\Delta x_1/d$	$\Delta(X/\tau)_{num}$ for 90–10% conc. change	$N_{Gr} = -148$ α	$\Delta(X/\tau)_{obs}$	K_{nat}/K_{num}	$N_{Sc} = 842$ K_{obs}/K_{num}
1	0.0825	4,500	118	0.635	66	0.88	1.44	1.58
2	0.0825	1,350	36	0.635	218	(?)		
3	0.241	1,350	80	0.49	98	0.84	1.42	2.94
4	0.241	225	13.2	0.49	590	(?)		
5	0.93	225	28.5	0.27	274	(?)		
6	2.8	90	20	0.158	392	0.75	3.1	22.5
7	6.2	27	9.9	0.12	788	(?)		
8	12.4	27	14	0.086	556	(?)		

(?) $\Delta(x/\tau)_{obs}$ could not be measured for 90 to 10% concentration change.

with either vertical or horizontal systems as long as no natural convection effects were present.

Some of the data obtained in 1.5-mm. tubes with two fluids of different densities are plotted on probability coordinates in Figures 2 to 6. The ordinate represents the concentration of the displacing fluid injected from the syringe. In these experiments, $K_2Cr_2O_7$ solution (6 g./liter) and K_2SO_4 solutions of different concentrations were used, as indicated in the table below. Density and viscosity data for the two solutions (at 25°C.) were obtained from International Critical Tables (3, 88, 92).

Fig. 4. Comparison of results of dispersion experiments with 1½- and 5-mm. tubes at small values of τ.

Solute	Conc., g./liter	ρ, g./ml.	$\Delta\rho/\rho \times 10^4$	$\nu \times 10^2$, sq. cm./sec.	N_{Gr} in 1.5-mm. tube
$K_2Cr_2O_7$	6	1.00146	—	0.918	—
K_2SO_4	10	1.00506	36	1.015	128
K_2SO_4	35	1.02475	230	1.045	810
K_2SO_4	70	1.05165	490	1.088	1,700
K_2SO_4	105	1.07782	735	1.1132	2,510

Experimental data were obtained in the 1.5-mm. tube at given τ and N_{Pe} values for the four different Grashof numbers above, for the case when the heavier fluid displaced the lighter one (that is, lighter fluid on top). The value of the Grashof number was calculated on the basis of an average value of the viscosity and density of the two fluids. For the case when the lighter fluid displaced the heavier one (that is, heavier fluid on top), data were obtained only for $N_{Gr} = -128$.

Complete tabulated and plotted data may be found elsewhere (12) where τ varied from 0.08 to 37 and N_{Pe} varied from 14,000 down to 27, but the shorter times are associated with the higher N_{Pe}.

It was desirable to obtain data in the 5-mm. tube at the same values of N_{Gr} as those used in the 1.5-mm. tube to see if the tube diameter is a parameter of the system as appeared to be the case in our earlier work with horizontal tubes. By adjusting the densities of the solutions used, two pairs were made up so that the N_{Gr} in the 5-mm. tube equalled as nearly as possible the highest and lowest N_{Gr} in the 1.5-mm. tube. The solution properties are shown in in the table below and $\Delta\rho/\rho$ are the measured values.

Solute	Conc., g./liter	ρ, g./ml.	$\Delta\rho/\rho \times 10^4$	$\nu \times 10^2$ sq. cm./sec.	N_{Gr} in 5-mm. tube
$K_2Cr_2O_7$	2.00	0.99843	—	0.90	—
K_2SO_4	1.82	0.99853	1.0	0.92	148
K_2SO_4	3.92	1.00021	17.8	0.95	2,540

