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CONVECTIVE DIFFUSION FROM A NON-UNIFORMLY DISTRIBUTED SOURCE IN NON-NEWTONIAN FLUIDS: A THEORETICAL INVESTIGATION AND EXPERIMENTAL CONFIRMATION

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This work presents a complete theoretical study of the process of convective diffusion from a non-uniformly distributed source in a non-Newtonian fluid flowing through a straight tube. The influence of the governing parameters on the dispersion process is studied and the results are examined in terms of the interaction of rheology, fluid mechanics and diffusion. Experimental studies have been conducted to obtain the diffusivity values of NaOH in aqueous polymer solutions and provide the first test of the potential of this technique for diffusivity measurement in difficult systems.

Measurement of molecular diffusion coefficients of solutes in non-Newtonian media is crucially important from the point of view of being able to understand the pragmatically important convective diffusion processes, which occur in these media. The state-of-the-art in this area was recently reviewed by Astarita and Mashelkar (1977). This review clearly emphasizes the need for more experimental data on molecular diffusivities in non-Newtonian media but more importantly, it showed that reliable *flow* techniques are essential for the diffusivity determination in highly viscous non-Newtonian media since in the conventional static techniques a very long time is required to attain an appreciable concentration gradient. In the present work, we analyse theoretically the possibility of using the technique of convective diffusion from a non-uniformly distributed source and set up experiments to simulate the model conditions. We then provide experimental results to show how such a technique can be used for diffusivity measurements.

BACKGROUND

It appears that the experimental techniques which have been used so far for diffusivity determination by using flowing fluids involve the use of essentially

two-phase systems. Thus, Clough *et al.* (1962) used solid dissolution from soluble walls of a pipe for the measurement of molecular diffusivity of β -naphthol and benzoic acid in flowing non-Newtonian polymer solutions. Astarita (1966) and Desai (1975) used the technique of solid dissolution from a falling film. Mashelkar and Soylu (1974) used the technique of gas absorption in films of dilute polymer solutions flowing over a wetted sphere for diffusivity measurement of carbon dioxide in a variety of polymer solutions. Such measurements essentially rely upon the solution of the pertinent convective diffusion problem (e.g. Graetz-Leveque solution in the case of a dissolving wall in a pipe) and the back calculation of the diffusivity from the observed exit concentrations. However, for such interplane transport to occur, it is crucially important that the solute be available either in the solid or in the gaseous form. Obviously, when the solute is a miscible liquid (e.g. a monomer in a polymer solution), such simple techniques cannot be used. The present technique essentially involves the preparation of a solution containing the soluble solute, its non-uniform injection in the bulk of the fluid and the measurement of concentration variation at fixed axial positions.

The theoretical problem has received some attention in the literature. Harlacher and Engel (1970) solved the convective diffusion problem from a non-uniformly distributed source. However, their solution suffered from certain limitations. Their series solution was truncated after three terms, which led to negative

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concentrations for radial distances greater than about one-third of the tube. Secondly, their solution is valid only for integer values of the reciprocal power-law index (N). The final solution is incomplete with respect to the eigenvalue corresponding to the zeroth value and lastly it is inaccurate for small axial distances. For Newtonian as well as non-Newtonian liquids analytical solutions to the pulse disturbance were offered by Subramanian and Gill (1975, 1976) and by Booras and Krantz (1976). Mashelkar and Venkatsubramanian (1975) obtained a numerical solution for a Casson fluid with special attention on the problem of urea diffusivity determination in flowing blood. However, these authors did not report any experimental data for such systems.

It is thus clear that so far there is no correct and complete solution to the problem of convective diffusion from a non-uniformly distributed source in a non-Newtonian fluid, neither are there any experimental data reported. The purpose of this work is to get an accurate solution by solving the convective diffusion equation for non-Newtonian liquids and to provide experimental support for the theory developed to measure diffusivity values. The advantages of using such a technique have been already enumerated (Harlacher and Engel, 1970). Any solute may be used for measurement and the solute concentration values may simulate process conditions.

THEORY

The general equation of conservation of mass can be expressed as given by Bird *et al.* (1960):

$$\frac{\partial c}{\partial t} + (\mathbf{V} \cdot \nabla c) = D \nabla^2 c + r_A \quad (1)$$

(the explanation of the symbols used can be found in "Notations").

The physical situation involved is described in Figure 1.

We will assume the following:

- 1) Steady-state conditions prevail, i.e. $\frac{\partial c}{\partial t} = 0$.
- 2) The flow is fully developed, i.e. $V_r = V_\theta = 0$.
- 3) Isothermal conditions prevail, so that physical properties may be assumed to be constant.
- 4) No reaction occurs or no source-sink effect is present.

Under the above assumptions, Eq. (1) can be reduced to Eq. (2) if the term representing the axial

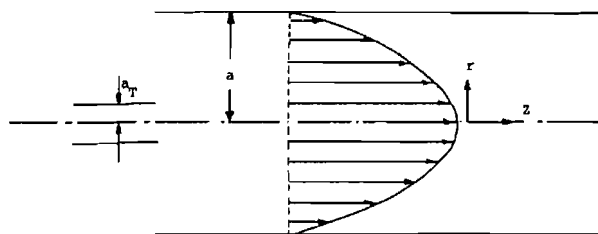


FIGURE 1 A sketch of the non-uniformly distributed source.

transport due to diffusion is considered negligible compared to the convective transport.

$$V_Z \frac{\partial c}{\partial Z} = D \left(\frac{\partial^2 c}{\partial r^2} + \frac{1}{r} \frac{\partial c}{\partial r} \right) \quad (2)$$

The convective field represented by V_Z will be different for different non-Newtonian liquids. We will choose two fluid models, which have been found to be quite useful in describing the shear flow of non-Newtonian fluids. For an Ostwald-de-Waele power-law model

$$\tau = K \dot{\gamma}^n \quad (3)$$

we have

$$V_Z = \left(\frac{3+N}{1+N} \right) V_{\text{avg}} (1 - (r/a)^{N+1}) \quad (4)$$

and for an Ellis fluid described by

$$\frac{1}{\tau} = \frac{1}{\eta_0 \dot{\gamma}} [1 + (\tau/\tau_{1/2})^{\alpha-1}] \quad (5)$$

$$V_Z = \frac{2V_{\text{avg}}}{1 + 4m(\alpha-1)} \left[1 - (r/a)^2 + \frac{2m(\alpha-1)}{\alpha+1} (1 - (r/a)^{\alpha+1}) \right] \quad (6)$$

where $m = \tau_R/\tau_{1/2}$ and τ_R represents the shear stress evaluated at the wall.

