ACKNOWLEDGMENT

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Dialysis Study of Diffusion in a Flowing Suspension

Comparison of solute concentrations at the outlet of a model dialyzer with values calculated from the appropriate convective diffusion equation yielded effective diffusion coefficients for mass transfer of sodium chloride in sheared suspensions of 37 to 74 μ spheres of a copolymer of styrene-divinylbenzene. Significant mass transfer augmentation was observed over that attributable to molecular diffusion, and the effective diffusivity increased with increasing concentration of suspension but remained relatively constant with respect to shear rate over the experimental range.

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SCOPE

The augmentation of heat or mass transfer in a sheared suspension has been well established (Singh, 1968; Collingham, 1968; Keller, 1971; Turitto et al., 1972). The secondary fluid motion associated with the rotation of the suspension particles may result in a greater transfer of solute transverse to the direction of bulk flow than would be produced by simple diffusion. These considerations are essential to an optimal design of artificial kidneys.

The present work studied the mass transfer of sodium chloride through a neutrally buoyant suspension of spheres of a copolymer of styrene-divinyl-benzene in a model dialyzer. The fluid was treated as being homogeneous, and the situation was formulated as a convective diffusion problem involving an effective diffusion coefficient. In a more elaborate formulation this effective diffusivity would vary with position as a function of local shear rate and concentration of suspension. In this work the effective diffusivity was regarded as a constant dependent upon shear rate at the wall and average particle concentration. This relationship was evaluated by comparing the value of the concentration of solute measured at the dialyzer outlet with the value predicted from the mathematical formulation involving the effective diffusivity.

CONCLUSIONS AND SIGNIFICANCE

The dialyzer studies showed that the presence of 50 μ particles served to augment the diffusion of a solute in laminar flow. The effective diffusivity increased with increasing particle concentration but remained relatively constant with increasing shear rate at the wall. These results are in the direction indicated by a model in which the particles do not interact (Leal, 1973) but differ in the nature of the dependence. From Figure 5 the effective diffusivity is seen to be almost constant with shear rate as opposed to the result expected on the basis of an independent particle model. The value of $D/D_o - 1$ is seen from Figure 6 to be negligible at particle concentrations of 5%; it increases rapidly with increasing particle concentration over the concentration range of 5 to 10% and approaches a limit at a concentration of about 30%. At the particle concentrations studied, particle interactions undoubtedly have a strong effect on mass transfer augmentation, and particle migration as well as particle rotation is expected to be important. These movements seemed to depend mainly on the particle concentration and were not significantly affected by the shear rate within the range of shear rates at the wall employed in the experiment.

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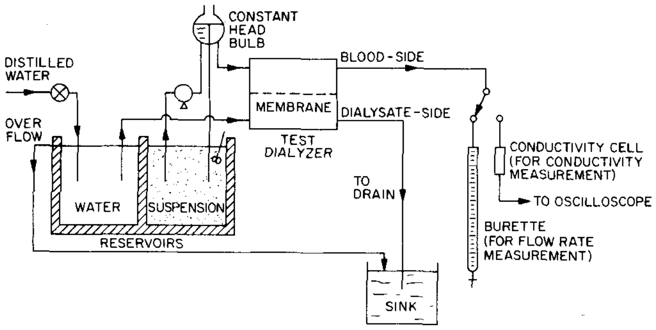

Fig. 1. Schematic flow diagram of a test dialyzer.

Since removal of toxic materials in hemodialysis is often limited by transport through the blood (Leonard and Bluemle, 1958), an enlightened choice of design parameters requires a detailed understanding of the transport phenomena within the blood. Blood itself is too complex to permit isolation of the individual features of interest, so a study was undertaken of the diffusion of sodium chloride through a neutrally buoyant suspension of particles in a simplified dialyzer. The objective was to determine the effect of particles in shear flow on the transport of solute in the presence of shear flow.

During the last several years much effort has been spent on the theoretical description of the bulk rheological properties of flowing suspensions (Cox and Brenner, 1968, 1971; Otterman and Lee, 1970), and extensive experimental investigations have been carried out. In particular, Karnis et al. (1966*b*) and Goldsmith and Mason (1969) studied the velocity profile in suspensions of rigid and deformable particles, and Karnis et al. (1966*a*) and Maude and Yearn (1967) considered the radial migration of flowing particles. Comprehensive review articles on flowing suspensions have been prepared by Goldsmith and Mason (1967) and Cox and Mason (1971).