Experimental data were obtained for the 5-mm. tube at various τ and N_{Pe} values for both Grashof numbers when the heavier fluid was on the bottom, but only for $N_{Gr} = -148$
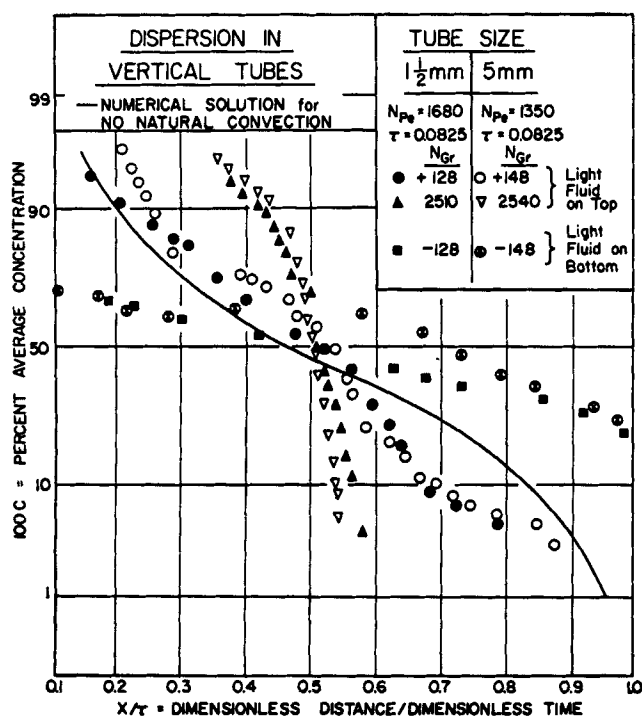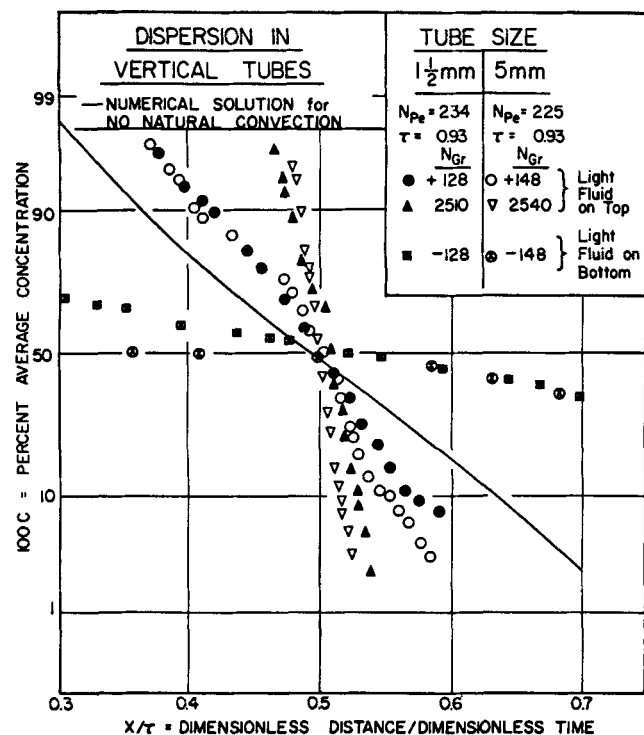
Fig. 5. Comparison of results of dispersion experiments with 1½- and 5-mm. tubes at intermediate values of τ.

when the heavier fluid was on top. These data are plotted on probability coordinates in Figures 4 to 6. The ordinate represents the concentration of the displacing fluid. Additional tabulated and plotted data may be found elsewhere (12), where in τ varied from 0.08 to 12.4 and N_{Pe} varied from 4,500 to 9.

