## Turbulent and Laminar Mass Transfer in a Tubular Membrane

NICHOLAS C. KAFES

Department of Chemical Engineering Manhattan College, Bronx, New York 10471

and CURTIS W. CLUMP

Department of Chemical Engineering Lehigh University, Bethlehem, Pennsylvania 18015

An experimental method has been developed which offers promise for mass transfer studies in high  $N_{Sc}$  systems. A tubular dialysis membrane was suspended in a column and the effect of flow conditions on the tube side mass transfer coefficient determined by passing water through the tube countercurrent to a circulating saturated salt solution in the column annulus. Experimentation was conducted under conditions that an adaption of the Wilson technique for film coefficients evaluation was valid to isolate the tube side coefficient from measurements of the overall mass transfer coefficient. The system is characterized by a transfer area which is always smooth and well defined, a net mass flux of zero by virtue of the membrane's permeability to all species of the system, and a negligible transfer of mass resulting in a minimal change in fluid properties.

Suitability of this technique is demonstrated with data covering a range of  $N_{Re}$  from 150 to 45,000 for a system  $N_{Sc}=800$ . The laminar regime results fill in a missing area between the streamline data collected in wetted wall column and pipe dissolution systems. Results in the turbulent regime are within creditable agreement of commonly accepted transport models and expectedly fall below coefficients data obtained in soluble pipe studies. The observed wall roughness and inherent unidirectional mass transfer mechanism of the pipe dissolution apparatus act

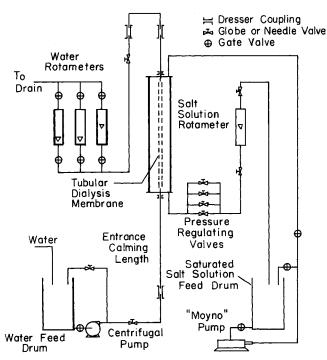


Fig. 1. Flow diagram of experimental system.

as increasingly disturbing influences the higher the  $N_{Sc}$ ; however, with the subject membrane system, a smooth interface and a counterdiffusion mechanism prevail.

#### **EXPERIMENTAL PROCEDURE**

The apparatus consisted of the column/tube contact section and ancillary circulation and measurement components as shown in Figure 1. The column was constructed with two parallel sides of plexiglass and the halves of a 12.7-cm diam. pipe split along its length. The cellulose dialysis tubing employed had a nominal diam. of 2.86 cm and an average pore diam. of 48 A°, and is available from the A. H. Thomas Co. of Philadelphia, Pa. (Cat. No. 4465-A2). Tubing lengths of the order of 125 cm were used; the transfer area provided gave an exit water concentration of 75 to 15,000 ppm (conductivity cell measurement) over the flow range considered in this study. Before installation, the membrane was steeped in a salt solution for at least 48 hours. The extremities of the membrane were slipped over steel tubes (I.D. = 2.76 cm) and the junctures sealed. These tubes acted as calming lengths to establish and maintain normal velocity gradients in the water stream; the entrance/exit tube lengths were 50/25 times the membrane diameter, respectively. Dresser coupling assemblies fastened to the end plates of the column served to align and secure the tubes.

Once startup was executed, two hours were permitted to attain steady state conditions. Each run lasted 45 minutes; rotameter readings were taken and inlet/exit water stream samples collected; a half hour was allowed between runs. The salt solution was circulated through the annulus at  $N_{Re} = 830$  (less than 2% variation maintained) by means of a Moyno screw pump. City water was pumped through the tubular membrane and discharged to the sewer. The pressures of the streams in the contact section were balanced to maintain a constant membrane geometry by manipulating the downstream valves of the system. An average tube diameter of 2.88 cm was photographically determined with a 2% difference noted between the top and bottom of the column. The system temperature averaged 19°C, being dictated by the available city water; during the course of a set of runs the water stream temperature held constant to within 1.5°C. The circulating solution had a tendency to increase in temperature. Direct addition of chipped ice to the salt solution feed drum was used to maintain the difference between the two feeds below 0.5°C; salt was also added to assure a saturation concentration (0.317 g/cm<sup>3</sup>) of the annular stream at all times.

