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HYDRODYNAMICALLY INDUCED CROSS STREAM MIGRATION OF DISSOLVED MACROMOLECULES (MODELLED AS NONLINEARLY ELASTIC DUMBBELLS)†

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Abstract—The hydrodynamically induced cross-stream migration of a nonlinear elastic dumbbell in non-homogeneous flows is considered. A generally valid expression for the migration velocity is derived and the result applied to viscometric flows. Effects of elasticity and size of the particle, as well as the strength of the flow field, are studied in detail for channel flow and for circular Couette flow. Estimates of the dynamics of developments of non-uniform concentration profiles are discussed.

1. INTRODUCTION

Essentially all flow fields of practical interest involve non-homogeneous flows, i.e. flows, for which the velocity gradients vary spatially over the domain of interest. To know if and to what extent suspended particles or macromolecules migrate systematically across streamlines is therefore of great importance. Such behavior would not only affect the rheology and the engineering of polymer solutions and suspensions but also influence processes such as heat and mass exchange between the suspension or solution and the bounding walls. There is a growing body of experimental evidence that such cross-stream migration: (i) may be important in flows of polymer solutions through porous media (Aubert *et al.* 1980); (ii) leads to molecular fractionation in tube flow (Busse 1964, Schreiber *et al.* 1965); and (iii) is responsible for the decrease of the apparent viscosity with decreasing dimensions of the viscometer (Kemblowski 1969, Porter *et al.* 1966).

Furthermore, the decay in the degree of extrudate swelling with increasing die length also has been attributed to the cross-stream migration (Schreiber *et al.* 1966). Finally, more on a laboratory scale we mention chromatography experiments (Quano *et al.* 1971), slip in gravity flow along an inclined plane (Astarita *et al.* 1964; Therien *et al.* 1970) the Uhlenhopp effect (Shafer *et al.* 1974) and the radial migration of droplets (Goldsmith *et al.* 1962).

By carefully examining all of these different experimental results two common features are detected: first of all, the particles or macromolecules in question are nonrigid (i.e. deformable) and secondly the flow field clearly is non-homogeneous.

It is the purpose of this communication to take these two features into consideration by analyzing the hydrodynamics of one particular system: a nonlinear elastic dumbbell with unstretched length L_0. This kind of particle mimics some features of dissolved macromolecules (Bird *et al.* 1977) and is simple enough to be treated in detail, yet rich enough to furnish information about various effects. Besides the influence of non-zero length L_0 we mention spring-stiffness (fast elastic response), finite and nonlinear extensibility (up to the limit of a rigid dumbbell), and weak and strong flow fields. Furthermore, as soon as the hydrodynamics is fully understood one can rigorously formulate a statistical mechanical treatment of a suspension of such particles. The present work represents a first step in that direction.

In section two we shall formulate the problem mathematically and derive, for the particles at hand, a general result, i.e. one which is valid in arbitrary non-homogeneous flows. This will then be applied to channel flow (section 3) and to circular Couette flow (section 4). Finally, in section five the results will be discussed and general conclusions will be drawn.

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2. FORMULATION OF THE PROBLEM

The particle considered in this study consists of two identical spheres (radius a) joined by some kind of nonbendable, frictionless spring. If $\mathbf{r}_i$ is the instantaneous position vector of the center of sphere i, $i = 1, 2$, then $\mathbf{R} = \mathbf{r}_2 - \mathbf{r}_1$ specifies the actual configuration (see figure 1). The force law for the spring, i.e. the force $\mathbf{F}^c$ on sphere "2" due to tension in the connector will be collinear with $\mathbf{R}$ and we write

$$\mathbf{F}^c = F^c\hat{\mathbf{R}} = -\frac{d\Phi}{dR}\hat{\mathbf{R}}, \qquad [2.1]$$

where $\Phi = \Phi(|\mathbf{R}|)$ is the connector potential.

Moving through the fluid, each sphere will experience a hydrodynamic force $\mathbf{F}_i$ and a torque $\mathbf{G}_i$, $i = 1, 2$. If

$$\hat{\mathbf{R}} \cdot (\mathbf{F}_2 - \mathbf{F}_1) - 2\frac{d\Phi}{dR} = 0, \qquad [2.2a]$$

$$\mathbf{F} \equiv \mathbf{F}_1 + \mathbf{F}_2 = 0, \qquad [2.2b]$$

$$\mathbf{G}_c = 0, \qquad [2.2c]$$

where $\mathbf{F}$ is the net (total) hydrodynamic force and $\mathbf{G}_c$ the net total torque (relative to the center $\mathbf{r}_c$), the dumbbell is said to be freely suspended. From these equations, the state of motion of the dumbbell, i.e. the translation of the center (velocity $\dot{\mathbf{r}}_c$), the rotation around the center (angular velocity $\mathbf{w}$) and the vibration of the spring (characterized by $\dot{R}$, the change in length) can be deduced. This requires that we know the relation between the "forces" $\mathbf{F}, \mathbf{G}_c$ & $\hat{\mathbf{R}} \cdot (\mathbf{F}_2 - \mathbf{F}_1)$ and the "momenta" $\dot{\mathbf{r}}_c$, $\mathbf{w}$ & $\dot{R}$.

To this end we assume that $\mathbf{V}^0$, the undisturbed and prescribed flow field in the vicinity of the particle is of the form

$$\mathbf{V}^0(\mathbf{r}) = \mathbf{V}_c^{\,0} + (\mathbf{r} - \mathbf{r}_c) \cdot \mathbf{E}_c + \boldsymbol{\Omega}_c \times (\mathbf{r} - \mathbf{r}_c) + \frac{1}{2}(\mathbf{r} - \mathbf{r}_c)(\mathbf{r} - \mathbf{r}_c):\boldsymbol{\Gamma}_c \qquad [2.3]$$

In this equation, the velocity $\mathbf{V}_c^{\,0}$, the rate of strain dyadic $\mathbf{E}_c$, the vorticity vector $2\boldsymbol{\Omega}_c$ and the second order velocity gradient $\boldsymbol{\Gamma}_c$ are all evaluated at the center $\mathbf{r}_c$. The quadradic variation of $\mathbf{V}^0(\mathbf{r})$ with position need not be valid everywhere, as in channel flow or tube flow, but only in the vicinity of the particle. If we then assume that the Reynolds number based on particle dimensions is very small, the velocity-pressure disturbances caused by the dumbbell satisfy the Stokes-equations, which are linear. Thus, if the spring force $\mathbf{F}^c$ is such as to guarantee that the two spheres are far apart at each instant, the method of reflections can be employed. This