The range of data for the two tube sizes was restricted by the physical limitations of the apparatus. However the data were taken systematically at each Grashof number so that: (1) a range of τ was covered for several, essentially fixed, N_{Pe}; (2) a range of N_{Pe} for fixed τ was studied; and (3) data for the 5- and 1.5-mm. tubes overlap signifi-
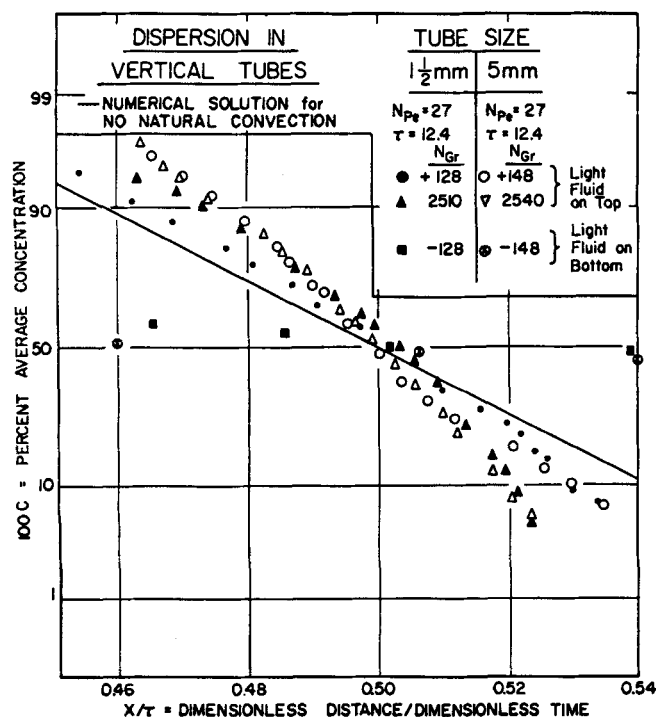
Fig. 6. Comparison of results of dispersion experiments with 1½- and 5-mm. tubes at large values of τ.
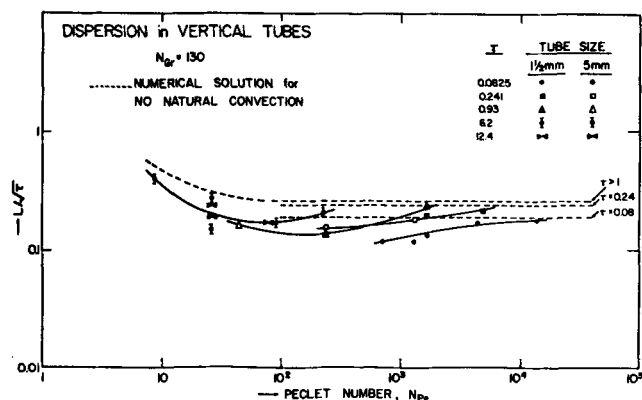
Fig. 7. Effect of N_{Pe} and τ on mixing length in 1½- and 5-mm. tubes at $N_{Gr} = 130$.

cantly at essentially fixed τ and N_{Pe}, so that all three parameters were fixed for several pairs of runs in the large and small tubes.

DISCUSSION OF RESULTS AND CONCLUSIONS

Experimental data, which were taken over a wide range of variables, for dispersion in vertical tubes with natural convection are plotted on probability coordinates in Figures 1 to 6. These plots compare the experimental results with the numerical solution obtained in Parts I and II (2, 3) for no natural convection. When reference is made to the numerical solution in subsequent discussion, it is understood that natural convection was neglected in obtaining it. Clearly, in the cases shown in Figure 1, natural convection is essentially negligible and the theory and experiments agree very well.

The data in Figures 2 and 3 differ significantly from the numerical solution for larger values of N_{Gr}. Note that for the heavy fluid on the bottom ($\alpha > 0$), the mixing length over which appreciable concentration change takes place decreases as N_{Gr} increases at a given τ and N_{Pe}. This is correctly predicted by Equation (33) and by the expression for the velocity profile, Equation (26), which indicates that the velocity profile is flattened and thus causes less dispersion to occur. For the light fluid on the bottom ($\alpha < 0$), velocity is enhanced in the center and retarded near the wall. Hence this elongated velocity profile increases dispersion.

It is obvious that a heavier fluid on top of, and being displaced by, a lighter one is an unstable situation in which natural convection would be expected to play a major role. Perhaps it is not as obvious intuitively that natural convection will inhibit dispersion when a heavy fluid displaces a light one in a vertical system. However, one can understand easily that this is exactly the behavior which
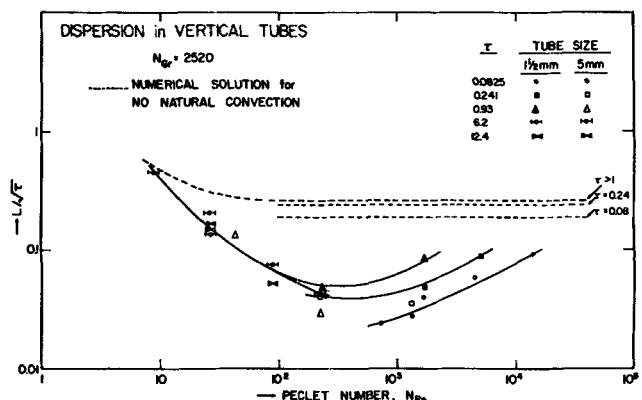