In contrast to the rheological problem, relatively little attention has been devoted to the other bulk transport processes of diffusion and heat transfer in suspensions. Diffusion of oxygen and helium through a suspension of polystyrene spheres in 7.4 wt. % sodium chloride solution was studied by Collingham (1968), and the corresponding measurement of thermal conductivity was carried out by Singh (1968). Both observed augmentation effects attributable to rotation of the suspended particles. Keller (1971), in reporting experiments on the effect of fluid shear on mass transport in flowing blood, also concluded that particle rotation could cause mass transfer augmentation. An experimental study on diffusion of urea in flowing blood was conducted by Colton et al. (1971*a*), and the augmentation of platelet diffusion by red blood cell motion was studied by Turitto et al. (1972). Navari et al. (1971) introduced a predictive theory for diffusion in polymer and protein solutions, and recently Leal (1973) performed a theoretical investigation on the effective conductivity of a dilute suspension in the limit of low particle Peclet number.

Hemodialysis has been a research subject of much interest in recent years, and many efforts were directed to establish appropriate analytical models to interpret data for diffusive and convective transport. The fundamental description of convective diffusion in laminar flow was presented by Graetz in 1885, and the techniques were applied to dialysis by Grimsrud and Babb (1966) and Colton et al. (1971*b*). Formulations for more specific applications to hemodialysis have been presented by Popovich (1970), Gaylor et al. (1970), and Ramirez et al. (1971). Informative reviews of the development and performance of hemodialyzers were given by Colton (1966) and Corcoran (1968). The determination of membrane permeabilities and solute diffusivities in a dialyzer were studied carefully by Babb et al. (1968) and Smith et al. (1968).

EXPERIMENTAL

Dialysis experiments were conducted with a flat plate dialyzer. It was a one-sided version of the basic design described by Grimsrud and Babb (1966), which permitted observation of the flow through the Lucite of the blood channel. Highly porous nickel foam introduced by Babb and Grimsrud (1964) was used as the mebrane support. Area for mass transfer was 5.75 in. in width and 2 in. in length. The height of the blood-side channel could be varied by changing the spacers between two Lucite blocks and was maintained as 0.032 in. in most cases. The steady shear rate was in the range of 7 to 25 s $^{-1}$. Cuprophane PT-150 was the membrane, and the pressures holding the membrane flat were sufficiently small that ultrafiltration was negligible (Popovich et al., 1971).

The overall flow system is shown schematically in Figure 1. The basic measurements made were of flow rates of suspension and concentrations of sodium chloride at the dialyzer outlet. Since air dissolved in tap water tended to come out of solution in the nickel foam and settle against the membrane, distilled water was used as dialysate. During each run the flow rate of dialysate was held constant at about 500 ml/min. According to Grimsrud and Babb (1966), this flow is well inside the turbulent flow regime for a packed bed. Runs made under identical conditions at 500 and 900 ml/min revealed no measurable dependency of the mass transfer rate on dialysate flow rate. The sodium chloride concentration in the dialysate side of the membrane could therefore be assumed to be essentially zero. The blood-side flow rate was varied over a wide range, from as low as 0.5 to as high as 50 ml/min. During a dialysis run, samples were withdrawn constantly from the exit of the blood-side channel.

All experiments were conducted at ambient temperature, which never varied more than 2 deg. from 26°C. Temperature was measured with thermometers at the blood-side exit and at the dialysate reservoir, and the entire system was assumed to be in thermal equilibrium. During a single run, temperature remained constant to within ± 1°C.

The concentration of sodium chloride in the blood-side fluid leaving the dialyzer was determined by conductivity measurements after the particles were filtered from the sample. The measurements were taken in a temperature regulated (within ± 0.1°C) conductivity cell by using a low distortion oscillator, a Winston bridge, and a dual beam oscilloscope. Accuracy of the conductivity measurements was ±0.01 mmho. Since calibrations indicated that the concentration of sodium chloride was linearly proportional to conductivity over the range of concentrations of interest, the dimensionless outlet concentration needed in the analysis was obtained as the ratio of conductivity of a sample to its initial conductivity.

Lightly cross-linked styrene-divinylbenzene-copolymer beads, manufactured by Dow Chemical Company as Dowex 50W8, were selected to form the suspension. Particle density was found to be 1.05 g/ml, which at room temperature was matched by the density of a solution of 7.5 wt. % sodium chloride in water. The particles were spherical with sizes widely distributed in the range of 37 to 74 μ. The particles tended to settle upon standing for several hours. Gentle agitation would, however, bring them back to the dispersed state. Neither particle aggregation nor significant solute binding by particles was observed.