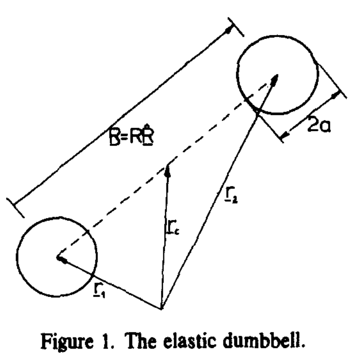

Figure 1. The elastic dumbbell.

method has received considerable attention in the literature and we use the general recursion formulas for the $(j+1)$st reflection in terms of the jth reflection of Brunn (1980). After 3 reflections (the third one only partially in order to get consistent (a/R)-terms) the result reads

$$\dot{\mathbf{r}}_c = \mathbf{V}_c^0 + \frac{1}{8}\mathbf{RR}:\mathbf{\Gamma}_c + \frac{1}{6}a^2\nabla^2\mathbf{V}^0 + \frac{1}{2}\frac{a^3}{R}\{(\hat{\mathbf{R}}\cdot\frac{\partial}{\partial \mathbf{r}_c}\mathbf{\Omega}_c)\times\hat{\mathbf{R}} + \frac{5}{2}\hat{\mathbf{R}}\hat{\mathbf{R}}\hat{\mathbf{R}}\hat{\mathbf{R}}:\frac{\partial}{\partial \mathbf{r}_c}\mathbf{E}_c\}, \qquad [2.3a]$$

$$\mathbf{w} = \mathbf{\Omega}_c + \left[1 - \frac{16}{3}\left(\frac{a}{R}\right)^2\right]\hat{\mathbf{R}}\times(\hat{\mathbf{R}}\cdot\mathbf{E}_c) \qquad [2.3b]$$

$$\dot{R} = R\mathbf{E}_c:\hat{\mathbf{R}}\hat{\mathbf{R}} + \frac{2F^c}{\zeta}\left(1 - \frac{3}{2}\frac{a}{R}\right). \qquad [2.3c]$$

We should point out that in [2.3a] the terms neglected are of order $(a/R)^4$, while for the internal motion, i.e. for [2.3b] and [2.3c] the error terms are of order $(a/R)^3$. The friction coefficient stands for $6\pi\eta a$. It is interesting to see that up to terms of order $(a/R)^3$ the second order velocity gradient (a measure of the inhomogeneity of the flow field) directly affects only the translation and not the rotation-vibration. For homogeneous flows $(\mathbf{\Gamma}_c = 0)$, we have $\dot{\mathbf{r}}_c = \mathbf{V}_c^0$, i.e. the dumbbell moves with the fluid. This, in turn, implies that in homogeneous flows none of the effects talked about in the introduction is possible.

On the other hand a migration relative to the imposed flow field is predicted by [2.3a] for non-homogenous flow fields. The actual value of the migration velocity, $\mathbf{r}_c - V_c^0$, depends upon the instantaneous particle vector $\mathbf{R}$, and the time rate of change of $\mathbf{R}$ itself depends—through $\mathbf{\Omega}_c$ and $\mathbf{E}_c$—upon $\mathbf{r}_c$. In order to see whether a net migration or merely a fluctuation around V_c^0 (without a net-migration) is predicted by [2.3], we first consider [2.3b] and [2.3c] in more detail.

Putting, with respect to an arbitrary cartesian base system (unit vectors $\boldsymbol{\delta}_i$, $i = 1.2.3$)

$$\mathbf{R} = R[\sin\Theta\cos\phi\boldsymbol{\delta}_1 + \sin\Theta\sin\phi\boldsymbol{\delta}_2 + \cos\Theta\boldsymbol{\delta}_3], \qquad [2.4]$$

and realizing that

$$\dot{\mathbf{R}} = \mathbf{w}\times\mathbf{R} + \dot{R}\hat{\mathbf{R}}, \qquad [2.5]$$

we obtain from (2.3b)

$$\dot{\Theta} = \hat{\boldsymbol{\phi}}\cdot\mathbf{\Omega}_c + \left[1 - \frac{16}{3}\left(\frac{a}{R}\right)^2\right]\hat{\mathbf{R}}\hat{\boldsymbol{\Theta}}:\mathbf{E}_c, \qquad [2.6a]$$

$$\sin\Theta\dot{\phi} = -\hat{\boldsymbol{\Theta}}\cdot\mathbf{\Omega}_c + \left[1 - \frac{16}{3}\left(\frac{a}{R}\right)^2\right]\hat{\mathbf{R}}\hat{\boldsymbol{\phi}}:\mathbf{E}_c. \qquad [2.6b]$$

Here the unit vectors $(\hat{\boldsymbol{\Theta}}, \hat{\boldsymbol{\phi}}, \hat{\mathbf{R}})$ are the orthogonal unit vectors for spherical polar coordinates.

So far, essentially no restrictions have been imposed upon the flow field. In order to proceed further we focus attention on viscometric flows, i.e. flows for which the velocity gradient relative to the orthogonal shear axes $\tilde{\boldsymbol{\delta}}_i$ $(i = 1, 2, 3)$ is given by (e.g. Bird *et al.* 1977)

$$\frac{\partial}{\partial \mathbf{r}}\mathbf{V}^0(\mathbf{r})|_{\mathbf{r}_c} = q_c\tilde{\boldsymbol{\delta}}_2\tilde{\boldsymbol{\delta}}_1, \qquad [2.7]$$

with q_c the local shear rate. Associating instantaneously $\tilde{\boldsymbol{\delta}}_i$ with $\boldsymbol{\delta}_i$ $(i = 1, 2, 3)$ furnishes

$$\dot{\Theta} = q_c\left[1 - \frac{16}{3}\left(\frac{a}{R}\right)^2\right]\sin\Theta\cos\Theta\sin\phi\cos\phi, \qquad [2.8a]$$

$$\dot{\phi} = -q_c\left\{\left[1-\frac{8}{3}\left(\frac{a}{R}\right)^2\right]\sin^2\phi + \frac{8}{3}\left(\frac{a}{R}\right)^2\cos^2\phi\right\}, \quad [2.8b]$$

$$\dot{R} = Rq_c\sin^2\Theta\sin\phi\cos\phi + \frac{2F^c}{\xi}\left(1-\frac{3}{2}\frac{a}{R}\right). \quad [2.8c]$$

By [2.8b] the time rate of change of the azimuthal angle ϕ is strictly one-sided (i.e. $\dot{\phi} \lesseqgtr 0$ if $q_c \gtreqless 0$). Thus ϕ uniformly increases or decreases and this implies a rotation of the particle around the $\boldsymbol{\delta}_3$ axis. On the other hand, the polar angle Θ changes according to

$$\frac{d}{d\phi}[\ln\tan\Theta] = -\frac{[1-(16/3)(a/R)^2]\cos\phi\sin\phi}{[1-(8/3)(a/R)^2]\sin^2\phi + (8/3)(a/R)^2\cos^2\phi}. \quad [2.9]$$

This equation, which is independent of the position $\mathbf{r}_c$ can be solved in two cases.