Equation (2) is to be solved with the following boundary conditions

$$\left. \begin{aligned} \frac{\partial c}{\partial r} = 0 & \quad \text{at } r = 0 \\ \frac{\partial c}{\partial r} = 0 & \quad \text{at } r = a \end{aligned} \right\} \text{—for all } Z \quad (7)$$

$$\left. \begin{aligned} c = c_f & \quad 0 \leq r \leq a_T \\ = 0 & \quad a_T < r \leq a \end{aligned} \right\} \text{—at } Z = 0$$

The first one signifies symmetry about the axis, the second an impermeable wall and the third the non-uniform but symmetric distribution at the source.

Equation (2) is non-dimensionalized by letting

$$\theta = c/c_I$$

$$\rho = r/a$$

$$\xi = ZD/V_{avg}a^2$$

Substituting these in Eq. (2), the non-dimensional equations for a power-law liquid reduce to

$$\left(\frac{3+N}{1+N}\right)(1-\rho^{N+1})\frac{\partial\theta}{\partial\xi} = \frac{\partial^2\theta}{\partial\rho^2} + \frac{1}{\rho}\frac{\partial\theta}{\partial\rho} \tag{8}$$

and for an Ellis fluid they reduce to

$$\frac{2}{\left(\frac{1+4m^{\alpha-1}}{\alpha+3}\right)} \left[(1-\rho^2) + \frac{2m^{\alpha-1}}{\alpha+1}(1-\rho^{\alpha+1}) \right] \times \frac{\partial\theta}{\partial\xi} = \frac{\partial^2\theta}{\partial\rho^2} + \frac{1}{\rho}\frac{\partial\theta}{\partial\rho} \tag{9}$$

The non-dimensional boundary conditions are

$$\left. \begin{aligned} \frac{\partial\theta}{\partial\rho} &= 0 & \text{at } \rho = 0 \\ \frac{\partial\theta}{\partial\rho} &= 0 & \text{at } \rho = 1 \end{aligned} \right\} \text{-at all } \xi \tag{10}$$

$$\left. \begin{aligned} \theta &= 1 & 0 \leq \rho \leq \beta \\ &= 0 & \beta < \rho \leq 1 \end{aligned} \right\} \text{-at } \xi = 0$$

where $\beta = a_T/a =$ ratio of source radius to radius of pipe.

Equation (8) or (9) is solved in conjunction with the boundary conditions given by Eq. (10) by the Crank-Nicolson (1947) implicit finite difference method. This consists in averaging the radial derivatives over two axial increments and replacing them by a central difference formula and representing the axial derivative by a forward difference scheme. The resulting set of equations together with two boundary conditions at $\rho = 0$ and $\rho = 1$ form a tridiagonal system, which is solved by the Thomas algorithm (Lapidus, 1962).

EXPERIMENTAL

A schematic diagram of the experimental set-up is given in Figure 2. It consisted of storage vessels for

the tracer and ambient liquids, which were pumped in through a series of rotameters via surge tanks. The rotameters were used to keep a steady flow rate, while the actual flow rate was measured by collecting a measured amount of liquid in a known time. A T-arrangement before entry into the test pipe helped in removing any entrapped air bubbles in the tracer liquid. The temperatures of both the streams were measured before entry into the test pipe.

The test section was divided into an entrance head and a measurement part. The entrance head was an ABS pipe consisting of a series of wire meshes placed at 45° to each other. The ambient liquid was introduced through two diagonal inlets and passed through the wire meshes before entry into the measuring part. The tracer liquid was introduced through a thin-walled stainless steel pipe 2.39 mm id which passed through the centre of the wire meshes and its length could be adjusted so that the tracer fluid made its entry under fully developed conditions. The entrance head fitted snugly into the test pipe. The measurements were carried out in a 12.7 mm id perspex tube 3.6 m long. It was held vertical by a series of clamps attached to a main frame, which itself was firmly supported. The verticality was periodically tested and necessary adjustments were made. The bottom end of the test pipe was connected to a return bend through a flexible rubber pipe, which maintained a column of liquid and thus prevented air bubbles rising up from the bottom of the tube at very low flow rates.

The measured property was the conductivity of the solute at the centre of the tube. Two conductivity probes were mounted at distances of 1500 and 3000 mm from the source. The probe was made of a 0.2 mm platinum wire electrode encased in a porcelain capillary. The other electrode was a 19 gauge hypodermic tubing. One end of the platinum wire was looped and insulated from the other electrode. The probe was mounted perpendicular to the pipe through a standard ABS fitting.

The conductivity was measured by means of a conductivity bridge through a selector switch which enabled the proper selection of the probe under use. The conductivity bridge chosen was a Wayne Kerr Autobalance Universal Bridge model and had an accuracy of 0.1%.

The tracer used was an indicator, which could colour the tracer stream, so that the spreading process could be visually monitored. The colouring also was helpful in pinpointing any channelling and local turbulence effects due to vibrations and proper care was taken to avoid these conditions.

