one may relate the measured dipole moment to those of the two lowest energy conformers approximately by using eq 2 with $\omega = 1.0$ (since the degeneracy is the same in both states). In Figure 2b we plot $\mu^2$ against $T$ for E134 along with a best fit curve of the data to eq 2. The best fit parameters are $\mu_1 = 1.20 \pm 0.06$ D (gauche–gauche conformers), $\mu_2 = 2.83 \pm 0.04$ D (gauche–cis conformers), and $\Delta E = 2.98 \pm 0.2$ kJ mol$^{-1}$.

For R123a we initially estimated $P_\alpha$ and found $P_\alpha = 1.17P_{\alpha e} = 24.2$ cm$^3$ mol$^{-1}$. We found that the dipole moment of R123a is $\mu = 1.302 \pm 0.007$ D and does not vary noticeably with temperature, although in principle it could because of the existence of nondegenerate conformers in its structure. Because of the constant value of the moment of R123a, we reevaluated the dipole moment using the method for temperature-independent moments and placed it in Table III.

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# Diffusion Coefficient in Aqueous Polymer Solutions

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**The diffusion coefficients for benzoic acid in a number of aqueous solutions of poly(vinyl alcohol) (3.0 and 6.0% polymer by weight) and poly(ethylene oxide) (0.75–3.5% polymer by weight) are reported. The diffusivity values are obtained by measuring the rate of mass transfer of benzoic acid from the surfaces of rotating disks. Data on solubility of benzoic acid in poly(ethylene oxide) are also reported. The diffusivities in the polymer solutions are found to be independent of shear rate and are lower than those in water.**

## Introduction

Studies on mass-transfer characteristics of highly viscous Newtonian and non-Newtonian polymeric fluids have assumed importance in recent years due to their application in several chemical and biochemical processes, like diffusion-controlled polymerization, fermentation processes, and the life processes of microorganisms, animals and plants, etc. The diffusion coefficient of the solute in the fluid is the prime factor which influences the rate of mass transfer in such systems. Fairly efficient predictive correlations are available for Newtonian fluids (*1*). But, apparently due to the very diverse nature of the polymeric solutions, until now, developing a totally predictive theory for diffusion in such systems appears to be a remote possibility. Therefore, it becomes essential to resort to experimental measurement of diffusivity in such systems. Considerable experimental information is available in the literature on the diffusivity of gases in non-Newtonian fluids (*2*). Such information on liquid and solid solutes is lacking.

Out of the various methods used by previous workers, the diffusion-cell, interferometric, membrane, polarographic, refractometric, and other stagnant-medium techniques are ineffective in showing the effect of shear rate, if any, on the molecular diffusivity in polymer solutions (*3*). Thus flow techniques like laminar jet (*4–7*), falling films and wetted walls (*8–10*), rotating cones, disks, and spheres (*11–21*), dispersion in flowing fluid stream (*22*), and dissolution during laminar flow along plates and through tubes (*3*, *23*, *24*) have been extensively used for the measurement of molecular diffusivity in polymer solutions. In many of these the experimental conditions were such that mass transfer occurred in a constant-velocity (or zero-shear) field (*4–10*). Consequently, the influence of shear rate on the molecular diffusivity could not be assessed from these studies. Remaining techniques (*11–24*), however, do indicate that the molecular diffusivities remain unaffected in a finite shear field.

Some of the flow techniques, which have been successfully used with gases, are unsuitable for solid solutes. The diffusivity of such solutes in polymer solutions has been determined either by laminar flow dissolution from inclined plates, rotating disks, and tubes or by laminar dispersion techniques. The rotating-disk technique offers several advantages over other techniques. It

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represents a system of well-defined three-dimensional flow, which allows complete analytical solution to the equation of motion. Further, it has the distinct advantage of uniform surface accessibility because the diffusional field is one dimensional in laminar flow. Experimentally, also, the rotating disk has several advantages over other geometries. The need for lengthy test pieces and recirculation pumps, etc., is eliminated, and all that is needed is a small pool of fluid surrounding the disk. The disk itself is relatively easy to fabricate. In view of these advantages, this technique has been used in this work for obtaining the diffusivity of a sparingly soluble solute like benzoic acid in aqueous solutions of poly(vinyl alcohol) (PVA) and poly(ethylene oxide) (Polyox), which have been assumed to be inelastic power law fluids.