Case 1: *rigid dumbbell*

In this case R does not change and [2.8a] merely serves to determine the connector force F^c. From [2.9] we get

$$\tan\Theta\left[\left(1-\frac{8}{3}\left(\frac{a}{R}\right)^2\right)\sin^2\phi + \frac{8}{3}\left(\frac{a}{R}\right)^2\cos^2\phi\right]^{1/2} = \text{const.} \equiv C. \quad [2.10a]$$

Thus the orientation after one rotation (i.e. for a change of ϕ of 2π) coincides with the initial orientation (the orbit constant C stays constant). We shall see that for the flow fields studied, this implies that $\mathbf{r}_c$ for a rigid dumbbell merely fluctuates periodically across stream lines without any net migration. The period T of rotation is given by

$$\int_0^T dt|q_c| = \sqrt{\left(\frac{3}{2}\right)}\frac{\pi R}{a}. \quad [2.10b]$$

Case 2: *no hydrodynamic interaction*

Without any hydrodynamic interaction the terms (a/R) will all be zero. This leads to the aperiodic behavior

$$\tan\Theta\sin\phi = \text{const.} \quad [2.11]$$

The terminal orientation ultimately attained by the dumbbell is $\phi_\infty = 0$, $\Theta_\infty = (\pi/2)$, i.e. alignment in the flow direction. Thus, although we have always assumed the dumbbell to be large (a/R small), the limit $a/R = 0$ should not be taken: by destroying the periodicity inherent in [2.8a/b] it is totally atypical (and physically meaningless).

In order to make sure that hydrodynamic interaction is small (but non-zero), the beads have to be far apart. Thus, if we consider an elastic dumbbell (spring constant H) of unstretched length L_0, we not only have to require $L_0/a \gg 1$, but we have to make sure that with a finite force the dumbbell cannot be compressed beyond a certain limit. This requires a non-linear force law and we choose

$$F^c = -H\frac{R-L_0}{1-\left(\frac{R-L_0}{L_1-L_0}\right)^2}, \quad 2L_0 - L_1 < R < L_1 \quad [2.12]$$

where L_1 is the length of the fully extended spring. Figure 2 illustrates our choice.

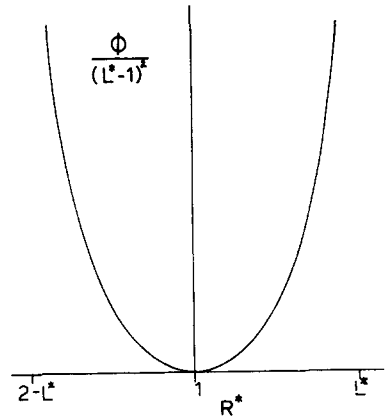

Figure 2. The Interaction potential Φ, $(F^c/HL_0) = -(d\Phi/dR^*)$ for the dumbbell with non-zero unstretched length L_0.

We should point out that various special limits of F^c have successfully been employed in connection with kinetic theory treatments of dissolved macromolecules. For $L_0 \to 0$ and $L_1 \to \infty$ the widely used Hookean dumbbell (Gaussian spring) results. On the other hand, with $L_0 \to 0$ and finite L_1 one obtains the force law characteristic of finitely extendable nonlinear elastic dumbbells (FENE dumbbells), first used for kinetic theory calculations by Warner. Finally, for non-zero L_0 but $L_1 \to \infty$ the Fraenkel dumbbell is recovered, which accounts (crudely) for excluded volume effects (see Bird *et al.* 1977 for details on Warner and all the other various models). Thus, as far as macromolecules are concerned, the force law [2.12] takes excluded volume effects and finite extensibility of the chain into consideration.

As long as the length $2L_0 - L_1$ is larger than the sphere diameter $2a$, hydrodynamic interaction will be small. This then allows us to retain nothing but the dominant (a/R) terms in [2.8] (such that we still have a periodic motion). If q_0 denotes the magnitude of the maximum value of q_c, we introduce dimensionless quantities

$$\tilde{q}_c = q_c/q_0, \quad \tilde{t} = q_0 t, \quad \tilde{\alpha} = \frac{4H}{\zeta q_0},$$
$$R^* = \frac{R}{L_0}, \quad L^* = \frac{L_1}{L_0}, \quad a^* = \frac{a}{L_0}, \tag{2.13}$$

to obtain

$$\frac{d}{d\tilde{t}}\Theta = \tilde{q}_c \sin\Theta\cos\Theta\sin\phi\cos\phi, \tag{2.14a}$$

$$\frac{d}{d\tilde{t}}\phi = -\tilde{q}_c\left[\sin^2\phi + \frac{8}{3}\left(\frac{a^*}{R^*}\right)^2\cos^2\phi\right], \tag{2.14b}$$

$$\frac{d}{d\tilde{t}}R^* = R^*\tilde{q}_c\sin^2\Theta\sin\phi\cos\phi - \frac{1}{2}\tilde{\alpha}\frac{R^*-1}{1-\left(\frac{R^*-1}{L^*-1}\right)^2} \tag{2.14c}$$

The parameter $\tilde{\alpha}$ stands for the ratio of two characteristic time scales: q_0^{-1} is a characteristic time of the flow field (and for the particle rotation) and $\zeta/4H$ is a time constant for the dumbbell characteristic vibration time. The meaning of all other dimensionless quantities is obvious. Note that these equations imply that the time scale for changes in $\mathbf{R}^*$ is governed by the shear rate.

Since q_c depends upon position, we know

$$\mathbf{R}^* = \mathbf{R}^*(\tilde{t}, \tilde{\mathbf{r}}_c; a^*, L^*, \tilde{\alpha}), \tag{2.15}$$

where $\tilde{\mathbf{r}}_c$ is the dimensionless position of the center of mass (scaled with some length B characteristic for the flow).