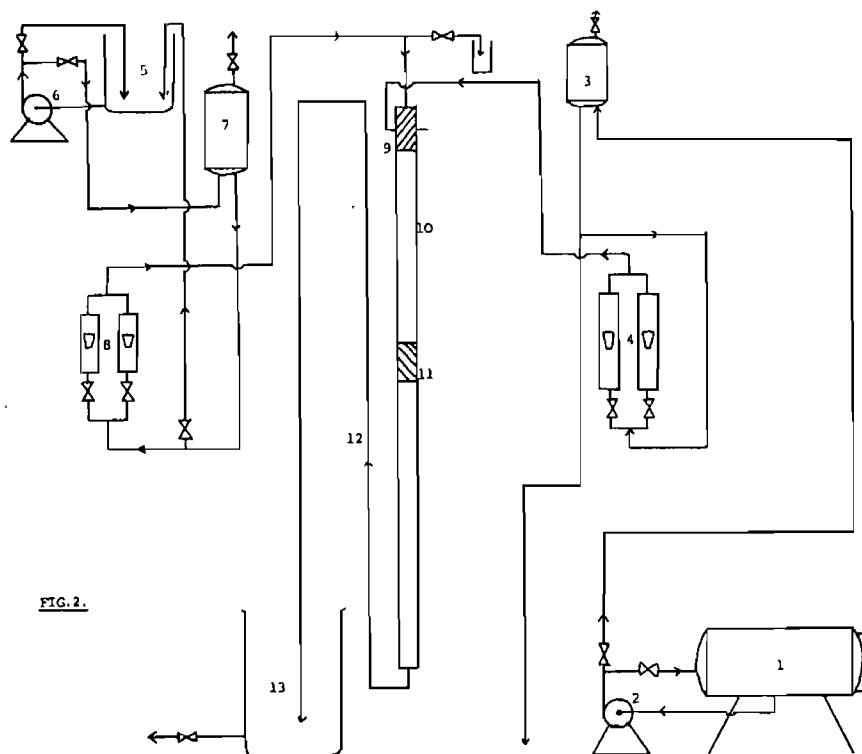


FIG. 2.

FIGURE 2 Schematic diagram of the experimental set-up. (1) Storage tank for the non-Newtonian liquid. (2) Mono pump for the non-Newtonian liquid. (3) Surge tank. (4) Ambient liquid side rotameters. (5) Storage tank for the tracer solution. (6) Mono pump for the tracer solution. (7) Surge tank. (8) Tracer side rotameters. (9) Entrance head. (10) Perspex pipe in which the measurements were made. (11) Test section. (12) Return U-bend. (13) Receiver for the spent solution.

The test liquid used was hydroxyethylcellulose (HEC)—Natrasol supplied by Hercules, NV—dissolved in water. The concentrations used were 0.7 and 1.2% by weight. The flow curves were measured on a Weissenberg Rheogoniometer (Model R18, Sangamo Western Controls Limited) placed in a constant temperature room. As the liquids passed through various shearing stages, rheological experiments were done before and after each set of experiments. During a day, the differences were not significant and hence an average value was used. The flow curves for the liquids are given in Figure 3 and fitted the power-law model with $n = 0.789$ and 0.314 and $K = 13.5$ and $32.5 \text{ dynes/cm}^2 \text{ sec}^{-n}$ for 0.7 and 1.2% HEC solutions respectively. The tracer solution was made by dissolving a known amount of NaOH in HEC solution to give 0.01 N strength. Phenol red was added to this solution to give a red colour.

The measured quantity was the local concentration on the axis of the tube as a function of the dimensionless distance ξ and by varying the average velocity

and carrying out measurements at two different axial distances, ξ was changed.

RESULTS AND DISCUSSION

Theoretical Results

The convective diffusion equations represented by Eqs. (8) and (9) for power-law and Ellis fluids respectively were solved on CDC7600.

The radial grid depended on the values of β , the dimensionless solute tube radius. The radius was initially divided into 200 parts and this facilitated the smoothing of the initial discontinuity. The axial step was initially set at 10^{-7} and progressively increased. The radial grid sizes were also varied and the solutions were found to be convergent. The results were also checked by comparing them with the Bessel function analytic solutions for a plug flow problem, i.e. $N \rightarrow \infty$ for power-law fluids.

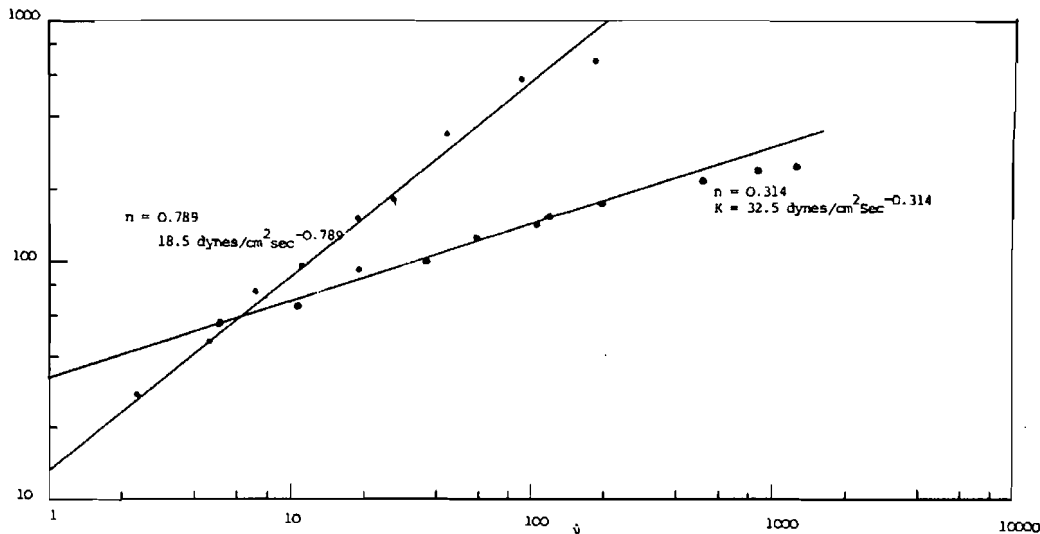


FIGURE 3 Rheograms for the test liquids.

