PMM U.S.S.R.,Vol.46,pp.58-65 0021-8928/83/1 0058 $7.50/0

UDC 532.529

CALCULATION OF THE EFFECTIVENESS OF GRAVITATIONAL COAGULATION OF DROPS WITH ALLOWANCE FOR INTERNAL CIRCULATION*

A.Z. ZINCHENKO

The relative trajectories of two liquid spherical particles of different radii moving in a viscous medium under the action of gravitational and Archimedean forces are considered in the domain of the quasi-stationary Stokes equation applicability. The effective capture cross section is determined using exact methods for calculating hydrodynamic forces.

Published results of calculations of effective coagulation of drops in emulsions were obtained using the solid spheres model /1,2/. Since in the Stokes flow convergence of solid particles under the action of finite forces in a finite time interval is impossible, additional forces of nonhydrodynamic interaction (electric or molecular), peculiar to contacting spheres were introduced for explaining coagulation. However such forces and even their order of magnitude are seldom known. It is shown in the present paper that allowance for internal circulation explains the possibility of gravitational coagulation without the introduction of additional interaction forces.

The radius of effective capture cross section is numerically calculated. The exact solution /3/ and the asymptotics /4/ are used for determining hydrodynamic forces in the case of axial symmetry and of small gap, and the above asymptotics are made precise. A numerical algorithm is developed for calculating the drag coefficient of particles moving the direction normal to their line of centers. This method is compared with the exact solution /5/. Estimates are given of the possibile effect of particle deformability and of molecular forces.

1. Statement of the problem. Consider the motion of two fluid spheres of radii a_1 and a_2 $(a_1 < a_2)$ in a viscous medium subjected to gravitation and Archimedean forces. The particles have the same viscosity μ, density ρ, and move in a medium of viscosity μ_e and density ρ_e. It is assumed that the quasi-stationary Stokes equations apply inside drops and in the outside medium. The tangential motion of drop surfaces is assumed not stabilized by surface-active substances, and their surface tension to be fairly high. Hence, as in /3—5/, we neglect the deviation of particle form from spherical, as the boundary condition take the absence of flow through the drop-medium interfaces. The velocities and tangential stresses are assumed continuous. Initially the particles are far away from each other and move at steady velocities $\mathbf{V}_1^\infty, \mathbf{V}_2^\infty$, while $\mathbf{V}_i^\infty \cdot \mathbf{l} > 0$, i.e. the spheres begin to converge (Fig.1). The problem is to determine the impact parameter d_∞ for which particle coagulation is possible.

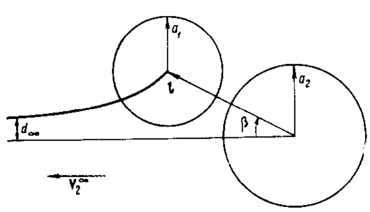

Fig.1

The density of particles is assumed equal to, or lower that of the medium. Hence it is reasonable to consider, when using Stokes equations, only the inertia-free equations of particles motion

$$\mathbf{F}_i + \frac{4}{3}\pi a_i^3(\rho - \rho_e)\mathbf{g} = 0 \tag{1.1}$$

where $\mathbf{F}_i$ are hydrodynamic forces.

In the case of slow motion of two solid spheres of similar material /6/ it is necessary to supplement Eqs. (1.1) by the condition of zero moments of hydrodynamic forces relative to particle centers (the condition of free rotation which independently of (1.1) makes it possible to express angular velocities of particles in terms of translational ones). To speak of liquid spheres rotation has no meaning, since the complicated internal motion is uniquely defined by the instantaneous velocities $\mathbf{V}_1, \mathbf{V}_2$ of the particles goemetric centers, and the absence of moments of hydrodynamic forces automatically follows from the boundary conditions and Stokes equations /5/.