On a dimensionless basis, some progress can even be made for the migration velocity $\dot{\mathbf{r}}_c$ without specifying the flow field in detail. Since $(\partial/\partial\mathbf{r})\boldsymbol{\Omega}_c$ and $(\partial/\partial\mathbf{r})\mathbf{E}_c$ can all be written as second order derivatives of $\mathbf{V}^0$ and since $\dot{\mathbf{r}}_c - V_c^0$ depend linearly on these derivatives, [2.3a] is of the form

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{r}_c = \mathbf{V}_c^0 + KL_0^2\mathbf{f}(\mathbf{R}^*, a^*), \tag{2.16}$$

where the vector function $\mathbf{f}$ is dimensionless. In this equation K is a measure of the curvature of the flow field, i.e. of the second order derivatives of $\mathbf{V}^0$. Scaling $\mathbf{r}_c$ with a characteristic flow dimension B, [2.16] becomes

$$\frac{\mathrm{d}\tilde{\mathbf{r}}_c}{\mathrm{d}\hat{t}} = \frac{1}{KL_0^2}\mathbf{V}_c^0 + \mathbf{f}(\mathbf{R}^*, a^*), \; \hat{t} = \frac{t}{t_m} \tag{2.17}$$

with

$$t_m = \left(\frac{B}{L_0}\right)^2 \frac{1}{KB}, \tag{2.18}$$

the time scale for migration. Thus, in constrast to the time dependent behavior of the orientation, for which the time scale is governed by the shear rate, the curvature of the flow field and the length of the particle dictate the time scale for migration. Since the particle length enters as $(L_0/B)^2$, it is quite clear that larger particles will be heavily favored in the migration.

For example, for tube flow (B = tube radius, v_m = fluid velocity at tube axis) we take $K = (2v_m/B^2)$ to obtain

$$t_m = \frac{B^3}{2L_0^2 v_m}. \tag{2.19}$$

Thus, a decrease in the time for migration can be achieved by decreasing the tube radius, increasing the size of the particles and increasing the fluid velocity. Reducing B is the most effective means while increasing v_m the least effective means to achieve that. It is interesting that the same conclusion has already been reached by purely thermodynamic arguments for the case of polymer fractionation (Tirrell & Malone 1977).

Since for tube flow we have $q_0 = 2(v_m/B)$, a particle will have undergone many rotations within the time interval t_m. Actually this is valid for any flow since the relation between $\tilde{t}$ and $\hat{t}$ is

$$\hat{t} = \beta^2 \tilde{t},$$

with

$$\beta^2 = \left(\frac{L_0}{B}\right)^2 \frac{KB}{q_0}. \tag{2.20}$$

As long as we regard second order derivatives of the velocity field $\mathbf{V}^0$ as spatially constant, the analysis simplifies considerably, the reason being that in this case q_c will be a linear function of position. Rescaling our variables by putting

$$\mathbf{r}_c{}^* = \frac{1}{\beta}\tilde{\mathbf{r}}_c = \frac{\mathbf{r}_c}{B\beta}, \qquad [2.21a]$$

$$t^* = \beta\tilde{t} = \beta q_0 t, \qquad [2.21b]$$

leaves (2.17) unchanged

$$\frac{d\mathbf{r}_c{}^*}{dt^*} = \frac{1}{L_0{}^2K}\mathbf{V}_c{}^0 + \mathbf{f}(\mathbf{R}^*, a^*), \qquad [2.22]$$

and leads to

$$q_c{}^* = \frac{1}{\beta}\tilde{q}_c. \qquad [2.23]$$

If we then introduce α^* by

$$\alpha^* = \frac{1}{\beta}\tilde{\alpha}, \qquad [2.24]$$

all three equations [2.14a–c] can be used if $\tilde{\alpha}$, $\tilde{q}_c$ and $\tilde{t}$ are replaced by α^*, $q_c{}^*$ and t^*, respectively. Recalling [2.15] we thus see that the function $\mathbf{f}$ of [2.22], which governs the cross-stream migration depends only upon the parameters a^*, L^* and α^* (besides t^* and $\mathbf{r}_c{}^*$). The curvature of the flow field and the size of the particle relative to the size of the system no longer appear explicitly.

3. UNIDIRECTIONAL FLOW

The velocity field for unidirectional motion is given by

$$\mathbf{V}^0(\mathbf{r}) = \{A_0 + A_1x_2 + A_2x_2{}^2\}\boldsymbol{\delta}_1. \qquad [3.1]$$

We shall concentrate on channel flow between two planes at a distance 2B apart. Taking the midplane as the zero of the x_2-coordinate, we get $A_0 = v_m$, the maximum (or centerline) speed, $A_1 = 0$ and $A_2 = -v_m/B^2$. Furthermore, we have $q_0 = 2v_m/B$ and $K = 2v_m/B^2$, i.e. $\beta = L_0/B$. Since no confusion is possible, we drop the subscript c on quantities evaluated at the center and thus write $q_c{}^* = -x_2{}^* = -x_2/L_0$. This expression has to be used in [2.14] for the starred quantities. In order to obtain $x_2{}^*$, [2.3a] has to be solved. Explicitly, it reads in dimensionless form

$$\frac{d}{dt^*}x_2{}^* = \frac{a^{*3}}{4R^*}\sin^2\Theta \sin\phi \cos\phi\,[1 - 5\sin^2\Theta \sin^2\phi]. \qquad [3.2]$$

Thus, in unidirectional flow a migration across stream-lines requires hydrodynamically interacting beads. By [2.14], we know that the azimuthal angle ϕ continuously increases ($x_2{}^* > 0$). This means that the particle axis rotates with period T^*, defined such that ϕ changes by 2π during the time interval T^*.† This being the case, the four coupled equations, [2.14a–c]

†Note that T^* changes during the course of the migration as it depends upon the local shear rate $q_c{}^*$.

and [3.2], have to be solved simultaneously in order to ascertain whether or not a net migration does indeed result.