One of the experimental methods to determine diffusion coefficients in a non-Newtonian liquid is by studying the centre-line concentration decay. A typical plot for the centre-line concentration decay as a function of dimensionless axial distance ξ is given in Figure 4 for power-law fluids. The process of decay appears to be slower for smaller ξ but speeds up for intermediate values of ξ . Then at large ξ it finally flattens and this would correspond to a uniform concentration across the cross-section of the tube. This value was calculated by a simple mass balance for the tracer. The final concentration in all cases agreed with the values obtained by the mass balance, thus adding to the credibility of the numerical technique. It is also clear from Figure 4 that the axial centre-line concentration decay is much more rapid as pseudoplasticity increases (or as N increases). This is understandable because the centre-line velocity is related to the average velocity as

$$V_{max} = V_{avg} \left(\frac{3 + N}{1 + N} \right) \quad (11)$$

Hence, the lower the value of N , the reciprocal power-law index, the higher is the centre-line velocity and the higher the effect of convection in the axial direction at the centre. Thus, for $N = \infty$, which corresponds to a plug flow case, the solute is spread solely due to molecular diffusion.

As mentioned earlier, the centre-line decay could be used as a useful method to determine the molecular diffusivity in a non-Newtonian liquid. If for a given ξ ,

which is defined as

$$\xi = ZD/V_{avg}a^2$$

Z , V_{avg} and " a " are treated as system parameters which can be varied at will, then " D " could be determined from a measurement of the decay of the centre-line concentration. More specifically, from the measurement of the centre-line concentration at different axial distances for different tubes and velocities for a liquid, whose rheology is fully known, a very good estimate of the diffusivity coefficient is obtained from a standard plot of θ_C vs. ξ .

The centre-line concentration decay also depends on β , the dimensionless solute tube radius. A typical figure to illustrate this effect of β on the spreading process on the centre-line concentration decay is shown in Figure 5 for a Newtonian liquid and a power-law liquid $N = 3.33$. It is evident that the smaller the value of β the faster is the decay. In addition to convection, the radial spread mechanism also depends on the molecular diffusion, which is essentially controlled by the concentration gradient available in the radial direction for the diffusion process to occur. Thus, the larger β , the lesser is the concentration gradient for large down-stream distances—thus, the fluid at $\rho = \beta$ has to spread before the centre-line concentration feels the effect of any appreciable gradient. Similar conclusions may be drawn for other non-Newtonian liquids.

For an Ellis fluid, the relevant rheological parameters are the Ellis fluid index and the shear stress

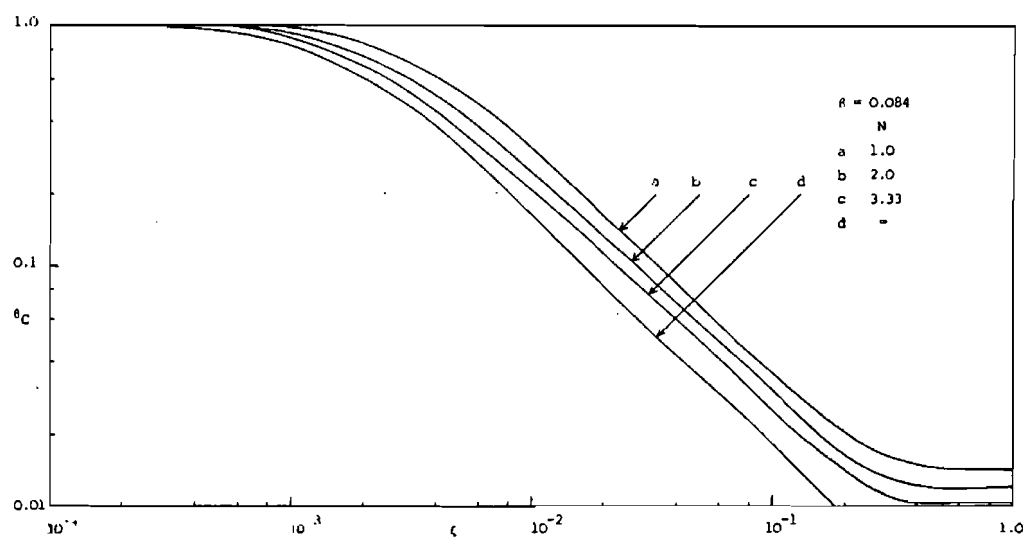


FIGURE 4 Influence of pseudoplasticity on centre-line concentration decay.