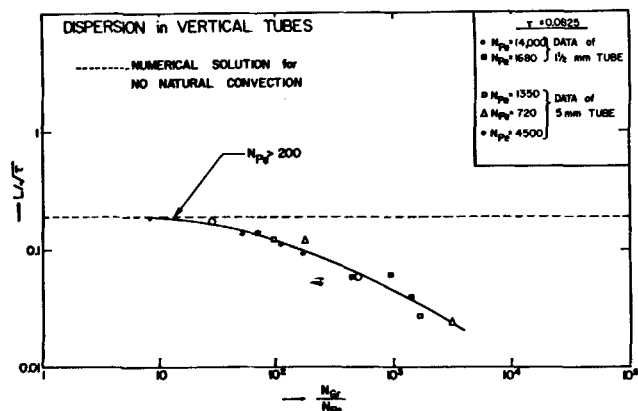**Fig. 8. Effect of N_{Pe} and τ on mixing length in 1½- and 5-mm. tubes at $N_{Gr} = 2,520$.**

Fig. 9. Effect of N_{Gr}/N_{Re} and N_{Pe} on mixing length in 1½- and 5-mm. tubes at small values of τ. Note that results are essentially independent of N_{Pe}.

Fig. 11. Effect of N_{Gr}/N_{Re} on mixing length in 1½- and 5-mm. tubes for large values of τ.

should be observed by considering that at small values of τ pure laminar convection causes a paraboloid finger of the displacing fluid to grow into the displaced fluid. Thus the central core is made rich in displacing fluid by convection. Subsequently, transverse molecular diffusion reduces the radial concentration gradients. Since the highest concentration of displacing, or bottom, fluid exists in the central core, gravity will accelerate this core if the lighter fluid is on the bottom and retard the core if the heavier fluid is on the bottom. In the former case, the velocity profile is obviously elongated and with the heavier fluid on bottom it is flattened. Consequently, if the lighter fluid is on bottom, the dispersion coefficient should be increased by natural convection; if the heavier fluid is on bottom the dispersion coefficient is decreased.

The observed dispersion coefficients are shown in Tables 1 and 2 and are compared with those calculated from Equation (33) as well as from the numerical results for no natural convection. The data are also plotted in Figure 12. The ratio K_{obs}/K_{num} is equal to the square of the ratio of the corresponding lengths over which 90 to 10% concentration change takes place. When the concentration vs. X/τ plot is not a straight line on the probability graph, this is merely a number useful mainly for comparison. In determining the values of α, $\Delta x_1/d$ was calculated with Equation (40) for $\tau > 1$, and for $\tau < 1$, it was calculated from the numerical solution for no natural convection.

In Tables 1 and 2 the adjacent columns of K ratios should be identical if Equation (33) holds, and the extent to which they differ is a measure of the inadequacy of the equation.

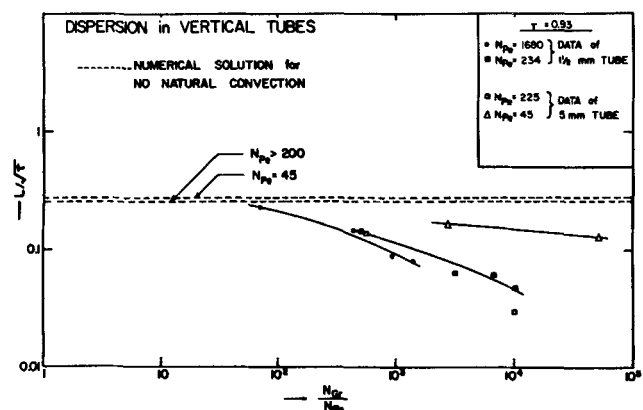As expected, the experimental results and mathematical analysis agree quite well for small values of α only. The

best fit lies in the region $-50 < \alpha < +300$. One would also expect the analysis to be adequate only for relatively large values of τ as is the case when natural convection is negligible. For large values of α, this analysis does not describe the phenomena adequately due undoubtedly to the many simplifying assumptions made.

It can be seen in Figures 4, 5, and 6 that the experimental results obtained in the 1.5- and 5-mm. tubes at essentially the same values of N_{Gr}, N_{Pe}, and τ, agree reasonably well at high Peclet numbers but differ to some extent at low Peclet values. This tends to verify the conclusions of the dimensional analysis which showed that only three dimensionless groups are important and that tube size is not a parameter. The Schmidt number was not varied in these experiments.

