RHEOLOGICAL EQUATIONS OF STATE OF WEAKLY CONCENTRATED SUSPENSIONS OF RIGID ELLIPSOIDAL PARTICLES

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Rheological equations of state of dilute suspensions of rigid ellipsoidal particles (ellipsoids of revolution) are derived [1-4] from the vantage point of the structural-continuum approach, with attention given both to rotational Brownian motion of particles and to their inertia and the outer force fields. Interaction between particles is ignored in those treatments given the low concentration of the suspended particles. In this paper, the earlier findings [1-4] are generalized to higher concentrations. The effect of hydrodynamical interaction between particles on the rheological behavior of the suspension is treated in the light of the Simha approach [5].

We resort to the structural-continuum model [6, 7] with fixed length of orientation vector n_i

$$t_{ij} = (a_0 + a_1 d_{km} n_k n_m)\,\delta_{ij} + a_2 n_i n_j + a_3 d_{km} n_k n_m n_i n_j + a_4 d_{ij} + a_5 d_{ik} n_k n_j + a_6 d_{jk} n_k n_i + a_7 n_i N_j + a_8 n_j N_i \tag{1}$$

$$\varepsilon(\ddot{n}_i + \dot{n}_k \dot{n}_k n_i) = \gamma\,[N_i - \lambda(d_{ij} n_j - d_{jk} n_j n_k n_i)] + \varepsilon_{ijk} M_j n_k \tag{2}$$

in deriving the rheological equations of state of weakly concentrated suspensions of rigid ellipsoidal particles.

Here t_{ij} is the stress tensor, d_{ij} is the strain-rate tensor, $N_i = \dot{n}_i - \omega_{ij} n_j$, ω_{ij} is the velocity vortex tensor, M_j is the moment of the forces acting upon an element of the substructure; $\alpha_i, \varepsilon, \lambda, \gamma$ are rheological constants, and δ_{ij}, ε_{ij} are the symmetric and skew-symmetric Kronecker symbols.

We take as our orientation vector n_i the unit vector oriented on the rotation axis of the ellipsoidal particle. Then the rheological constants figuring in Eqs. (1) and (2) can be determined for any case by generalizing Jeffery's results [8] with the aid of the Simha approach [5].

In accordance with [5], we consider a "constricted" flow of an ellipsoidal particle within a sphere whose center coincides with the center of the particle, and whose radius $R = (a b^2/V)^{1/3}$, where $2a$ and b are the rotation axis and the equatorial radius of the particle, respectively, and V is the volume concentration of suspended particles. The solution of the actual hydrodynamical problem in the Stokes approximation, satisfying the condition of "sticking" on the surface of the particle and the condition of velocity perturbations vanishing on the surface of the sphere in question, will be sought by the method of successive approximations.

As our first approximation we take the Jeffery solution [8] which, in a moving frame of reference x_i with origin at the center of the particle and axes coinciding in direction with the directed principal axes of the ellipsoidal particle, exhibits the form

$$u_i = u_{0i} + \frac{4}{3}\frac{R^3 - r^3}{R^3 r^3}(A_{ki} - A_{ik})\,x_k - 4x_i \frac{R^5 - r^5}{R^5 r^5}\Phi_0 + 5\frac{R^2 - r^2}{R^5}\frac{\partial \Phi_0}{\partial x_i} \tag{3}$$
$$p = p_0 - \frac{8\mu}{r^5}\Phi_0 - \frac{42\mu}{R^5}\Phi_0$$

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where u_i is the velocity, u_{0i} is the velocity of the unperturbed stream; p is the pressure; p_0 is the pressure in the unperturbed stream; r is the modulus of the radius vector; μ is the dynamical viscosity of the solvent; A_{ki} are quantities defined in [2]; $\Phi_0 = A_{pq}x_p x_q$. This solution accurately satisfies the boundary conditions on the surface of the sphere; the velocity perturbation on the surface of the ellipsoid does not exceed a quantity of the order $O(R^{-3})$.