Particle concentrations in the suspensions were determined with a Beckman model B spectrophotometer. The blank for comparison consisted of dimethylaminobenzaldehyde in ethanol and sulfuric acid. Calibration with known particle concentrations provided a relationship between particle concentration and light absorbance which was useful for concentrations ranging from 4 to 40 vol. %.

DATA ANALYSIS

The suspension containing an initial concentration c_0 of sodium chloride flowed through the dialyzer in a flat duct of width w and height a. The flow was laminar, and the velocity profile was assumed to be parabolic

(Newtonian fluid), although the experimental results of Karnis et al. (1966*a*) indicated that the profile may be blunted for particle concentrations greater than 11%. The solution flowed over a supported semipermeable membrane, characterized by a constant solute permeability P. Distilled water flowed through the dialysate channel under the membrane. Flow of the dialysate was turbulent, so the solute concentration was assumed to be zero on the dialysate side of the membrane. If one assumes steady state conditions; no ultrafiltration, which combined with the osmotic effect was less than 1% of the gross flow; and no axial diffusion, which is clearly negligible in this situation (Schneider, 1957), the convective-diffusion equation describing the concentration of the solute in the dialyzer may be written as

$$\frac{3}{2}\bar{u}\left[1-\left(\frac{2y}{a}-1\right)^2\right]\frac{\partial c}{\partial x} = D\frac{\partial^2 c}{\partial y^2} \quad (1)$$

with boundary conditions

$$x = 0: \quad c = c_o \quad (2)$$

$$y = 0: \quad \frac{\partial c}{\partial y} = 0 \quad (3)$$

$$y = a: \quad -D\frac{\partial c}{\partial y} = P'c \quad (4)$$

where $P' = P/(1-\phi)$, and ϕ = volume fraction of particles. The quantity $\bar{u}$ is the average velocity and is related to the volumetric flow rate Q by

$$\bar{u} = \frac{Q}{wa} \quad (5)$$

D is the effective diffusivity. Under these circumstances it depends on the flow rate Q.

The solution to this problem is available (Lindgren and Corcoran, 1973). The dimensionless mixing cup concentration $<\psi>$ ($= <c>/c_o$) at the dialyzer outlet $x = x_e$ is given in terms of dimensioned quantities by

$$<\psi> = \sum_{n=1}^{\infty} B_n\, e^{-\frac{\lambda_n D W X_e}{6Qa}} \quad (6)$$

where the coefficients B_n and the eigenvalues λ_n depend upon the wall Sherwood number N_s defined as

$$N_s = P'a/D \quad (7)$$

In a dialyzer run with no suspended particles, and for laminar flow, D is simply the molecular diffusivity of sodium chloride and is independent of Q. Measured values of $<\psi>$ for various Q from such a run were compared to values calculated from Equation (6), and D was varied until the best fit was obtained according to a least-squares criterion. This procedure provided a reference value D_o to which effective diffusivities in suspensions were compared. In runs with suspended particles, Equation (6) must be applied separately at each flow rate because D is a function of Q. By solving the equation for D for each set of experimental values for $<\psi>$ and Q, the effective diffusivity was obtained as a function of flow rate for a dialyzer run with a specific concentration of suspended particles. The flow rate is related to the shear rate at the wall by the equation

$$\gamma = \frac{6Q}{wa^2} \quad (8)$$

and the experimental results are presented in the later sections as graphs of the diffusion augmentation D/D_o vs. γ.

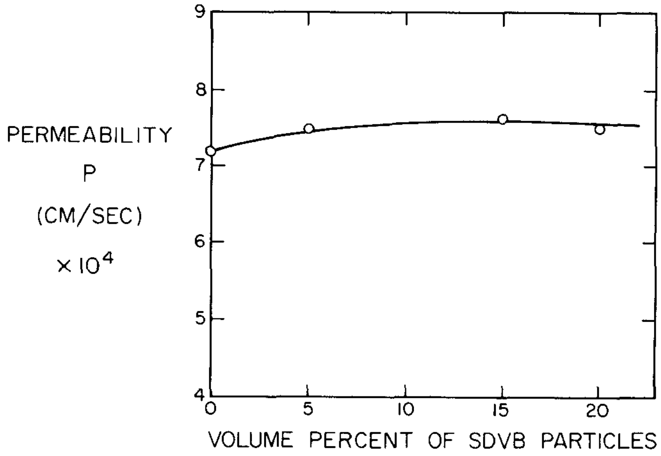

Fig. 2. Membrane permeability vs. particle density.