## Theoretical Background

When a disk rotates steadily in a large reservoir of a Newtonian fluid, which is otherwise at rest apart from the motion imparted by the rotation of the disk, the heat or mass transfer between the fluid and the disk–fluid interface obeys the relation (*25–27*)

$$J_h \text{ or } J_m = (\rho\mu\omega)^{1/2} f(Pr \text{ or } Sc) \qquad (1)$$

where $\omega$ is the angular velocity (radians/s) and $f(Pr$ or $Sc)$ is a function of Prandtl ($Pr$) or Schmidt ($Sc$) number for which particular values are given by several workers (*25–28*).

For laminar flow dissolution ($Re < 5 \times 10^5$) of a sparingly soluble solute from a rotating disk surface in a fluid like water, i.e. for systems with large $Sc$, eq 1 gives (*28–30*)

$$J_m = 0.62048 C_s D_{M,s}^{2/3} \mu^{-1/6} \rho^{1/6} \omega^{1/2} \qquad (2)$$

This agrees well with experiments (*28–30*). It is interesting to note that the radial location of the point at which mass transfer is being measured does not appear in eq 2, and thus the mass flux has the same value at all points on the disk surface. Spalding (*28*) suggested that eq 2 could be used for calculating the diffusivity by measuring the dissolution rate from the surface of a rotating disk.

Several investigators have considered the flow of a non-Newtonian fluid on a rotating disk (*31–33*). Mitschka and Ulbrecht (*33*) obtained exact solutions for flow around cones and disks rotating in a power-law fluid. The average shear stress at the surface of the disk can be given as

$$\tau_{av} = \frac{n+1}{2n+1} \beta(n)\, K(\omega)^{3n/(n+1)} \left(\frac{K}{\rho}\right)^{-1/(n+1)} R^{2n/(n+1)} \qquad (3)$$

where

$$\beta(n) = 0.1539(5n+3)/(n+1)(6.13)^{(n-1)/[2(n+1)]} \qquad (4)$$

It is seen that $\tau_{av}$ is a strong function of angular velocity because, if all other parameters are fixed,

$$\tau_{av} \propto (\omega)^{3n/(n+1)} \qquad (5)$$

Hansford and Litt (*12*) considered the case of mass transfer from a disk rotating in non-Newtonian solutions. They obtained the following expression for the average mass flux

$$J_m = \left(\frac{a'}{3}\right)^{1/3} \left(\frac{6n+6}{3n+7}\right) \left(\frac{C_S D_{M,PS}^{2/3}}{0.893}\right) \times \left(\frac{K}{\rho}\right)^{-1/3(n+1)} R^{(1-n)/3(1+n)} (\omega)^{1/(1+n)} \qquad (6)$$

For $n = 1$, eq 6 reduces to eq 2. When all other parameters are fixed, it can be shown that

$$J_m \propto R^{(1/3)[(1-n)/(1+n)]} \qquad (7)$$

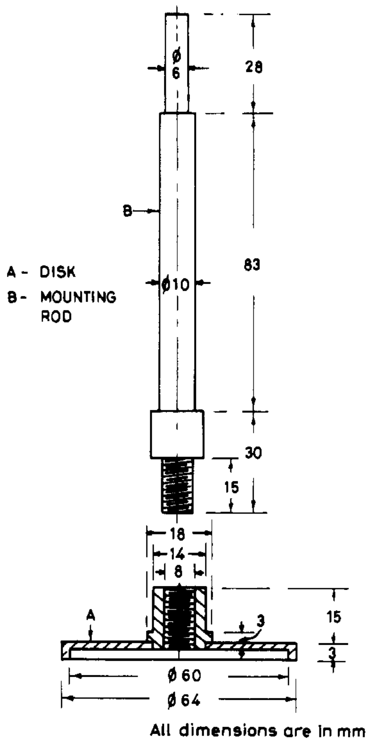

**Figure 1.** Details of the disk.

Thus for non-Newtonian fluids the disk surface is no more uniformly accessible for mass transfer. Hansford and Litt (*12*) and Grief et al. (*14*) showed that eq 6 can be used for evaluating the diffusivity of solutes from mass-transfer measurements. Mishra and Singh (*19*) extended the above analysis and obtained a generalized relation for mass transfer from disks rotating in power-law fluids. Pathak et al. (*20*) and Lal et al. (*21*) used this technique for measuring the diffusivity of benzoic acid in aqueous carboxymethyl cellulose solutions. The present experimental data will be analyzed in the light of this background.