We now find the second approximation in the problem, such that the velocity perturbation on the surface of the ellipsoid will not be greater than a quantity of the order $O(R^{-6})$. For that, we add to Eq. (3) the following solution obtained by the method employed in [8]:

$$u_i^* = \frac{4}{3}\frac{1}{r^3}(B_{ki} - B_{ik})x_k - \frac{4x_i}{r^5}\Phi + U_i, \qquad p^* = P - \frac{8\mu}{r^5}\Phi \tag{4}$$

Where $\Phi = B_{pq}x_p x_q$

$$B_{11} = \frac{5d_{11}}{18\beta_0''^2R^3}$$
$$B_{22} = \frac{5d_{22}}{8b^4\alpha_0'^2R^3} + \frac{5(\beta_0'' - \alpha_0'')(2b^2\alpha_0' + 3\beta_0'')d_{11}}{72b^4\alpha_0'^2\beta_0''^2R^3} \tag{5}$$
$$B_{33} = \frac{5d_{33}}{8b^4\alpha_0'^2R^3} + \frac{5(\beta_0'' - \alpha_0'')(2b^2\alpha_0' + 3\beta_0'')d_{11}}{72b^4\alpha_0'^2\beta_0''^2R^3}$$
$$B_{12} = \{[15\alpha_0(\alpha_0 + \beta_0) - 4b^2\beta_0'(\beta_0 - \alpha_0)]d_{12} + [4b^2(a^2 + b^2)\beta_0'^2 - 15(a^2 - b^2)\alpha_0\beta_0'](\omega_{12} + \omega_3)\} / 12\beta_0'^2B^2R^3$$
$$B_{21} = \{[15\beta_0(\alpha_0 + \beta_0) + 4a^2\beta_0'(\beta_0 - \alpha_0)]d_{21} + [4a^2(a^2 + b^2)\beta_0'^2 + 15(a^2 - b^2)\beta_0\beta_0'](\omega_{21} - \omega_3)\} / 12\beta_0'^2B^2R^3$$
$$B_{13} = \{[15\alpha_0(\alpha_0 + \beta_0) - 4b^2\beta_0'(\beta_0' - \alpha_0)]d_{13} + [4b^2(a^2 + b^2)\beta_0'^2 - 15(a^2 - b^2)\alpha_0\beta_0'](\omega_{13} - \omega_2)\} / 12\beta_0'^2B^2R^3$$
$$B_{31} = \{[15\beta_0(\alpha_0 + \beta_0) + 4a^2\beta_0'(\beta_0 - \alpha_0)]d_{31} + [4a^2(a^2 + b^2)\beta_0'^2 + 15(a^2 - b^2)\beta_0\beta_0'](\omega_{31} + \omega_2)\} / 12\beta_0'^2B^2R^3$$
$$B_{23} = \frac{5d_{23}}{8b^4\alpha_0'^2R^3}, \qquad B_{32} = \frac{5d_{32}}{8b^4\alpha_0'^2R^3}$$

$B = a^2\alpha_0 + b^2\beta_0$, α_0, β_0, α_0', β_0', α_0'', β_0'' are functions of the ratio a/b defined in [8]; ω_i is the angular velocity of the particle, U_i and P are the next terms in the expansion of the Jeffery solution having quantities of the order $U_i \sim O(R^{-4})$ and $P \sim O(R^{-5})$ on the surface of the sphere.

Addition of the solution (4) to equations (3) makes it possible to satisfy the boundary conditions on the surface of the ellipsoid exactly, but the solution so constructed fails to satisfy the boundary conditions on the surface of the sphere.

Once one other particular solution useful in satisfying the boundary conditions on the spherical surface exactly has been found, together with the solutions (3) and (4), we arrive at the definitive solution of the problem in the second approximation

$$u_i = u_{0i} + \frac{4}{3}\frac{R^3 - r^3}{R^3r^3}(A_{ki} + B_{ki} - A_{ik} - B_{ik})x_k - 4x_i\frac{R^5 - r^5}{R^5r^5}(\Phi_0 + \Phi) + 5\frac{R^2 - r^2}{R^5}\frac{\partial(\Phi_0 + \Phi)}{\partial x_i} + U_i + U_i' \tag{6}$$
$$p = p_0 - \frac{8\mu}{r^5}(\Phi_0 + \Phi) - \frac{42\mu}{R^5}(\Phi_0 + \Phi) + P + P'$$

where U_i', P' is the particular solution of the problem satisfying the following boundary condition on the surface of the sphere: $U_i' = -U_i$. Given the cumbersomeness of the expression, U_i, U_i', P, and P' are not calculated out. As the investigation shows, these expressions make no contribution to the averaged stress tensor constructed below for the problem in question.