In Figures 7 and 8 plots of $L/\sqrt{\tau}$ vs. N_{Pe} are made for $N_{Gr} = 130$ and 2,520, respectively. It is observed that these plots, for a given value of τ, pass through a minimum and approach the numerical solution asymptotically both at very high and very low values of N_{Pe}. At high N_{Pe} values, the amount of solute transported by forced convection greatly exceeds that by pure axial diffusion and natural convection combined, that is, the high forced velocities make the small natural convection velocities unimportant. However, at very low values of N_{Pe}, the explanation is not as clear. With the heavy fluid on the bottom and at zero velocity $(N_{Pe} = 0)$ the only mechanism for dispersion is

Fig. 10. Effect of N_{Gr}/N_{Re} on experimental mixing length results for 1½- and 5-mm. tubes at $\tau = 0.93$.

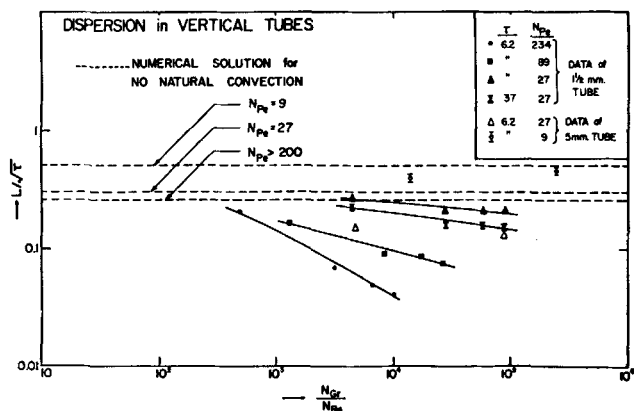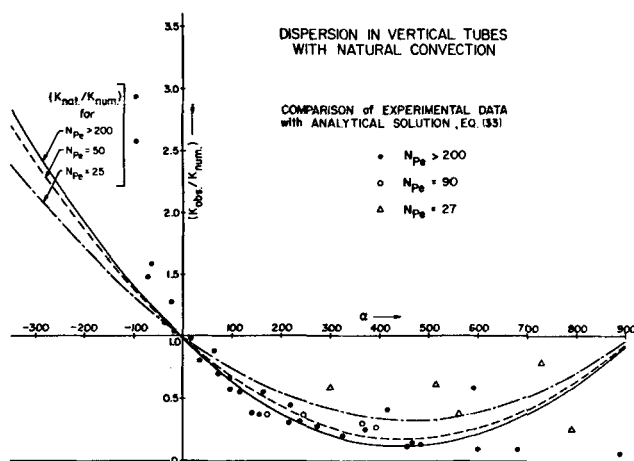Fig. 12. Comparison of experimental data with the analytical solution developed in the present work. The lines for various N_{Pe} represent the ratio of the dispersion coefficient predicted by Equation (33) to that determined numerically by neglecting natural convection. As expected, agreement is better at smaller values of α.
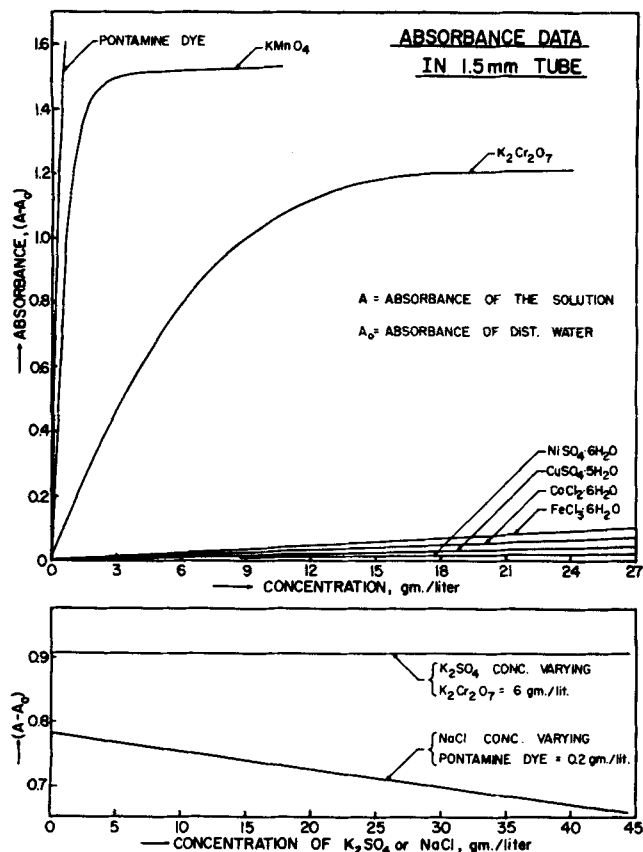
Fig. 13a (top) and 13b (bottom).