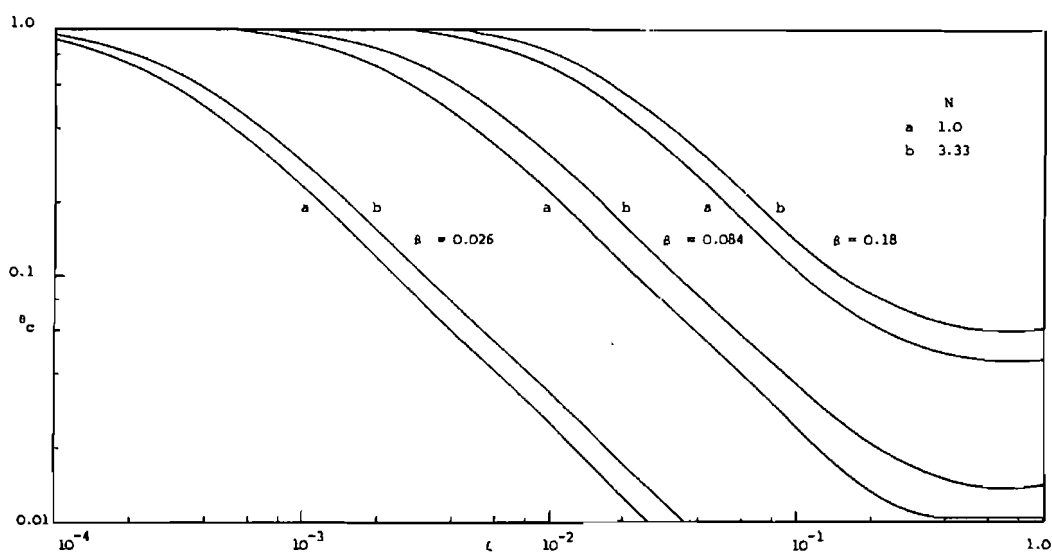


FIGURE 5 Effect of solute tube radius on centre-line concentration decay Newtonian and power-law fluids.

ratio, $m(\tau R/\tau_{1/2})$. For $\alpha = 1$, the solution reduces to the Newtonian case. For low shear stress values of the order of 1, the Ellis fluid index α has only a marginal effect and the differences cannot be graphically depicted clearly. For higher shear stress ratios, the effect is as in the case of power-law liquids as shown in Figures 6 to 8. In general, for a given shear stress ratio, the centre-line concentration decay is faster as the Ellis fluid index increases. This is because as α increases, the centre-line velocity decreases as

compared to the average velocity and hence the faster the decay. However, it is interesting to consider the effect of shear stress ratio m for a given α . A typical illustration is given in Table I for $\alpha = 1.5$. The effect is marginal. Thus it seems that the centre-line concentration decay is affected much more by the Ellis fluid index than by the shear stress ratio.

The radial concentration profiles obtained are plotted in Figures 9 to 11 for power-law fluids and Figures 12 and 13 for an Ellis fluid. These figures

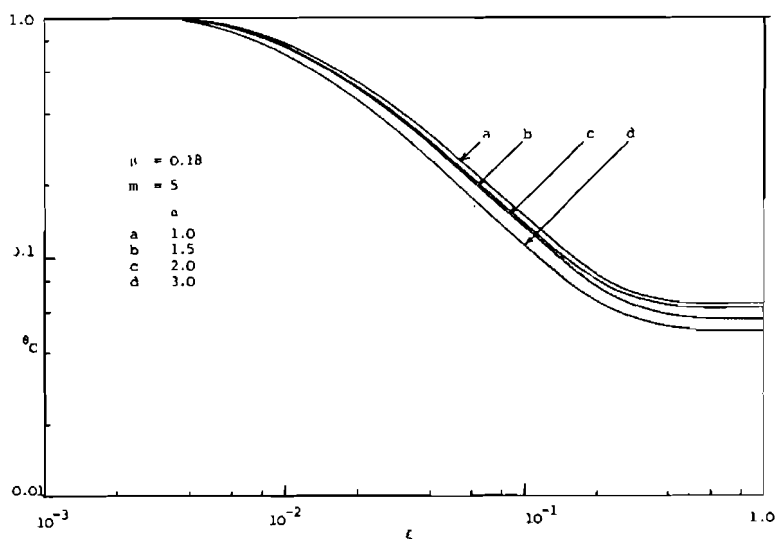


FIGURE 6 Effect of Ellis fluid index on centre-line concentration decay ($m = 5$).

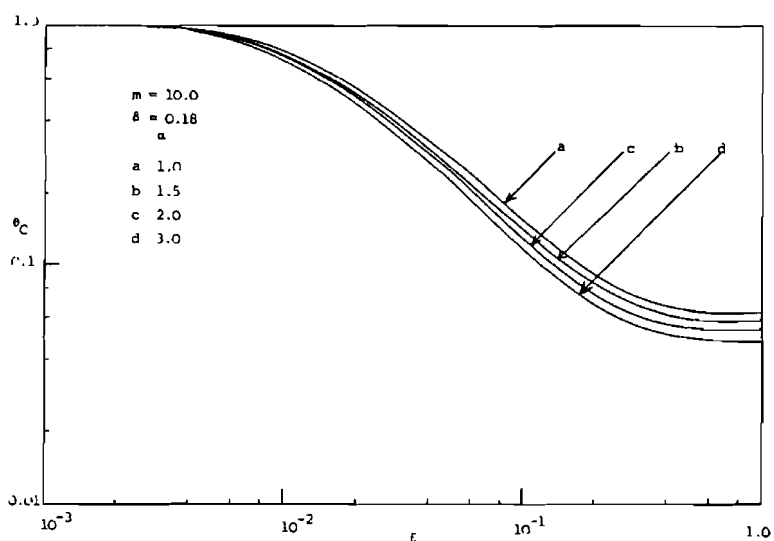


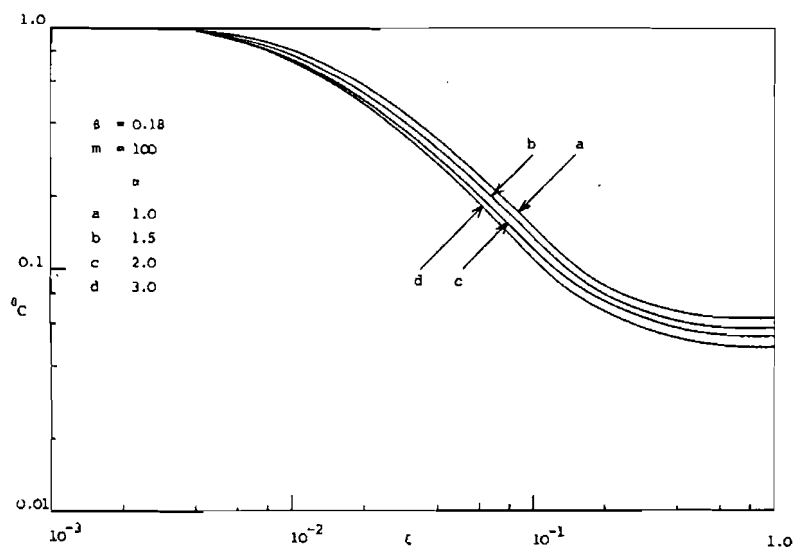
FIGURE 7 Effect of Ellis fluid index on centre-line decay ($m = 10$).