The stress tensor σ_{ij} for the medium [9] can be found from the velocity and pressure perturbation in the moving system of coordinates x_i. By averaging the stress tensor determined by the solution (6) over the volume of the sphere, and proceeding from integration over the volume to integration over the surface of the sphere [9, 10, 2] we get

$$\sigma_{ij} = -p_0\delta_{ij} + 2\mu d_{ij} + \frac{8\mu V}{ab^2}(A_{ij} + B_{ij}) \tag{7}$$

The orientation equations for an ellipsoidal particle, when the moment of inertia about the rotation axis of the particle (prolate ellipsoids) is neglected appear in the moving system of coordinates x_i in the form

$$0 = M_1^* + M_1^0, \quad I(\dot{\omega}_2 - \omega_1\omega_3) = M_2^* + M_2^0 \tag{8}$$
$$I(\dot{\omega}_3 + \omega_1\omega_2) = M_3^* + M_3^0$$

where I is the moment of inertia of the ellipsoid about the axis lying in the equatorial plane, M_i^0 is the moment of the external forces, and M_i^* is the moment of the hydrodynamical forces whose components, in the case in point, exhibit the form

$$
\begin{aligned}
M_1^* &= -\frac{16\pi\mu}{3\beta_0}\left(1+\frac{2V}{3ab^2\beta_0}\right)(\omega_{23}+\omega_1)\\
M_2^* &= -\frac{32\pi\mu}{3}(A_{31}+B_{31}-A_{13}-B_{13})\\
M_3^* &= -\frac{32\pi\mu}{3}(A_{12}+B_{12}-A_{21}-B_{21})
\end{aligned}
\tag{9}
$$

Considering Eqs. (1) and (2) in the moving system of coordinates x_i($n_1 = 1$, $n_2 = n_3 = 0$, $\dot{n}_1 = 0$, $\dot{n}_2 = \omega_3$, $\dot{n}_3 = -\omega_2$, $\ddot{n}_1 = -\omega_2^2 - \omega_3^2$, $\ddot{n}_2 = \dot{\omega}_3 + \omega_2\omega_1$, $\ddot{n}_3 = -\dot{\omega}_2 + \omega_3\omega_1$), and comparing Eqs. (1) with (7), and (2) with (8), (9), we find the rheological constants appearing in Eqs. (1), (2), $\varepsilon = 1$