There is one exception where we know the answer. This concerns a rigid dumbbell where the θ–ϕ relation is known (see [2.10]). With R^* the constant length of the dumbbell, we can combine [3.2] with [2.9] and [2.13b] to obtain an ordinary first order differential equation for $x_2^* = x_2^*(\phi)$ of the form

$$\frac{d}{d\phi} x_2^{*2} = f(\phi).$$

Since $f(\phi)$ so defined has the property $f(2\pi \pm \phi) = \pm f(\phi)$, we immediately see that there is no net migration during one complete rotation (although the dumbbell fluctuates back and forth across a streamline).† Elasticity (or deformability) and hydrodynamic interaction are two prerequisites for any hydrodynamically induced cross-stream migration of dumbbells in nonuniform unidirectional flows. With $R^* = 1$, the period of rotation R^* is thus (see [2.10b])

$$T^* = \sqrt{\left(\frac{3}{2}\right)} \frac{\pi}{a^*|q_c^*|}. \qquad [3.3]$$

For the general case, a numerical integration of [2.14a–c] and [3.2] is in order. Choosing values for the three parameters α^*, L^*, and a^*, the quantities ϕ, θ, R^*, and x_2^* can be evaluated. Although the numerous results obtained depend upon the parameters and upon the initial conditions, they share one thing in common: during the course of one rotation the particle center fluctuates back and forth across a streamline. The eight stationary points of x_2^* are characterized by any multiple of $\pi/2$ and, for the case shown ($\theta = \pi/2$), ϕ being $(n \pm 0.148)\pi$, where n is an integer. The fluctuations are unsymmetric with the final value of x_2^* (called x_{2f}^*) being less than x_{20}^* (the initial value). This implies a net migration towards the channel axis (i.e. towards the region of low shear). Figure 3 shows a typical result. It is interesting to see that the actual period for a rotation is of the same order of magnitude as given by [3.3]. This implies that for a single rotation, the effect of variable q_c^* and R^* on T^* is small.

Since without hydrodynamic interaction ($a^* = 0$) the dumbbell aligns with the flow (aperiodic motion) and does not show any cross-stream migration, we expect a decrease in the migration and an increase in the period of rotation if the interaction parameter a^* is decreased. Figure 4 dramatically illustrates that point.

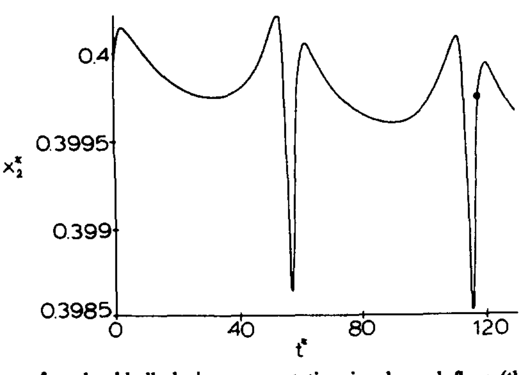

Figure 3. The migration of a dumbbell during one rotation in channel flow (the circles characterize $x_{2f}^* = x_2^*(T^*)$, i.e. the value of x_2^* after one rotation) parameters: $\alpha^* = 0.1$, $a^* = 0.1$, $L^* = 1.8$; initial conditions: $\Theta_0 = \pi/2$; $\phi_0 = (3/4)\pi$; $R_0^* = 1$; $x_{20}^* = 0.4$.

†Rigid particles of revolution lacking fore-aft symmetry would undergo a similar sinuous motion even in homogeneous flows (Brenner 1972).

From figures like these for multiple rotations, it becomes apparent that the initial value of ϕ (called ϕ_0) as well as R_0^* (the initial value of R^*) plays essentially no role at all. On the other hand, θ_0 (i.e. θ at $t^* = 0$) is important: for dumbbells with the initial polar angle ranging from zero to $\pi/2$, the corresponding migration uniformly increases from zero (for $\theta_0 = 0$) to its maximum value (for $\theta_0 = \pi/2$, i.e. dumbbells in the flow-shear plane).† It is due to this reason that explicit results are reported only for $\theta_0 = \pi/2$.

The parameter L^* is a measure of the nonlinearity of the force law. Decreasing L^* (i.e. increasing the nonlinearity) decreases the period of rotation (figure 5: $T^* = 118$ for $L^* = 1.8$ compared to $T^* = 101$ for $L^* = 1.15$). At the same time, the net migration increases and the actual fluctuations decrease. Apparently the reason is that a dumbbell with L^* large passes more slowly (faster) through orientations corresponding to its maximum extension (compression) than one with a smaller L^*. And for larger separation of the beads the hydrodynamic interaction is very small, an interaction which is needed in unidirectional flows for any net migration to occur.

The influence of the parameter α^* is shown in figure 6. While the period T^* of rotation uniformly decreases with increasing α^*, the net migration does not show such a one-to-one correspondence. For the three cases studied, the migration is least for $\alpha^* = 10$, largest for $\alpha^* = 1$, and somewhere in between for $\alpha^* = 0.1$. Such results can be comprehended if we look at the limits $\alpha^* \to 0$ and $\alpha^* \to \infty$, respectively. For $\alpha^* \to \infty$ (e.g. very weak flows), we need $R^* \to 1$

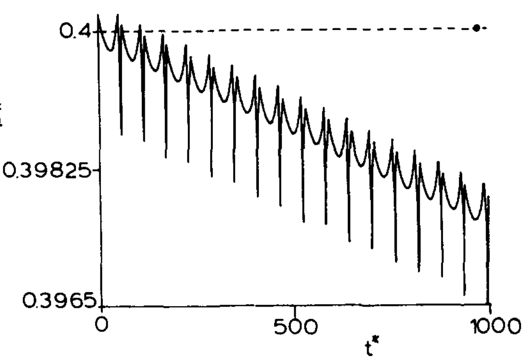

Figure 4. The dependence of migration on the inverse aspect ratio a^* (The period $T^* = 976$ for dumbbells with $a^* = 0.01$ as indicated by the circle), parameters: $\alpha^* = 0.1$, $L^* = 1.8$; same initial conditions as in figure 3, (a) solid: $a^* = 0.1$; (b) dashed: $a^* = 0.01$.

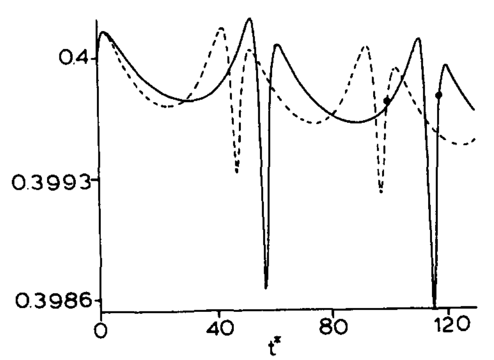

Figure 5. The dependence of migration on L^* (the circle indicates one full rotation), parameters: $\alpha^* = 0.1$, $a^* = 0.1$; same initial conditions as before, (a) solid line: $L^* = 1.8$; (b) dashed: $L^* = 1.15$ ($T^* = 10.1$).

†Note also that for $\theta < 0.148\pi$, there are only four stationary points in the $x_2^* = x_2^*(t^*)$ curve during one rotation.