*Prikl.Matem.Mekhan.,46,No.1,72-82,1982

According to /3,5/

$$\mathbf{F}_1 = -6\pi\mu_e a_1 [\Lambda_{11}(\mathbf{V}_1 - \mathbf{V}_2)'' + \Lambda_{12}\mathbf{V}_2'' + T_{11}(\mathbf{V}_1 - \mathbf{V}_2)^{\perp} + T_{12}\mathbf{V}_2^{\perp}] \tag{1.2}$$
$$\mathbf{F}_2 = -6\pi\mu_e a_2 [\Lambda_{21}(\mathbf{V}_2 - \mathbf{V}_1)'' + \Lambda_{22}\mathbf{V}_2'' + T_{21}(\mathbf{V}_2 - \mathbf{V}_1)^{\perp} + T_{22}\mathbf{V}_2^{\perp}]$$

where $\mathbf{V}_i''$ is the vector component of velocity $\mathbf{V}_i$ along the line of centers, $\mathbf{V}_i^{\perp}$ is the projection of vector $\mathbf{V}_i$ on a plane normal to the line of centers. Coefficients Λ_{ij}, T_{ij} depend on ε, k, λ (εa_1 is the gap between spheres, and $k = a_1 / a_2$, $\lambda = \mu / \mu_e$) and are considered in greater detail in Sects.2 and 3.

The projection of relations (1.1) on the line of centers and on the plane normal to that line, with allowance for (1.2), enables us to eliminate from the obtained equalities the components of velocity $\mathbf{V}_2$, and express the equations of relative motion in the form

$$\Lambda(l)\, dl / dt = -\varkappa \cos\beta, \quad l\,[T(l)]^{-1} d\beta / dt = \varkappa \sin\beta \tag{1.3}$$
$$\Lambda = \frac{\Lambda_{11}\Lambda_{22} + \Lambda_{21}\Lambda_{12}}{\Lambda_{12} - k^2\Lambda_{22}}, \quad T = \frac{T_{12} - k^2 T_{22}}{T_{11}T_{22} + T_{12}T_{21}}, \quad \varkappa = \frac{2a_2^2 |\rho - \rho_e| g}{9\mu_e}$$

where the first two formulas are similar to equations in /6/ of relative motion of solid spheres. Eliminating time from (1.3) and taking into account the meaning of the aiming parameter d_∞ (Fig.1), we obtain equations of relative trajectories of the form

$$F(l) = \ln\left(\frac{d_\infty}{\sin\beta}\right), \quad F(l) = \ln l - \int_l^\infty \frac{T(l')\Lambda(l') - 1}{l'}\, dl' \tag{1.4}$$

Convergence of integral in (1.4) is implied by the asymptotic formulas /5,7/ for T_{ij}, Λ_{ij} according to which $\Lambda(l)\,T(l) = 1 + O(a_i / l)$ as $l \to \infty$.

Calculations show that Λ, $T > 0$, always (see Sects.2 and 3), hence $dF / dl > 0$. Moreover, as $\varepsilon \to 0$ function Λ has a singularity of order not higher than $\varepsilon^{-1/2}$ (Sect.2), and function T remains finite (Sect.3), therefore $F(a_1 + a_2)$ is finite. It follows from this and (1.4) that when $\ln d_\infty \leqslant F(a_1 + a_2)$, any relative trajectory arriving from infinity reaches the sphere $l = a_1 + a_2$ (i.e. there is coagulation) and, as implied by Eqs.(1.3), the time of motion taken from any point of the relative trajectory to reach the sphere $l = a_1 + a_2$ is finite. When $\ln d_\infty > F(a_1 + a_2)$ the relative trajectory does not reach the sphere $l = a_1 + a_2$ but moves into infinity and is symmetric relative to the plane $\beta = \pi / 2$. The critical value of the aiming parameter d_∞^* is, thus, defined by the equality

$$\frac{d_\infty^*}{a_1 + a_2} = S = \exp\left[-\int_{a_1+a_2}^\infty \frac{\Lambda(l)\,T(l) - 1}{l}\, dl\right] \tag{1.5}$$

In the case of solid spheres Λ has a sigularity of the order of ε^{-1} as $\varepsilon \to 0$ /8/, hence contact of spheres cannot occur within a finite time without the introduction of additional interaction forces that are singular as $\varepsilon \to 0$.