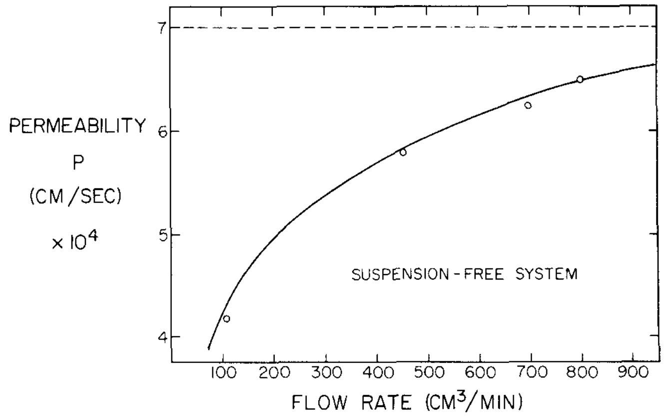

Fig. 3. Apparent membrane permeability vs. flow rate.

RESULTS

Membrane Permeability

Cuprophane PT-150, which has been widely adopted in hemodialysis, was used for the membranes in the dialyzers. The membrane permeability P enters the analysis of data through Equation (7), where $P' = P(1-\phi)$. Since its value may vary considerably from one batch of Cuprophane to another, P was determined in experiments by using a standard NBS cell (Babb et al., 1968), and all dialyzer membranes were taken from the same sheet. Equal volumes of fluid were circulated on either side of the membrane in the NBS cell. One was distilled water initially, and the other was a solution of sodium chloride. Periodically, equal samples were withdrawn from each side during the experiment, and concentrations of both samples were determined by conductivity measurements. Concentrations were recorded as functions of time and were compared with values generated from an analytical model which described mass transfer in the finite sampling system. The analytical values were functions of the permeability P, and the value of P was determined by minimizing the differences between experimental data and model predictions by means of a least-squares treatment.

For a particle free system, the membrane permeability was found to range from 7.0×10^{-4} to 7.2×10^{-4} cm/s and was independent of the solute concentration. A series of permeability tests was carried out for various concentrations of particles ranging from 0 to 20 vol. %. A very slight increase in permeability with increasing particle density was observed as shown in Figure 2. This result was most probably caused by the existence of a very thin boundary layer in the proximity of the membrane in spite of the rapid flow used to justify the assumption of perfect mixing within the cell. The motion of the particles may

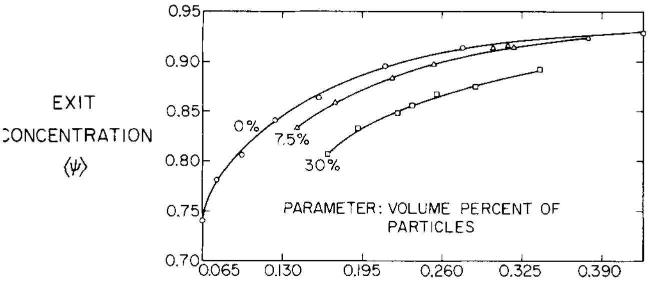

Fig. 4. Exit concentration vs. flow rate for different particle densities.

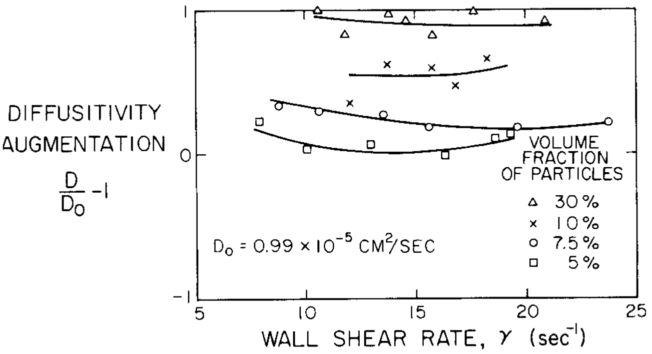

Fig. 5. Diffusivity augmentation vs. shear rate as a function of particle density.

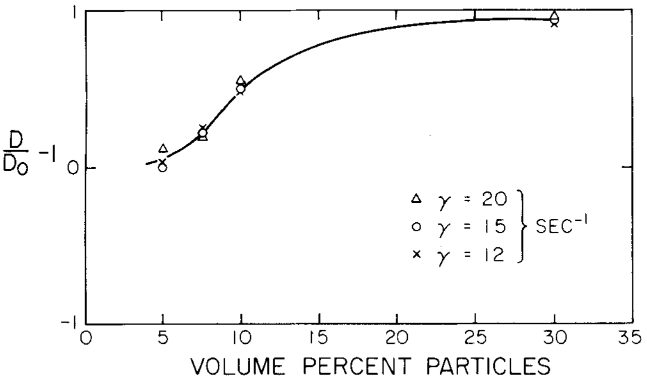

Fig. 6. Diffusivity augmentation vs. particle density for different shear rates.

have facilitated mixing and thus may have reduced the effect of the boundary layer. Assignment of these effects to a boundary layer adjacent to the membrane becomes more convincing after observation of permeability measurements taken at lower flow rates. Figure 3 shows a dramatic decrease in apparent membrane permeability with a decrease in the flow rate, and this effect is clearly the result of imperfect mixing in the cell.