give some indication of the way in which radial spread occurs at different axial distances depending on β and the rheological parameters. In general, the radial profiles for Newtonian liquids tend to be sharper than the others and as the velocity profiles tend away from the Newtonian case, the concentration profiles become flatter. For a given β and a liquid of known rheological characteristics, the concentration profiles are skewed for short axial distances. This is when most of the material is concentrated in the region $\rho \ll \beta$. The concentration profiles become

flatter as the axial distances increase because of the spreading process. Finally, at large downstream distances, the concentration is uniform throughout the cross-section of the tube as can be predicted by a simple mass balance.

Experimental Results

The dimensionless solute radius used in the experiment was 0.18. The centre-line concentration decay curves were obtained by solution of the convective

FIGURE 8 Effect of Ellis fluid index on centre-line concentration decay ($m = 100$).

diffusion equation for $n = 0.789$ and 0.314 and 0.7 and 1.2% HEC, respectively, and are shown in Figures 14 and 15. The concentration was obtained from the conductivity values and the corresponding ξ values were obtained from Figures 14 and 15. The diffusivity values were evaluated from the definition of ξ .

The average diffusivities for 0.01 N NaOH diffusing into 1.2% Natrasol solution was 0.989×10^{-5} cm^2/sec and for 0.7% Natrasol was 1.004×10^{-5} cm^2/sec .

These average values were used to recalculate the dimensionless axial distance ξ at each concentration obtained experimentally and were replotted as shown

TABLE I
Centre-line concentration decay for an Ellis fluid—effect of shear stress ratio
 α : Ellis fluid index = 1.5
 ξ : Dimensionless axial distance

ξ	Shear stress ratio, m		
	5	10	100
0.0001	1.0	1.0	1.0
0.0010	1.0	1.0	1.0
0.0050	0.9512	0.9501	0.9476
0.0090	0.8143	0.8119	0.8067
0.0100	0.7803	0.7779	0.7723
0.0155	0.6236	0.6208	0.6146
0.0255	0.4478	0.4453	0.4397
0.0355	0.3484	0.3463	0.3415
0.0555	0.2413	0.2393	0.2361
0.0755	0.1851	0.1838	0.1808
0.0955	0.1506	0.1495	0.1470
0.1505	0.0997	0.0989	0.0972
0.2505	0.0705	0.0699	0.0687
0.7505	0.0602	0.0598	0.0589
0.9505	0.0602	0.0598	0.0589

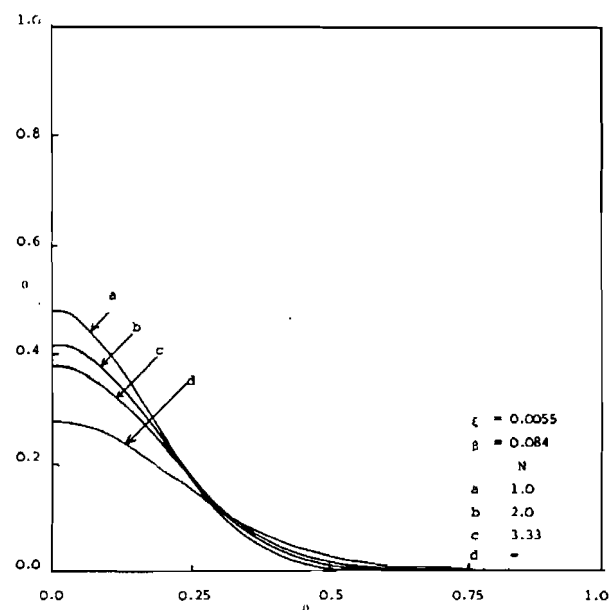


FIGURE 9 Typical radial concentration profiles for power-law fluids.

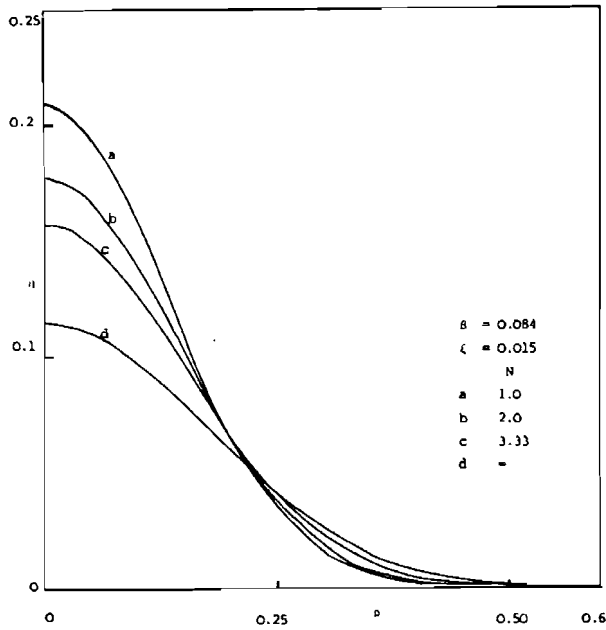


FIGURE 10 Typical radial concentration profiles for power-law fluids.