$$
\begin{aligned}
\gamma &= -\frac{16\pi\mu}{3B}\left[(a^2+b^2)+\frac{15(a^2-b^2)^2+4(a^2+b^2)^2}{6ab^2B}V\right]\\
\lambda &= \left[a^2-b^2+\frac{15(a^2-b^2)(\alpha_0+\beta_0)+4(a^2+b^2)(\beta_0-\alpha_0)}{6ab^2\beta_0'B}V\right]\left[a^2+b^2+\frac{15(a^2-b^2)^2+4(a^2+b^2)^2}{6ab^2B}V\right]^{-1}\\
a_0 &= -p_0, \qquad a_2 = 0\\
a_1 &= \frac{2\mu V(\beta_0''-\alpha_0'')}{3ab^4\beta_0''\alpha_0'}+\frac{5\mu V^2(\beta_0''-\alpha_0'')(2b^2\alpha_0'+3\beta_0'')}{9a^2b^8\beta_0''^2\alpha_0'^2}\\
a_3 &= \frac{2\mu V}{ab^2}\left[\frac{\alpha_0''+\beta_0''}{b^2\alpha_0'\beta_0''}-\frac{2(\alpha_0+\beta_0)}{\beta_0'B}\right]+\frac{2\mu V^2}{3a^2b^4}\left[\frac{5(\beta_0''^2+\beta_0''\alpha_0''+\alpha_0''^2)}{b^4\alpha_0'^2\beta_0''^2}-\frac{15(\alpha_0+\beta_0)^2+4\beta_0'^2(a^2-b^2)^2}{\beta_0'^2B^2}\right]\\
a_4 &= 2\mu\left(1+\frac{V}{ab^4\alpha_0'}+\frac{5V^2}{2a^2b^8\alpha_0'}\right)\\
a_5 &= \frac{4\mu V}{ab^2}\left(\frac{\beta_0}{\beta_0'B}-\frac{1}{2b^2\alpha_0}\right)+\frac{2\mu V^2}{a^2b^4}\left[\frac{15\beta_0(\alpha_0+\beta_0)+4a^2\beta_0'(\beta_0-\alpha_0)}{3\beta_0'^2B^2}-\frac{5}{2b^4\alpha_0'^2}\right]\\
a_6 &= \frac{4\mu V}{ab^2}\left(\frac{\alpha_0}{\beta_0'B}-\frac{1}{2b^2\alpha_0'}\right)+\frac{2\mu V^2}{a^2b^4}\left[\frac{15\alpha_0(\alpha_0+\beta_0)-4b^2\beta_0'(\beta_0-\alpha_0)}{3\beta_0'^2B^2}-\frac{5}{2b^4\alpha_0'^2}\right]\\
a_7 &= \frac{4b^2\mu V}{ab^2B}+\frac{2\mu V^2[4b^2(a^2+b^2)\beta_0'-15(a^2-b^2)\alpha_0]}{3a^2b^4\beta_0'B^2}\\
a_8 &= -\frac{4a^2\mu V}{ab^2B}-\frac{2\mu V^2[4a^2(a^2+b^2)\beta_0'+15(a^2-\beta^2)\beta_0]}{3a^2b^4\beta_0'B^2}
\end{aligned}
\tag{10}
$$

Since the orientation vector characterizes the behavior of the substructure (microparticle, macromolecule), upon constructing the rheological equations of state we have to perform an averaging process in Eq. (1) with the aid of the distribution function of the angular positions of the rotation axis of the suspended particle F which, together with determinate forces acting on the particle (hydrodynamical forces, external force fields), can be taken under consideration, and also forces due to the rotational Brownian motion. As shown in [3], we have to introduce into Eq. (2), in M_j, the moment

$$
M_j^0 = -kT\varepsilon_{jik}n_i\frac{\partial \ln F}{\partial x_k}
$$

where k is Boltzmann's constant, and T is the absolute temperature.

Accordingly, we take, as the rheological equations of state of weakly concentrated suspensions of rigid ellipsoidal particles, Eq. (1) with coefficients a_i related to the parameters which characterize the geometry and concentration of the suspended particles by equations (10); Equation (1) is averaged with the aid of the distribution function F satisfying the equation

$$
\partial F/\partial t = \mathrm{Dr}\Delta F - \mathrm{div}(F\omega)
\tag{11}
$$

where t is the time, $\mathrm{Dr} = -kT/\gamma$, ω is the angular velocity vector of the particle determined from Eq. (2),

$$
\begin{aligned}
T_{ij} = \langle t_{ij}\rangle &= (a_0 + a_1 d_{km}\langle n_k n_m\rangle)\delta_{ij} + a_3 d_{km}\langle n_k n_m n_i n_j\rangle +\\
&+ a_4 d_{ij} + a_5 d_{ik}\langle n_k n_j\rangle + a_6 d_{jk}\langle n_k n_i\rangle + a_7\langle n_i N_j\rangle + a_8\langle n_j N_i\rangle
\end{aligned}
\tag{12}
$$

Since γ was obtained with "constricted" flow taken into account, $\mathrm{Dr} = -kT/\gamma$ takes into account the effect of the hydrodynamical interaction of suspended particles on the rotational Brownian motion of the particles.

We note in summary that, when we take the hydrodynamical interaction of the suspended particles into account on the basis of the procedure outlined in the article, the rheological equations of state of the weakly concentrated suspensions of ellipsoidal particles coincide in form with the equations of state of dilute suspensions of ellipsoidal particles. The hydrodynamical interaction between the suspended particles is manifested in the changes experienced by the distribution function F and by the rheological constants.

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