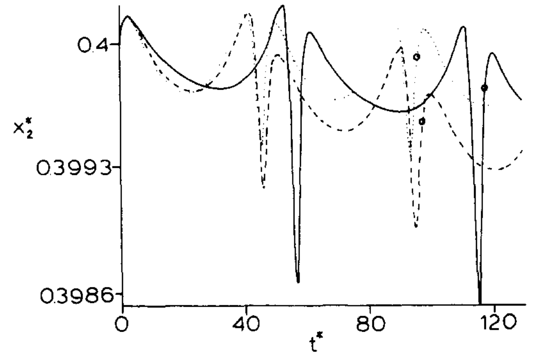

Figure 6. The dependence of migration on α^* (each circle characterizes one full rotation for the corresponding dumbbell), parameters: $a^* = 0.1$, $L^* = 1.8$; same initial conditions as before; (a) solid: $\alpha^* = 0.1$ ($T^* = 118$); (b) dashed: $\alpha^* = 1$ ($T^* = 98$); (c) dotted: $\alpha^* = 10$ ($T^* = 96$).

(corresponding to freezing the spring into a rod). No net migration is possible in that case. On the other hand, for $\alpha^* \to 0$ (e.g. very strong flows), the dumbbell rotates too fast for the spring to respond. Again, no net migration can result. Thus, the migration will be small for very small and for very large α^* (i.e. when the elastic response time of the dumbbell and the characteristic time for rotation (inverse shear rate) differ drastically).

Having established the dependence of the parameters on the cross-stream migration, we finally look at an actual particle trajectory. For the channel flow under consideration, $\mathrm{d}x_1^*/\mathrm{d}t^*$ increases with decreasing x_2^*. Consequently, $\mathrm{d}x_2^*/\mathrm{d}x_1^*$ decreases with decreasing x_2^* (i.e. the net migration (as a function of axial distance travelled) decreases with decreasing distance from the axis (see figure 7)). Physically, this was to be expected. Recalling the remarks made at the end of the last section, it is not surprising that on the scale at which appreciable cross-stream migration takes place, the fluctuations do not show up at all.

4. CIRCULAR COUETTE FLOW

For circular Couette flow, i.e. the flow between two concentric cylinders with radii R_1 and R_2, one of them steadily rotating with angular velocity Ω relative to the other, we have

$$\mathbf{V}^0 = \rho\omega(\rho)\boldsymbol{\delta}_\varphi. \tag{4.1}$$

Here ρ is the radial coordinate measured from the common axis and ω is given by

$$\omega(\rho) = A\left[\left(\frac{R_2}{\rho}\right)^2 - 1\right], \tag{4.2a}$$

$$A = \frac{\Omega R_1^2}{R_2^2 - R_1^2}. \tag{4.2b}$$

We shall assume that the inner cylinder is rotating counterclockwise relative to the outer one ($\Omega > 0$), so that A is a positive constant.

The shear axes are related to the cylindrical coordinate axes by

$$\tilde{\boldsymbol{\delta}}_1 = \boldsymbol{\delta}_\varphi, \quad \tilde{\boldsymbol{\delta}}_2 = \boldsymbol{\delta}_\rho, \quad \tilde{\boldsymbol{\delta}}_3 = -\boldsymbol{\delta}_z,$$

and relative to these axes the local shear rate is (Bird *et al.* 1977)

$$q_c = \rho\frac{\mathrm{d}\omega}{\mathrm{d}\rho} = -2\frac{AR_2^2}{\rho^2}, \tag{4.3}$$

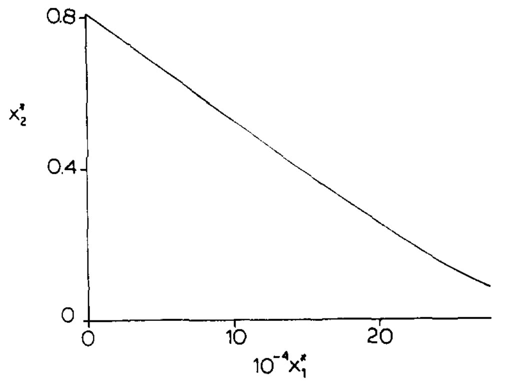

Figure 7. The actual particle trajectory for $L_0/B = 1/2$, parameters: $\alpha^* = 0.05$, $(a^* = 0.1)$, $L^* = 1.8$.

i.e.

$$q_0 = 2A\left(\frac{R_2}{R_1}\right)^2. \qquad [4.4]$$

Equations (2.14) require that we know ρ, the radial coordinate of the center of mass, $\mathbf{r}_c$. By [2.3a], ρ satifies

$$\frac{d\rho}{dt} = -\frac{1}{4}R^2\frac{d\omega}{d\rho}\sin^2\Theta\sin\phi\cos\phi. \qquad [4.5]$$

This being the case we put

$$r^* = \frac{\rho}{L_0}, \quad q_c{}^* = \left(\frac{L_0}{R_1}\right)^2\tilde{q}_c = -\frac{1}{r^{*2}},$$

$$t^* = \left(\frac{R_1}{L_0}\right)^2 q_0 t, \quad \alpha^* = \left(\frac{L_0}{R_1}\right)^2\tilde{\alpha}. \qquad [4.6]$$

This enables us to use [2.14] (for the starred quantities) and to supplement this set of equations by the equation for r^*,

$$\frac{d}{dt^*}r^{*2} = \frac{1}{2}\left(\frac{R^*}{r^*}\right)^2\sin^2\Theta\sin\phi\cos\phi. \qquad [4.7]$$

Since [4.7] is independent of a^*, hydrodynamic interaction does not directly enter into the expression for the migration velocity. This, however, does not imply that hydrodynamic interaction can be neglected altogether. As emphasized previously, the particle rotation does depend upon this interaction. If it is neglected altogether the resulting terminal orientation of the dumbbell ($\phi_\infty = 0, \Theta_\infty = (\pi/2)$) is such that no cross-stream migration can result. This is in striking contrast to kinetic theory treatments for this type of flow (Shafer *et al.* 1974, Bird 1979, Aubert *et al.* 1980). Dealing exclusively with a Gaussian spring and neglecting the finite size of the spheres (i.e. modeling them as volumeless friction centers, and thus using $a^* = 0$) these authors end up with a cross-stream migration in circular Couette flow. Actually the reason is not hard to see. With Brownian motion included the forces and couples of [2.2] are non-zero, but balanced by Brownian motion forces and couples. This immediately implies that the $a^* = 0$ dumbbell does not align with the flow and this, by [4.5], necessitates a non-zero migration velocity.