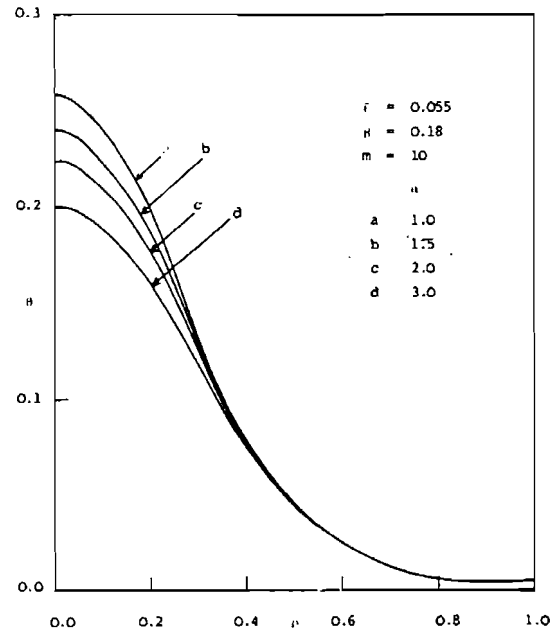


FIGURE 12 Typical radial concentration profiles for Ellis fluid ($m = 10$).

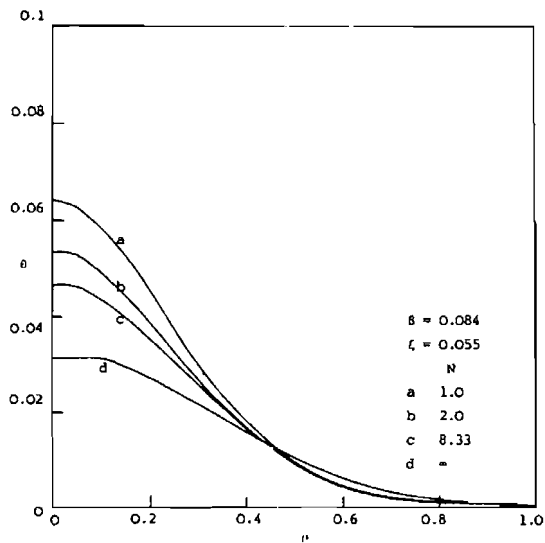


FIGURE 11 Typical radial concentration profiles for power-law fluids.

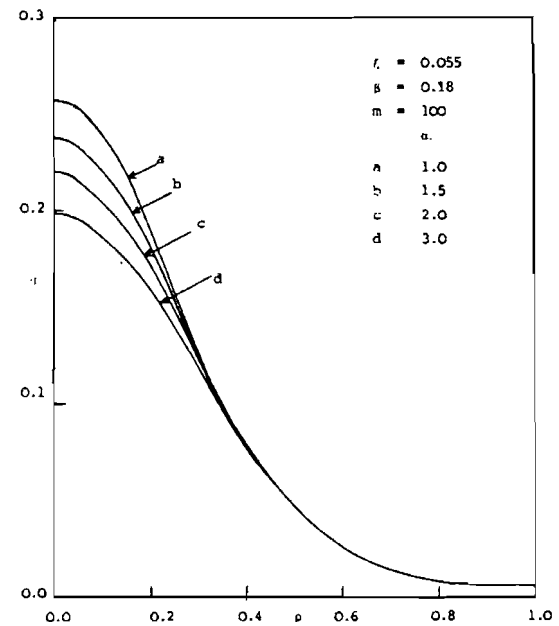
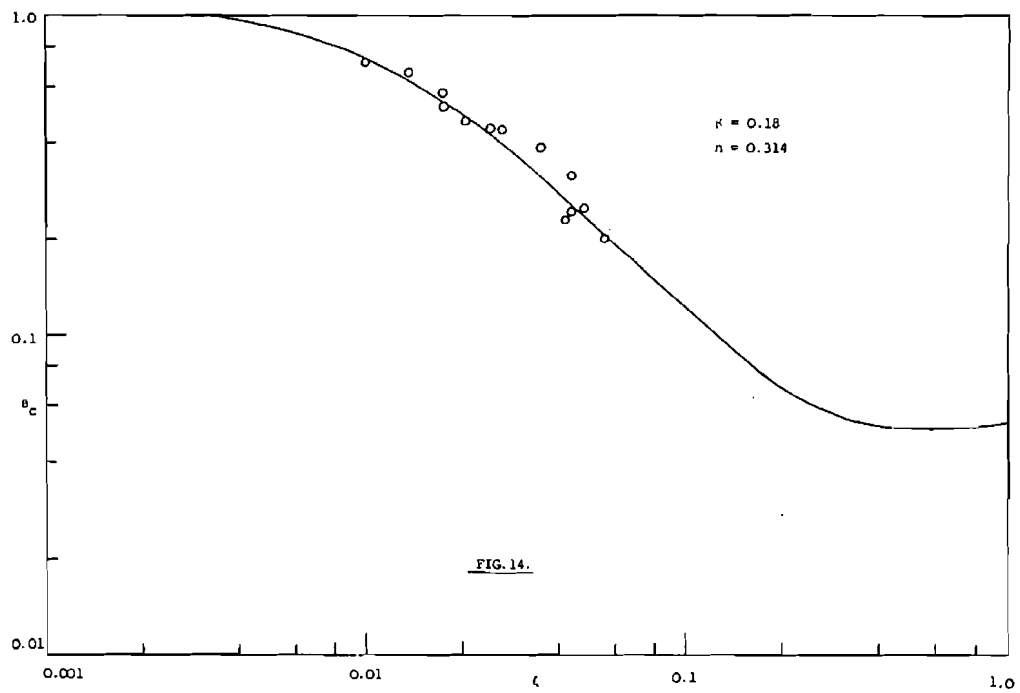
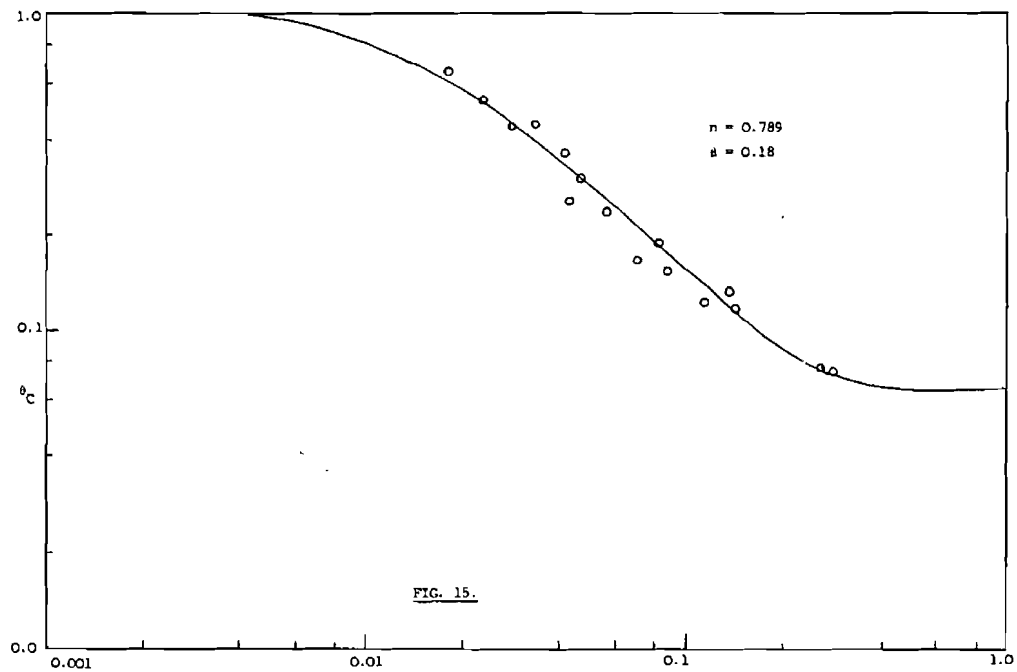