## Experimental Section

Mass-transfer rates were determined by rotating a disk of a solid organic solute supported on aluminum dishes in aqueous polymer solutions. Dishes of diameter 4.5–10.5 cm and each having 3-mm-high rims were used for casting the disks. All such dishes had a threaded connecting bush on the center of their back for mounting them onto the shaft of the rotor. The details of a dish are shown in Figure 1. Special care was taken during the machining of these support dishes to ensure that the entire assembly rotated in the vessel without wobbling throughout the entire speed range covered in the experiments. All the disks were cast, surface-finished, washed, and dried to a constant weight before being used in the runs. The procedure adopted for this purpose was the same as that detailed elsewhere (*19–21*).

The dissolution rate was determined as the weight loss of a disk over a timed run. A schematic diagram showing details of the experimental setup used is given in Figure 2. A disk weighed accurately to the nearest 0.05 mg was mounted onto the rotor and run for a predetermined time in the solution kept in a thermostated cylindrical vessel. The vessel was a 28-cm-diameter and 40-cm-height flat-bottom cylindrical glass vessel having four standard baffles (width = $^1/_{10}$th of the vessel diameter). The depth of immersion of the disk was kept at a height of one-third of the vessel diameter from the bottom. The duration of the run was timed to give a minimum weight loss of 0.1 g. At the end of the run, the disk was removed from the

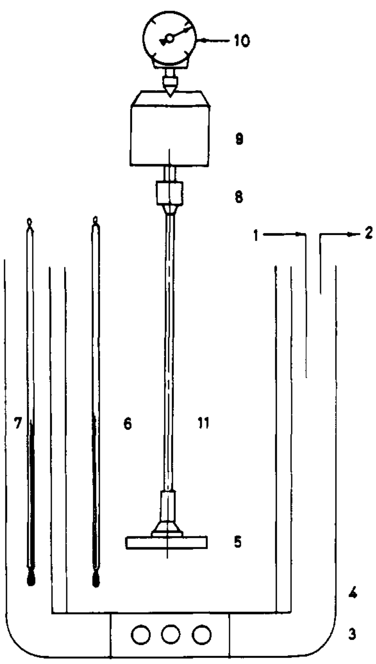

**Figure 2.** Schematic diagram of the diffusivity setup: (*1*) water inlet, (2) water outlet, (3) constant-temperature water bath, (4) baffled vessel, (5) coated disk, (6) inner-vessel thermometer, (7) outer-vessel thermometer, (8) drill chuck, (9) motor, (10) tachometer, and (11) shaft.

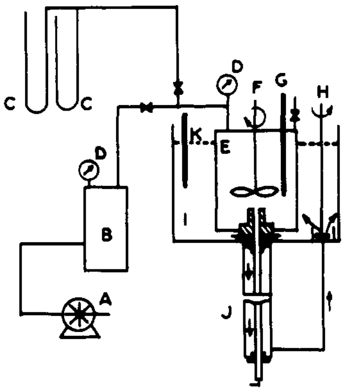

**Figure 3.** Capillary tube viscometer system: (A) air compressor, (B) air tank, (C) manometer, (D) pressure gauge, (E) fluid container, (F) stirrer, (G) thermometer, (H) stirrer, (I) constant-temperature water bath, (J) capillary tube, and (K) thermometer.

solution and washed with a saturated solution of benzoic acid to remove the excess viscous polymer, blotted lightly with filter paper and dried in a dessicator for 24 h before reweighing. All the runs were made in duplicate; some were also made in triplicate. In a separate set of blank runs, losses in weight during installation of the disks in the vessel, during their removal from the vessel, and during their washing were determined and were used as corrections. The polymer solutions were prepared by dissolving an appropriate amount of polymer in a known amount of distilled water. Special care was taken to prevent the formation of "cat's eyes" during solution preparation. Polymers used were poly(vinyl alcohol) (BDH, product no. 30573; MW = 14 000; viscosity of 4% aqueous solution at 20 °C = 4-6 cP) and polyox (BDH, product no. 29760; MW = 300 000, viscosity of 5% aqueous solution at 25 °C = 550-900 cP).