In our purely hydrodynamic treatment we thus see that $a^* \neq 0$ is still vital for any cross-stream migration to occur. But even for $a^* \neq 0$ it is important that the spring is elastic. For a rigid dumbbell it is easy to prove, along the same line of reasoning used in section 3, that no net migration is possible during the course of one rotation.

In order to evaluate $r^* = r^*(t^*)$ numerically for elastic springs, it is important to recall that r^*, as defined by [4.6], is defined as the distance of the center of the particle from the axis of the cylinders relative to the unstretched length of the dumbbell. We shall report results only for $r_0^* = 120$, which, for $R_1 = 2.3$ cm, $R_2 = 2.5$ cm (the system of Shafer *et al.* 1974) means that a dumbbell with $L_0^* = 0.02$ cm is initially halfway between the cylinders. This implies, by [4.6], that the local shear rate q_c^* is of the order of 7×10^{-5}. If [3.3] is used to obtain a (rough) estimate for the period of rotation then $T^* \sim 5.54 \times 10^4 (a^*)^{-1}$. Choosing $a^* = 0.1$ and $a^* = 0.01$, respectively, leads to periods of rotation so large that we found it advantageous not to calculate $r^* = r^*(t^*)$ but rather $r^* = r^*(\phi)$ (ϕ increases by 2π during the time interval T^*).

This kind of representation requires some care to put the resulting curves into proper perspective. Thus, while for elastic dumbbells the migration always is towards the inner cylinder (see figures 8–10), i.e. towards the high shear region, a comparison of figures 8(a) with 8(b) shows that during the course of one rotation, it is the longer dumbbell which has migrated most. Only by realizing that the period of rotation varies approximately linearly with the particle aspect ratio a^{*-1} does it become clear that during equal time intervals the actual migration decreases with decreasing a^*. On an order of magnitude basis, this effect is far less dramatic than in the channel flow situation (see figure 4). This was to be expected since for circular Couette flow the migration velocity does not directly depend upon the particle aspect ratio, while for channel flow it is proportional to a^{*3}.

From the channel flow results we also see that the period of rotation decreases with decreasing L^* (figure 6). This will to some extent counterbalance the decrease in the migration with decreasing L^* during one rotation (figure 9).

Finally, the effect of the parameter α^* is shown in figure 10. As found in channel flow, the migration is largest if α^* is of the same order of magnitude as q_c^*, i.e. when the elastic response time of the dumbbell and the characteristic time of rotation are of the same order.

5. DISCUSSION

Elastic dumbbells in non-homogeneous flows have been studied. Concentrating solely upon the hydrodynamics of that problem we found deformability and hydrodynamic interaction an indispensible prerequisite for any net-migration to take place in viscometric flows. Without interaction the orientation of the dumbbell changes aperiodically such that in the terminal

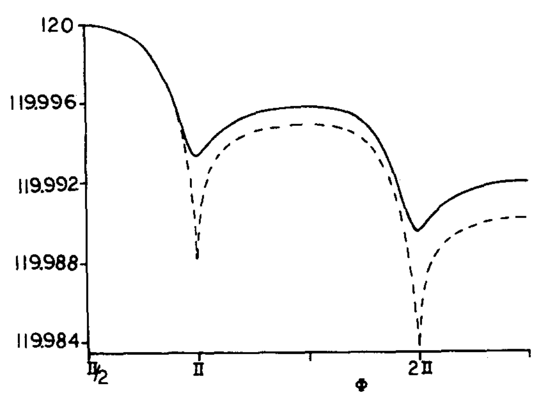

Figure 8. The dependence of migration on a^* during one rotation in circular Couette flow, parameters: $\alpha^* = 1.4 \times 10^{-5}$, $L^* = 1.6$; initial conditions: $\Theta_0 = \pi/2$, $\phi_0 = \pi/2$, $R_0^* = 1$, $r_0^* = 120$, (a) solid line: $a^* = 0.1$; (b) dashed line: $a^* = 0.01$.

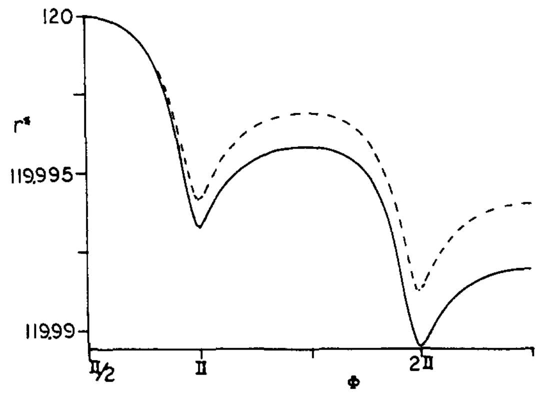

Figure 9. The dependence of migration on L^* during one rotation parameters: $\alpha^* = 1.4 \times 10^{-5}$; $a^* = 0.1$; same initial conditions as in figure 8. (a) solid line: $L^* = 1.6$; (b) dashed line: $L^* = 1.3$.

orientation attained no migration is possible. With hydrodynamic interaction the dumbbell orientation changes periodically and in this case a net migration towards the high shear region results in circular Couette flow. On the other hand, in channel flow (and in tube flow) the hydrodynamic interaction enters not only into the expression for the change in orientation but also into the expression for the change of position. A migration towards the low shear region is the consequence. In contrast to the hydrodynamic migration of droplets, which attain a steady orientation relative to the streamlines (e.g. Chan *et al.* 1979), the dumbbells always rotate. Since the details of the migration depend upon the instantaneous length and orientation of the particle it is clear that any attempt to correlate our predictions with experimental results requires that we know the probability to find a dumbbell at time t in some orientation-and-length range. This, however, is the subject of a statistical-mechanical treatment, a treatment which will be the subject of a separate study. Thus, all we can do for now is to speculate whether the migration phenomena can in principle lead to observable effects.

On an order of magnitude basis the residence time for a particle in tube (or channel) flow is

$$t_R = \frac{2l}{V_m} \qquad [5.1]$$

where l is the length of the tube. Since, the time scale for migration is†

$$t_m = \frac{B^3}{2L_0^2 v_m a^{*3}}, \qquad [5.2]$$

it is clear that no effects are possible unless the restriction

$$\frac{t_m}{t_R} = \frac{1}{4}\left(\frac{B}{l}\right)\left(\frac{B}{L_0}\right)^2\left(\frac{L_0}{a}\right)^3 \ll 1 \qquad [5.3]$$

is met. We expect this condition to be violated, since within the limits of and approximations of our theory, $a^* \ll 1$ and $(L_0/B) \ll 1$.