pure molecular diffusion, and one would then expect the solutions with and without natural convection to be identical. Also, one would expect this condition to be approached as N_{Pe} approaches zero. At our lowest velocities, however, $(N_{Pe} = 9$ and $\tau = 6)$ we are not yet in the region where we would expect the natural convection data to be negligible. Note, however, in Table 2 that K_{obs}/K_{num} changes rapidly from 0.255 to 0.66 in runs 9 and 11 as N_{Pe} changes from 27 to 9 and thus the effect of natural convection has diminished markedly as N_{Pe} is lowered in this range.

In Figures 9 to 11, plots of $L/\sqrt{\tau}$ vs. N_{Gr}/N_{Re} are made for various values of τ. Note that the points approach the numerical solution asymptotically as $N_{Gr}/N_{Re} \longrightarrow 0$ as expected. In Figure 9, all the data lie on a single curve for the N_{Pe} range of 720 to 14,000 at $\tau = 0.0825$, whereas in Figures 10 and 11 the effect of decreasing N_{Pe} at a given value of τ is marked. One may also note from these plots the effect of increasing τ at a given value of N_{Pe}. For example, at $N_{Pe} = 1680$, the values of $L/\sqrt{\tau}$ increase as τ increases from 0.0825 to 0.93. However, at small Peclet values, for example, at $N_{Pe} = 27$, the opposite effect is observed when τ increases from 6.2 to 37. At intermediate values of N_{Pe}, the effect of increasing τ is irregular.

ACKNOWLEDGMENT

NOTATION

a = radius of the tube
C^+ = point concentration
C_o^+ = initial concentration of the solute
C = dimensionless point concentration, C^+/C_o^+
C_m = dimensionless average concentration across the tube section
ΔC_m = dimensionless concentration change equal -1 for 100 to 0% change

d = diameter of the tube = $2a$
D = molecular diffusion coefficient
g = gravitational component perpendicular to earth's surface
k = dispersion coefficient of Taylor = $a^2 U^2/48D$
K = dispersion coefficient of Aris = $D + k$
K_{nat} = dispersion coefficient as defined by Equation (33)
L = dimensionless mixing zone length = $DL'/(2a^2 U_m)$
L' = mixing length = Δx between $C_m = 0.1$ to 0.9
r = radial distance
N_{Fr} = Froude number = $U_m^2/(dg)$; the same groups defined by using the radius a instead of the diameter d are denoted with primes.
N_{Pe} = Peclet number = dU_m/D
N_{Sc} = Schmidt number = ν/D
N_{Gr} = Grashof number for mass transfer
$\qquad = d^3 g \beta (C^+ - C_o^+)/\nu^2 = d^3 g (\Delta\rho/\rho)/\nu^2$
N_{Re} = Reynolds number = dU_m/ν
t = time of flow
U_m = bulk mean velocity = U
u, v, w = components of the velocity vector in x, r, θ directions, respectively
u_s = steady state velocity in the axial direction
x = axial distance along the tube
$x_1 = x - U_m t$
X = dimensionless axial distance = $Dx/2a^2 U_m$
y = dimensionless radial distance = r/a

Greek Letters

$$\alpha = \frac{g \beta a^4 \dfrac{\partial C_m}{\partial x_1}}{\nu D}$$

$$\beta = -\frac{1}{\rho_o} \frac{d\rho}{dC^+}\bigg|_{C^+ = C_o^+}$$

Δ = incremental value of any variable
ν = kinematic viscosity of the solution
ρ = density of the solution
μ = viscosity of the solution
τ = dimensionless time = tD/a^2
$\Delta\rho/\rho = 2(\rho_1 - \rho_2)/(\rho_1 + \rho_2)$

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