When λ is large, the domain of very small ε provides a substantial contribution to the integral in (1.5) which becomes divergent as $\lambda = \infty$. Moreover, when ε is very small, it may prove essential to take into account additional effects, such as the Van-der-Waals forces, etc. Because of this, the most interesting is the determination of integrals (1.5) in the case of fairly small λ.

The methods of calculating coefficients Λ_{ij} and T_{ij} used in the determination of integrals (1.5) are presented below. These methods relate to the case of drops of generally different viscosities μ_1, μ_2 . They can be also of interest for a more general determination of coagulation effectiveness with allowance for additional nonhydrodynamic forces.

2. Calculation of coefficients Λ_{ij}. The exact solution of the axisymmetric problem constructed in bispherical coordinates in /3/ is used for determining coefficients Λ_{ij} . According to /3/

$$\Lambda_{11} = \frac{\sqrt{2}\,\mathrm{sh}\,\alpha}{3c^2} \sum_{n=1}^\infty \frac{\delta_0 + \lambda_1\delta_1 + \lambda_2\delta_2 + \lambda_1\lambda_2\delta_3}{\Delta}, \quad \lambda_i = \frac{\mu_i}{\mu_e}$$

The same formula with the substitution of $\bar{\delta}_i$ for δ_i is valid for $\Lambda_{12} - \Lambda_{11}$. Parameters $c, \alpha, \delta_i, \bar{\delta}_i, \Delta$ were defined in /3/. We would point out the misprints in formulas in /3/ have in the denominator c instead of c^2, and the expression for δ_3 is of the wrong sign. Calculation formulas for $\Lambda_{21}, \Lambda_{22}$ are obtained by interchanging the position of spheres, bearing in mind the validity of the reciprocity relation /7/

$$\Lambda_{11} - k^{-1}\Lambda_{21} = \Lambda_{12}$$

As shown in /6,8/, the convergence of series defined in bispherical coordinates worsens as $\varepsilon \to 0$ and can be indefinitely slow. Because of this the use of series for calculating coefficients Λ_{ij} with small ε is ineffective /3/, and asymptotic formulas are mainly used for such calculations. In the case of small ε coefficients $\Lambda_{12}, \Lambda_{22}$ were replaced by their limit values $\Lambda_{12}^t, \Lambda_{22}^t$ for touching spheres with reasonable accuracy. The method of calculating Λ_{12}^t, Λ_{22}^t and their numerical values appeared in /9/. For Λ_{11} the following asymptotics were used:

$$\Lambda_{11} \simeq \frac{\pi^2\sqrt{2}\,(\lambda_1+\lambda_2)}{32\,(1+k)^{3/2}\sqrt{\varepsilon}} - \frac{1}{3\,(1+k)}\left[1 + \frac{\lambda_1\lambda_2 - \lambda_1^2 - \lambda_2^2}{3}\right]\ln\varepsilon \; + c_0\,(k, \lambda_1, \lambda_2) \tag{2.1}$$

In a number of cases the first two terms of (2.1), obtained in /4/ did not ensure sufficient accuracy, and the next following term was determined. The internal expansion of the stream function obtained in /4/ is valid in the region of small gap between spheres. The contribution of that region to the coefficient Λ_{11} was also determined there, and a method constructing external expansion valid for the remaining region flow between spheres was roughly outlined there. The contribution of external expansion to coefficient Λ_{11} is determined as in the case of interaction between a solid sphere and a solid plane /8/. As the result, we have

$$c_0 = \frac{(3+\lambda_1\lambda_2-\lambda_1^2-\lambda_2^2)}{9\,(1+k)}\{\ln[2\,(1+k)] - 1\} + \tag{2.2}$$