Although disappointing there may be situations of practical significance where an observable phenomenon could result. Since the net migration cannot depend upon the direction of flow, a periodically varying pressure difference should, if applied long enough, lead to an observable effect.

†Note that in channel and tube flow we have by (2.16) and (3.2) $\mathbf{f}(\mathbf{R}^*, a^*) = a^{*3}\mathbf{f}(\mathbf{R}^*)$.

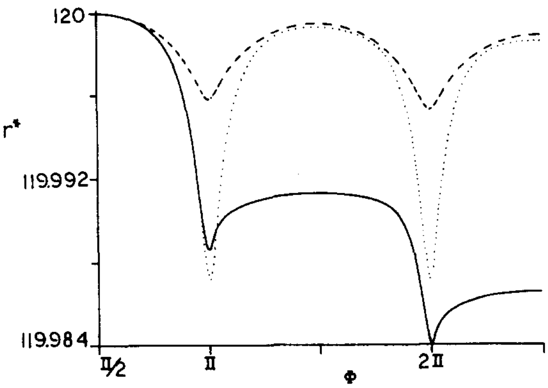

Figure 10. The dependence of migration on α^* during one rotation parameters: $a^* = 0.1$, $L^* = 1.6$; same initial conditions as in figure 9. (a) dashed line: $\alpha^* = 1.4 \times 10^{-3}$; (b) solid line: $\alpha^* = 1.4 \times 10^{-5}$; (c) dotted line: $\alpha^* = 1.4 \times 10^{-7}$.

Similarly, for the circular Couette flow time restrictions (such as (5.3)) play no role. In this case we directly turn to polymer concentration $c(\rho, t)$ which satisfies the conservation equation

$$\frac{\partial}{\partial t} c + v_\perp \frac{\partial c}{\partial \rho} = -\frac{c}{\rho} \frac{\partial}{\partial \rho} (\rho v_\perp), \qquad [5.4]$$

with

$$v_\perp = \frac{AR_2^2}{2} \frac{1}{\rho^3} \langle R^2 \sin^2 \Theta \sin \phi \cos \phi \rangle, \qquad [5.5]$$

the ensemble average of the migration velocity ($d\rho/dt$). Assuming now that outside a particular radius ρ_c the concentration is zero the motion of that outside boundary must move along the characteristic curve ($\rho_0 = \rho_c(t = 0)$)

$$t = \int_{\rho_0}^{\rho_c} \frac{d\rho}{v_\perp}. \qquad [5.6]$$

For example, taking $\rho_0 = R_2$ the time t_c (termed clearance time) required for all the molecules to have migrated to the inner cylinder is given by

$$t_c = \int_{R_2}^{R_1} \frac{d\rho}{v_\perp}. \qquad [5.7]$$

In order to estimate this clearance time the function $v_\perp$ has to be known. Unfortunately, this again requires a rigorous statistical-mechanical treatment. Without it any expression for $v_\perp$ must be taken as a conjecture. One possible choice is

$$\langle R^2 \sin^2 \Theta \sin \phi \cos \phi \rangle = \text{const. } L_0^2 \bar{q}_c, \qquad [5.8a]$$

i.e.

$$v_\perp = -\frac{\text{const. } AR_1^2 R_2^2 L_0^2}{2} \frac{1}{\rho^3} \equiv -\frac{\beta}{\rho^3}. \qquad [5.8b]$$

This choice is motivated by three facts: (a) The ensemble average [5.8a] vanishes in equilibrium ($q_c = 0$). (b) by using the distribution function

$$\Psi_{eq.}\left[1+\frac{\zeta}{4kT}\,q_c R^2 \sin^2\Theta \sin\phi\cos\phi + 0(\tilde{q}_c^2)\right], \qquad [5.9]$$

with

$$\Psi_{eq.} = \frac{1}{J}\left[1-\left(\frac{R-L_0}{L_1-L_0}\right)^2\right]^{(H(L_1-L_0)^2)/2kT}$$

and

$$J = 4\pi\int_{2L_0-L_1}^{L_1} \mathrm{d}\,RR^2\left[1-\left(\frac{R-L_0}{L_1-L_0}\right)^2\right]^{(H(L_1-L_0)^2)/2kT}, \qquad [5.10]$$

which, for slow flow conditions and q_c independent of position is valid for any spring force law (Bird *et al.* 1977),† [5.8a] results. In this case, the constant of [5.8a] turns out to be

$$\text{const} = \frac{AR_2^2\zeta}{30kTR_1^2L_0^2}\langle R^4\rangle_{eq.} \quad . \qquad [5.11]$$

(c) From a statistical-mechanical treatment for Rouse coils and for Zimm coils

$$v_\perp = -\frac{\tilde{\beta}}{\rho^3},\ \tilde{\beta} = \text{const}. \qquad [5.12]$$

follows, irrespective of the strength of the flow field (Shafer *et al.* 1974). Note that the constant $\tilde{\beta}$ does depend upon the actual choice of the model.

Thus, we feel that [5.8b] suffices for illustrative purposes. Using this expression in [5.7] leads to the clearance time:

$$t_c = -\frac{1}{24}\frac{(R_2^6-R_1^6)(R_2^2+R_1^2)(R_2+R_1)}{R_1^4R_2^4\bar{v}_\perp} \qquad [5.13]$$

where $\bar{v}_\perp$ denotes the average of $v_\perp$ over the annular gap. For the values reported by Shafer *et al.* (1974), $R_1 = 2.3$ cm, $R_2 = 2.5$ cm together with their calculated value $\bar{v}_\perp = -3\times 10^4$ cm/sec we thus get:

$$t_c = 11.27\ \text{min} \qquad [5.14]$$

The observed clearance time could differ significantly from this estimate (Shafer *et al.* (1974) do not report t_c), the reason being that for the spring force [2.12], [5.8b] for $v_\perp$ should only be used for slow flow conditions, i.e. $\tilde{\alpha} \gg 1$. This being the case, Brownian motion most certainly cannot be neglected. Thus, we expect any observed clearance time to exceed t_c as given by [5.13]. For the extreme case, that Brownian motion is so strong as to balance the inward radial convective flux steady state concentration profiles can be established. Aubert *et al.* (1980) studied exactly this case and reported theoretical results for a Gaussian chain.

†More accurately, we should say that up to order $\tilde{q}_c^2$ only the equilibrium function ψ_{eq} depends upon the connector potential. This implies that the equilibrium average $\langle R^4\rangle_{eq}$ will be of the form $\langle R^4\rangle_{eq} = L_0^4(L^*,b)$ with $b = H(L_1-L_0)^2/2kT$.

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