### RESULTS

As described, the experiment was conducted over a range of tube-side water flows while maintaining a constant salt solution flow rate in the annulus. This approach, suggested by Wilson for the analogous heat transfer operation, can be adapted to the present mass transfer situation for determining the tube side coefficient from measurements of the overall mass transfer coefficient. The general relation

$$n = \frac{Q_w \left(C_{w2} - C_{w1}\right)}{A} = K \left(C_s - C_w\right)$$

was employed to calculate the overall coefficient. Particularization of the overall resistance in terms of the resistance to mass transfer residing in the boundary layers of fluid adjacent to the surface and the resistance of the membrane to diffusion

$$\frac{1}{K} = \frac{1}{k_s} + \frac{1}{k_m} + \frac{1}{k_w}$$

follows from the linearity between the mass transfer rate and the concentration potential. Under the constraints of the experiment,  $k_s$  and  $k_m$  are constant while  $k_w$  is a function of velocity only.

$$\frac{1}{K} = a + b \, \frac{1}{V^c}$$

The turbulent regime data were employed to determine the constants of the above relation. Evaluation by the method of least squares requires that the equation be reduced to a form linear in these constants. The method of differential corrections (Nielson, 1956) was utilized to accomplish this. Initial values of a, b, and c are assumed and the correction  $\alpha$ ,  $\beta$ , and  $\gamma$ , for each of the constants determined. Application of Taylor's theorem results in a series of residual equations (one for each data point), linear in these corrections, which can then be subject to a least squares analysis. Successive trials were carried out using the results of each previous trial until values for the constants converged. The final values were

$$a = 80.94$$
  $b = 13450$   $c = 0.9005$ .

The resistance to mass transfer of the tube side fluid  $(1/k_w)$ , obtained by subtracting the intercept value of a from the overall resistance, can be expressed in terms of the generalized fluid flow/mass transfer parameters. These results are plotted in Figure 2 and are compared with the

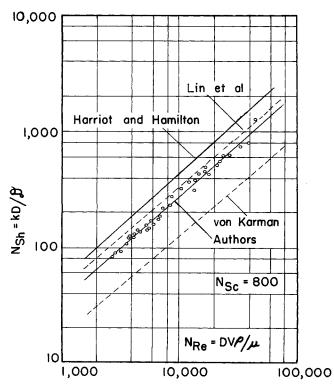


Fig. 2. Turbulent mass transfer.

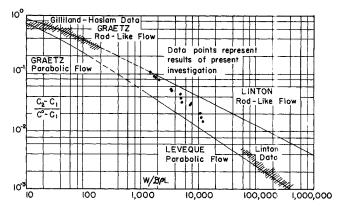


Fig. 3. Mass diffusion in laminar flow

turbulent eddy diffusivity model of Lin et al. (1953) and the film model of von Karman as adapted by Sherwood (1959). The data points are represented by the equation

$$N_{Sh} = 0.069 \ N_{Re}^{0.90}$$

which falls about 15% below the corresponding Lin relation at  $N_{Sc}=800$ . The Harriot and Hamilton (1965) correlation of their pipe dissolution data is also shown and found to be about 35% above the Lin model.

The resistance to transfer of the tube side fluid in streamline motion is similarly obtained by subtracting the intercept value from the overall resistance data collected in the laminar regime. The results are compared in Figure 3 with the mass transfer analogs of the classical Graetz solutions and the corresponding Leveque asymptotic approximations for the two cases of parabolic flow and plug flow in tubes. The Leveque solutions are applicable for the present mass transfer problem, since diffusion takes place a short distance into the fluid stream while in the contact zone. The shaded areas in the figure depict the laminar data of the Linton and Sherwood (1950) pipe dissolution study and the wetted wall column data of Gilliand (vaporization) and Haslam (absorption), as taken from Sherwood and Pigford (1952).

This investigation demonstrates the potential utility of the dialysis membrane system for studying the turbulent transport mechanism in the region of the wall. In particular, the limiting relation for the eddy diffusivity at the wall can be determined from a study of high  $N_{Sc}$  systems in an apparatus in which the smooth, permeable interface obviates the disadvantages of the more commonly employed pipe dissolution experiment.

## NOTATION

A = membrane transfer area, cm<sup>2</sup>

a, b, c = constants

C = concentration, g/cm³
 D = membrane diameter, cm
 D = molecular diffusivity, cm²/min

K = overall mass transfer coefficient, g/cm² min g/cm³
 E individual mass transfer coefficient, g/cm² min g/

 $m cm^{8}$ 

L = membrane length, cm  $N_{Re}$  = Reynolds number,  $DV\rho/\mu$   $N_{Sc}$  = Schmidt number,  $\mu/\rho D$   $N_{Sh}$  = Sherwood number, kD/D n = mass flux, g/min cm<sup>2</sup> Q = volumetric flow, cm<sup>3</sup>/min V = average velocity, cm/min

W = mass flow, g/min  $\alpha$ ,  $\beta$ ,  $\gamma$  = corrections to constants a, b, c = viscosity, g/min cm = density, g/cm<sup>3</sup>

#### **Subscripts and Superscripts**

= membrane

= saturated salt solution 2. = tube side water stream 11) 1. 2 = terminal conditions

= wall concentration, tube side

#### LITERATURE CITED

Harriot, P., and R. M. Hamilton, "Solid-Liquid Mass Transfer in Turbulent Pipe Flow," Chem. Eng. Sci., 20, 1073 (1965). Lin, C. S., R. W. Moulton, and G. L. Putnam, "Mass Transfer Between Solid Wall and Fluid Streams," Ind. Eng. Chem., **45,** 636 (1953).