The rheological parameters of the polymer solutions were determined from the flow curves prepared with the help of a locally constructed capillary-tube viscometer (*3*). A line diagram of the capillary-tube viscometer system is shown in Figure 3. The range of shear rate covered in these experiments was from 230 to 3956 $s^{-1}$. Densities of the solutions were determined with the help of a pycnometer. The saturation solubilities of benzoic acid in aqueous polyox solutions were determined by the equilibration technique used earlier (*33-36*), and those for PVA solutions were taken from Sahay et al. (*36*). Solubility and viscosity data for distilled water were those reported in the literature (*3*).

**Table I. Diffusivity and Rheological Constants**

| temp/ °C | solution (% by weight) | $n$ | $K$/ (kg/(m·$s^{2-n}$)) | $10^{10}D_M$/ ($m^2$/s) |
|---|---|---|---|---|
| 18.0 | water | 1.0 | 0.001 | 9.036 |
| 19.0 | 0.75% aq polyox | 0.966 | 75.56 × $10^{-4}$ | 8.679 |
| 17.5 | 1.00% aq polyox | 0.914 | 106.51 × $10^{-4}$ | 7.585 |
| 18.0 | 1.25% aq polyox | 0.863 | 176.95 × $10^{-4}$ | 7.556 |
| 19.0 | 1.40% aq polyox | 0.847 | 314.70 × $10^{-4}$ | 8.298 |
| 19.5 | 2.50% aq polyox | 0.810 | 608.74 × $10^{-4}$ | 8.193 |
| 19.5 | 3.50% aq polyox | 0.787 | 1448.41 × $10^{-4}$ | 7.401 |
| 30.0 | 3.00% aq PVA | 0.973 | 70.938 × $10^{-4}$ | 6.712 |
| 29.5 | 6.00% aq PVA | 0.765 | 595.4 × $10^{-4}$ | 2.140 |

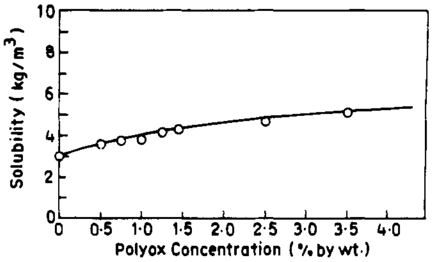

**Figure 4.** Solubility of benzoic acid in aqueous polyox solutions (effect of polymer concentration).

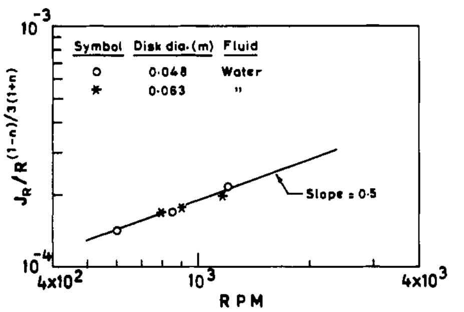

**Figure 5.** Plot of mass flux vs rpm (water).

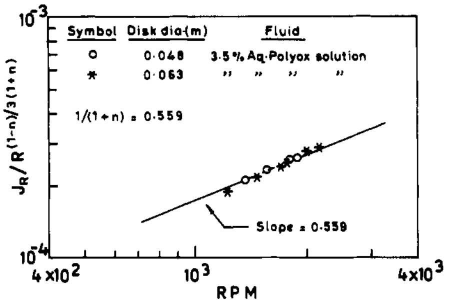

**Figure 6.** Plot of mass flux vs rpm (3.5% aqueous polyox solution).

## Results and Discussion

The rheological parameters obtained by analyzing the capillary flow data for the aqueous polyox and aqueous PVA solutions are summarized in Table I. It was observed that within the range of the shear rate studied both the solutions exhibited pseudoplastic behavior and obeyed the power-law model.

The results of solubility determination are given in Table II and Figure 4. It is seen from Figure 4 that the solubility of benzoic acid in aqueous poly(ethylene oxide) solutions increases with increasing polymer concentration and approaches a con-

**Table II. Saturated Solubility (kg/m³) of Benzoic Acid in Aqueous Poly(ethylene oxide) Solution**

| | | solubility for given polymer concn (wt %) | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| run no. | temp/°C | 0.0 | 0.5 | 0.75 | 1.00 | 1.25 | 1.40 | 2.5 | 3.5 |
| 1 | 17.5 | | | | 3.6547 | | | | |
| 2 | 18.0 | 3.0000 | 3.3906 | | | 3.921 | | | |
| 3 | 19.0 | | | 3.6045 | | | 4.1351 | | |
| 4 | 19.5 | | | | | | | 4.5073 | 5.037 |
| 5 | 20.0 | 3.2163 | 3.6066 | 3.7125 | 3.9247 | 4.1371 | 4.2431 | 4.5613 | 5.091 |