FIGURE 13 Typical radial concentration profiles for Ellis fluid ($m = 100$).

in Figures 14 and 15. The curve-fit appears to be excellent. It should be emphasized that the best region for diffusivity measurements should be the ones for which the curve of θ_c vs. ξ shows the maximum slope, since in this region, the sensitivity is maximum. Hence, diffusivity measurements should be confined to this region.

It would have been very useful to be able to compare the experimentally obtained diffusivity values with other data in the literature. Unfortunately, no data on NaOH diffusion in aqueous HEC solutions

FIGURE 14 Centre-line concentration decay curve for experimental solution ($n = 0.314$).FIGURE 15 Centre-line concentration decay curve for experimental solution ($n = 0.789$).

have been reported. The reported diffusivity values of NaOH in water at 15°C is $(1.54 \pm 0.04) 10^{-5}$ cm²/sec (*International Critical Tables of Numerical Data in Physics, Chemistry and Technology*, 1929). It would thus appear that even after the appropriate temperature correction, the diffusivity in HEC solutions is lowered. It is important to recognize, however, that these values are of the same order of magnitude as in water. This implies that in spite of the enormous rise in consistency due to the polymer addition, the diffusion coefficient appears to be unchanged. In other words, the macroscopic viscosity in such macromolecular solutions does not appear to represent the resistance to a diffusive process. This fact has been well established by now and the several experimental studies (see e.g. Astarita (1966); Astarita and Mashelkar (1977); Clough *et al.* (1962); Desai (1975); Mashelkar and Soylu (1974)) and the theoretical models (e.g., Navari *et al.* (1971)) confirm this finding.

The nature of the diffusivity reduction as a function of the polymer concentration appears to be approximately consistent with the theoretical model of Navari *et al.* (1971). These authors showed that after an initial sharp decrease in the diffusivity on initial polymer addition, the later decrease is much less sharp and indeed the diffusivity values level off to some extent. From the experimental observations reported in this work, we would appear to be confined to the latter region.

CONCLUSIONS

This work provides a theoretical study of the process of convective diffusion from a non-uniformly but symmetrically distributed source in the laminar flow field of a non-Newtonian fluid prevailing under steady conditions in a straight tube. An experimental set up was built up to simulate the model conditions and obtain molecular diffusivity values of NaOH in aqueous solutions of hydroxyethylcellulose. The measurements in this work are the first ones reported with this technique and consequently exploratory but they none-the-less demonstrate that this technique can be used successfully.

The technique studied here would be particularly suitable for diffusivity measurements in highly viscous media and settling suspensions for which the conventional static techniques may pose certain difficulties. Mashelkar and Venkatsubramanian (1975) have already elaborated the utility of such a flow technique for diffusivity measurement in blood at

low RBC concentrations, where the settling problem is acute. The potential of this method is thus worth exploring further not only with respect to mass diffusivity measurements but also the thermal diffusivity measurements, since the theoretical development in the foregoing is equally applicable to the corresponding heat transfer problem.

NOTATION

a	radius of tube
a_T	solute tube radius
c	local concentration
c_I	source concentration
D	diffusion coefficient
K	consistency index
m	shear-stress ratio for an Ellis fluid, $\tau_R/\tau_{1/2}$
n	power-law index
N	reciprocal of power-law index
r	radial distance
r_A	reaction or source-sink term in Eq. (1)
V_{avg}	average velocity
V_{max}	maximum velocity
V_Z	axial velocity component
Z	axial distance

Greek Letters

α	Ellis fluid index
β	dimensionless solute tube radius, a_T/a
η_0	Zero-shear viscosity
θ	dimensionless concentration, c/c_I
θ_C	centre-line concentration
$\dot{\gamma}$	shear rate
ξ	dimensionless axial distance, $ZD/V_{\text{avg}}a^2$
ρ	dimensionless radial distance, r/a
τ	shear stress
τ_R	wall shear stress
$\tau_{1/2}$	shear stress at which shear rate equals half the wall shear rate

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