Linton, W. H., and T. K. Sherwood, "Mass Transfer from Solid Shapes to Water in Streamline and Turbulent Flow," Chem. Eng. Progr., 46, 258 (1950).

Nielson, K. L., Methods in Numerical Analysis, pp. 309-313,

Macmillan, New York (1956).
Sherwood, T. K., "Mass, Heat and Momentum Transfer Between Phases," Chem. Eng. Progr. Symp. Ser., No. 25, 55, 71 (1959).

-., and R. L. Pigford, Absorption and Extraction, pp. 81-82, McGraw-Hill, New York (1952).

Manuscript received October 12, 1972; revision received and accepted

# Experimental Studies of Natural Convection Effects On Dispersion in Packed Beds

WILLIAM N. GILL

**Faculty of Engineering and Applied Sciences** State University of New York at Buffalo, New York

> and MARTIN POSNER

**Chemical Engineering Department** Clarkson College of Technology, Potsdam, New York

In two previous articles (Reejsinghani et al., 1968; Posner and Gill, 1973) it was shown that buoyancy effects in vertical open tubes can influence the extent of dispersion markedly. The purpose of this note is to study natural convection effects in packed beds which are used in a variety of industrial applications. The data cover most of the range of concentration differences (up to 26,000 ppm salt) of interest in washing the ice crystals produced in desalting by the freezing process. However, the  $Re_p$  range studied was limited to about twice the critical value  $(Re_{p \text{ critical}} \approx 3)$  for fluidization to occur.

#### PACKED BED APPARATUS

As an extension of the experimental work done with open tubes the experimental apparatus shown in Figure 1a and 1b was set up to investigate miscible displacement in packed beds. The apparatus is designed so that a close approximation to a step change in concentration and density can be obtained at the inlet of the bed after it has been filled with the solution that is to be displaced. The area mean concentration at the outlet of the bed is monitored continuously. Both upflow and downflow experiments have been carried out to determine the effect of viscosity.

The bed is constructed of a 3.81-cm glass pipe in which 3M Company, Superbrite, Class B, narrow size distribution (70% and 30%, respectively, are retained on U.S. sieves number 40 and 60) glass beads with a density of 2.5 g/cc have been packed in the pipe and the spheres are contained by wire screens at both ends; the bed length L is 15.24 cm and the average particle diameter  $d_p$  is 0.047 cm. Thus the ratio of bed length to particle diameter is sufficient for the dispersion model to apply. The entrance section to the bed is a length of glass pipe which has been filled with a nylon mesh that flattens the velocity profile of the fluid entering the bed. Above the bed is a glass pipe spacer which is opaque except for a small region at the bottom which serves as the window for the optical system which measures the output concentration. The region above the bed also is filled with nylon mesh except for a short distance of about 2 mm in which the optical measurements are

The optical system consists of a light source, a G.E. No. 93 bulb powered through a step-down transformer, and a light intensity measuring system which is a No. 631A photomultiplier tube in a housing that includes two parallel slits separated by a shutter; behind the second slit is a space for colored filters and behind this is the photomultiplier tube. The photomultiplier tube is connected to an Aminco microphotometer which translates the output from the tube into light transmission data. The output from the microphotometer is monitored continuously on a 0-50 millivolt recorder.

The experimental fluids used are K2SO4, which forms a colorless solution, and K2Cr2O7 which forms a colored solution. Since the dichromate solution shows maximum light absorption in the region of 4600 A, filters are placed in the photomultiplier tube housing so that only light in this region of the spectrum is mea-

## ANALYSIS OF THE PACKED BED DATA

We have employed the method of Hopkins, Sheppard, and Eisenklam (1969) to determine the dispersion coefficient from the experimental data. Once one knows the concentration vs. time data at the outlet  $c_2(t)$  and at the inlet  $c_1(t)$  of the system then one forms the LaPlace transform of the residence time distribution which is

$$F(s) = \frac{\int_0^\infty c_{m2}(t) e^{-st} dt}{\int_0^\infty c_{m1}(t) e^{-st} dt}$$
(1)

For axial dispersed plug flow, the dispersion equation is