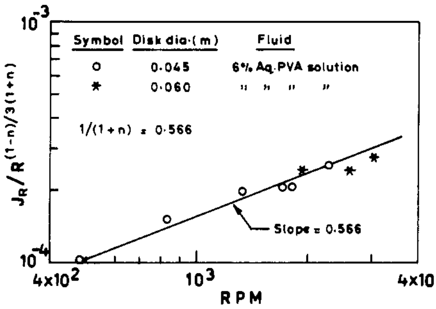

**Figure 7.** Plot of mass flux vs rpm (6% aqueous PVA solution).

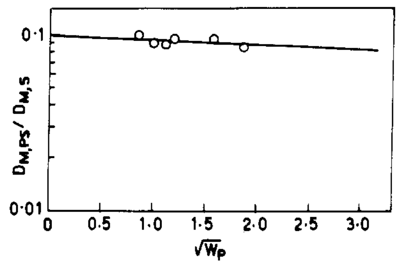

**Figure 8.** Effect of polymer concentration on diffusivity (benzoic acid–aqueous polyox solutions).

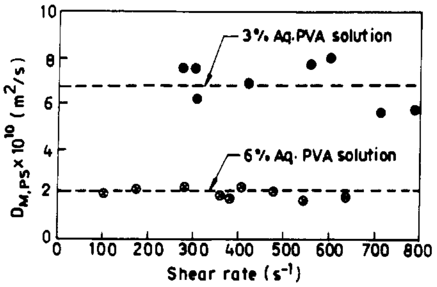

**Figure 9.** Effect of shear rate on diffusivity (aqueous PVA solutions).

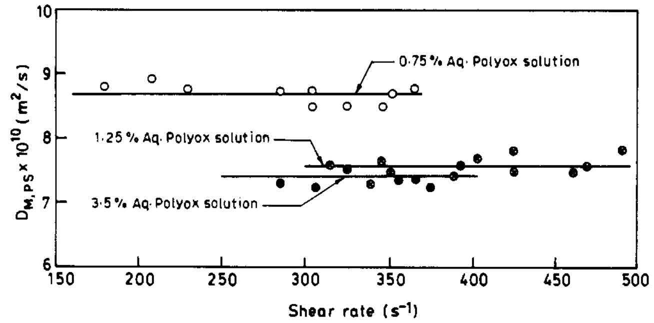

**Figure 10.** Effect of shear rate on diffusivity (aqueous polyox solutions).

stant value at higher concentrations, which is in agreement with the observations made earlier (*34–36*) for other aqueous polymeric solutions.

Typical plots showing the variation of the rate of mass transfer with the rotational speed of the disk are shown in Figures 5–7. As required by the theoretical analysis (*12*), experimental results show a $1/(1 + n)$ power dependence of the mass flux on the rotational speed. As the condition of uniform accessibility no longer exists and the mass flux for non-Newtonian fluids is dependent upon the radius, therefore, the term involving the disk radius has been included with mass flux to get a single line for all the disks used with a particular solution. These data were used to calculate the molecular diffusivities. The mean values of diffusivities for various polymer solutions are listed in Table I.

For both polymers studied, the diffusivity decreases as the polymer concentration increases. Similar effect of polymer concentration on diffusivity of small solutes diffusing in aqueous PVA and polyox solutions have been reported by others (*8*, *14*, *16*). This decrease has been reported to be to the extent of 30–40% of the diffusivity of the same solute in pure water. Lal et al. (*21*) showed that the ratio of diffusivity in polymer solution to that in water is a function of the square root of the polymer concentration and, for the benzoic acid–aqueous carboxymethyl cellulose (CMC) solution system, it can be expressed as

$$D_{M,PS}/D_{M,S} = 10^{-0.984(W_p)^{1/2}} \qquad (8)$$

where $W_p$ is the weight fraction. A similar relation appears to be true for aqueous polyox and PVA solutions as well. A typical plot for polyox solutions is shown in Figure 8. The diffusivity data for these fluids can also be represented by an equation similar to eq 8, differing only in the constant on the right-hand-side exponent, which will be a function of the nature of the polymer.