Effective Diffusivity

Results of experiments performed on the dialyzer with the smaller channel height are presented in Figures 4 to 6. The measured exit concentrations are plotted against flow rate in Figure 4, and it is apparent from the positions of the curves that mass transfer is enhanced by increasing the density of suspended particles. The effect of shear rate on mass transfer, however, cannot be determined from direct observation of these results. Analysis of the data for the run without particles yielded a reference diffusivity D_0 of 0.99×10^{-5} cm²/s with respect to which all effective diffusivities were normalized. Results of the analysis are presented in Figure 5 as plots of diffusivity augmentation D/D_0 vs. wall-shear rate and show that the effective diffusivity essentially remains constant as the shear rate varies. The curves of Figure 5 are replotted in Figure 6 to show the variation of effective diffusivity with particle concentration.

In addition to particle concentration, the particle size is an important parameter in the study of augmentation of mass transfer. Its effect was not seriously studied, but for a suspension with 10 vol. % of particles, preliminary results showed that the effective diffusivity decreased by a factor of 2 when the average particle size was reduced from 50 to 25 μ. More experimental results are needed to characterize quantitatively the effect of particle size on effective solute diffusivity over the range of shear rates of interest.

SUMMARY

Experiments were performed by using a flat plate dialyzer with a semipermeable membrane as one wall. A known concentration of sodium chloride was passed through the device, and the steady state, mixing cup concentration at the outlet was measured for various flow rates. The wall-shear rate is related very simply to the flow rate, and an effective diffusivity was obtained as a function of wall-shear rate by comparing the measured concentration with the theoretical model. Dialyzer dimensions, diffusivity of sodium chloride in water, and membrane permeability had to be known. The permeability of the membrane was determined in separate experiments by running solutions of sodium chloride through standard NBS test cells. The diffusivity was obtained by comparing measured and theoretical outlet concentrations for a dialyzer run without suspended particles. At a temperature of 26°C, values for the permeability of the membrane and for the diffusivity of sodium chloride were evaluated as 7.0×10^{-4} and 0.99×10^{-5} cm²/s, respectively. They agreed well with values in the literature (Babb et al., 1968).

Experiments with suspended spherical beads of styrene-divinylbenzene copolymer with sizes in the range of 37 to 74 μ were carried out for various concentrations of particles ranging from 0 to 30%. At the upper limit of particle concentration, the effective diffusivity exceeded the molecular diffusivity by a factor of about 2 for wall-shear rates in the range of 10 to 20 s^{-1} (Figure 5). No appreciable change in the effective diffusivity with shear rate was observed for the particle concentrations studied.

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NOTATION

a = channel height in dialyzer
B_n = coefficient of eigenvalue expansion
c = solute concentration
c_o = initial solute concentration
$<c>$ = mixing cup solute concentration
D = effective diffusivity
D_o = reference diffusivity (diffusivity of sodium chloride in water)
N_s = wall Sherwood number
P = membrane permeability
$P' = P(1 - \phi)$ = a permeability corrected for volume fraction of suspended particles
Q = volumetric flow rate
$\overline{u}$ = average velocity
w = dialyzer width
x = distance in direction of flow

y = distance transverse to flow
γ = wall-shear rate
λ_n = eigenvalue
$<\psi>$ = dimensionless mixing cup solute concentration
ϕ = volume fraction of particles

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The Effect of Surfactant on the Terminal and Interfacial Velocities of a Bubble or Drop

Stream functions are derived for a spherical droplet in creeping flow with an arbitrary surface tension gradient at its interface. The stream functions are used to show theoretically that the terminal velocity is reduced and that the interfacial velocity is retarded, especially near the rear of the droplet, when a trace of surfactant is present in the exterior phase and when surface aging is controlled by diffusion.

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SCOPE

Experimental investigations of droplet motion in liquid containing a surface active impurity have shown that, compared to motion in pure liquid, both the terminal velocity and the interfacial velocity of a droplet are reduced, the latter particularly near the rear of the droplet.

Here a theoretical investigation of droplet motion in systems contaminated by surfactant is presented. The ob-

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