Plots showing the effect of shear rate on diffusivity in polyox and PVA solutions are given in Figures 9 and 10. Within the range studied, no effect of shear rate is observed on the diffusivity values. This is in agreement with the observations made by Clough et al. (*23*), Mishra and Singh (*19*), and Kumar and Upadhyay (*3*), Lal et al. (*21*), and Pathak et al. (*20*) for benzoic acid diffusing in other non-Newtonian fluids.

A quite confusing situation prevails in the published literature on the effect of shear rate on diffusivity. Several workers have reported a strong dependence of diffusivity on shear rate while others have reported either a mild or no effect (*2*). According to Mashelkar (*37*), many of these conclusions appear to be doubtful. By analyzing the data of Grief et al. (*14*), he showed that over an approximately 50-fold change in shear stress the diffusivity remained essentially constant. Kumar and Upadhyay (*3*), Lal et al. (*21*), and Pathak et al. (*20*) showed that the diffusivity of benzoic acid in aqueous carboxymethyl cellulose solutions remained unaffected by the shear rate. Kumar and Upadhyay (*3*) covered a 7-fold (approximately) range of shear rate in their investigations.

Hubbard and co-workers (*2*, *38–40*) reported the existence of a critical shear rate below which the diffusivity decreased slowly with increasing shear rate and above which it decreased sharply. This was attributed to the change in the molecular conformation of polymer with increasing shear rate. Edwards and Smith (*41*) reported that the effect of the shear rate depended on the nature and concentration of polymer. For low concentrations of polymers they did not observe any effect of shear rate on diffusivity. The shear rate range covered by these workers, however, is very narrow, and the system geometry does not provide the advantages of a rotating-disk system.

In the present work, a 3–4-fold range of shear rate has been covered. In view of these observations and the inferences drawn by others (*3*, *20*, *21*, *23*, *37*), it is justified to conclude that diffusivity of small solutes in dilute polymer solutions is independent of shear rate.

The effect of viscoelasticity appears to have been a priori neglected by almost all workers. Several of the polymer solutions for which diffusivity data have been obtained are reported to be possessing elastic properties and thus the neglect of elastic properties may not be justified. A theoretical analysis of the mass transport from a disk rotating under laminar boundary layer flow conditions into a Walters B′ liquid indicated that elasticity enhanced the transport rate to some extent (*37*). This finite effect is likely to have some influence on the deduced values of diffusivities. Since the majority of the workers have obtained their data with highly dilute polymer solutions with experimental techniques unsuitable for resolving the nature and extent of the dependency of diffusivity on elastic properties, the issue remains unresolved. There appears to be a need for additional experimental information concerning this aspect.

## Conclusions

The solubility of benzoic acid in aqueous polyox solutions increases with increasing polymer concentration up to a maximum value; thereafter it tends to be almost constant.

Diffusivity of benzoic acid in the polymer solutions is independent of the shear rate, and it decreases with increasing polymer concentration.

## Nomenclature

| | |
|---|---|
| $a'$ | constant |
| $C_S$ | solubility ($ML^{-3}$) |
| $C_p$ | heat capacity ($L^2T^{-2}\theta^{-1}$) |
| $D_M$ | mean diffusivity ($L^2T^{-1}$) |
| $D_{M,PS}$ | diffusivity in polymer solution ($L^2T^{-1}$) |
| $D_{M,S}$ | diffusivity in pure solvent ($L^2T^{-1}$) |
| $J_h$ | average heat flux ($MT^{-3}$) |
| $J_m$ | average mass flux ($ML^{-2}T^{-1}$) |
| $K$ | consistency index ($ML^{-1}T^{n-2}$) |
| $k$ | thermal conductivity ($MLT^{-3}\theta^{-1}$) |
| $n$ | flow behavior index |
| $Pr$ | Prandtl number ($\mu C_p/k$) |
| $R$ | radius of disk ($L$) |
| $Sc$ | Schmidt number ($\mu/\rho D_M$) |
| $W_P$ | weight fraction of polymer in solution |

### *Greek Letters*

| | |
|---|---|
| $\beta(n)$ | function of $n$ |
| $\rho$ | density ($ML^{-3}$) |
| $\omega$ | angular velocity ($T^{-1}$) |
| $\mu$ | viscosity ($ML^{-1}T^{-1}$) |
| $\tau_{av}$ | shear stress ($ML^{-1}T^{-2}$) |

**Registry No.** PVA, 9002-89-5; Polyox, 25322-68-3; benzoic acid, 65-85-0.

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