$$\frac{1}{6}\int_0^\infty ds\left\{-\frac{2\,(\lambda_1+\lambda_2)}{(1+k)^2 s^2} + \frac{4e^{-2s}(\lambda_1^2+\lambda_2^2-\lambda_1\lambda_2-3)}{3\,(1+k)\,s} + \frac{\psi_0+\lambda_1\psi_1+\lambda_2\psi_2+\lambda_1\lambda_2\psi_3}{\varphi_0+(\lambda_1+\lambda_2)\,\varphi_1+\lambda_1\lambda_2\varphi_3}\right\}$$

$$\psi_0 = (1+2s)\,e^{2ks} + (2ks-1)\,e^{-2s}$$
$$\psi_1 = (1+2s+2s^2)\,e^{2ks} + (1-2ks)\,e^{-2s}$$
$$\psi_2 = (1+2s)\,e^{2ks} + (1-2ks+2k^2s^2)\,e^{-2s}$$
$$\psi_3 = (1+2s+2s^2)\,e^{2ks} - (1-2ks+2k^2s^2)\,e^{-2s}$$
$$\varphi_0 = \mathrm{sh}^2[(1+k)\,s],\quad \varphi_1 = {}^1/_2\,\{\mathrm{sh}\,[2\,(1+k)\,s] - 2\,(1+k)\,s\}$$
$$\varphi_3 = \mathrm{sh}^2[(1+k)\,s] - (1+k)^2 s^2$$

A comparison of approximate values of Λ_{11} determined by formula (2.1) with its exact value for $k = 0.5, \lambda_1 = 0.5, \lambda_2 = 1$ and various ε shows that the relative error does not exceed 3.5% when $\varepsilon \leqslant 0.1$ and 0.5% when $\varepsilon \leqslant 0.01$.

Remark. Formula (2.1) is not uniformly useful when $\lambda_1, \lambda_2 \to \infty$. When $\lambda_1\lambda_2 \gg 1$, $\varepsilon \ll 1$ a rough first approximation of the asymptotics of Λ_{11} is of the form /10/

$$\Lambda_{11} \simeq \varepsilon^{-1}(1+k)^{-2} f(p_1, p_2), \quad p_i = \lambda_i\sqrt{2\varepsilon\,(1+k)} \tag{2.3}$$

where $f(p_1, p_2)$ is expressed in terms of the logarithmic derivative of the gamma function. When $p_1, p_2 \to 0$, formula (2.3) is consistent with (2.1), and when $p_1, p_2 \to \infty$, with the asymptotics of solid spheres /8/.

Owing to the fairly slow decrease of the integrand in (1.5) as $l \to \infty$, it proved advantageous to use in calculations of further asymptotic expansions for coefficients Λ_{ij} /7/.

3. Calculation of coefficients T_{ij}.

An exact solution of the problem of slow motion of two liquid spheres whose instantaneous velocities are normal to their line of centers was constructed in bispherical coordinates in /5/. Coefficients T_{ij} are also represented by infinite series, but the terms of such series cannot be explicitly obtained, and have to be defined by solutions $\mathbf{w}_n$ of the system of difference equations

$$\sum_{k=-2}^{2} T_n^k \mathbf{w}_{n+k} = \delta_1 \mathbf{S}_n^1 + \delta_2 \mathbf{S}_n^2, \quad n \geqslant 1 \tag{3.1}$$
$$T_n^k = 0 \;\; (n+k < 1), \quad \mathbf{w}_n \to 0 \;\; (n \to \infty)$$

where T_n^k are some fourth order matrices, $\mathbf{w}_n$, $\mathbf{S}_n^j$ are four-dimensional vectors, and $\delta_l = 0$ or $\delta_l = 1$.

To obtain analytic expressions for the 88 elements of matrix T_n^k and vectors $\mathbf{S}_n^j$, although theoretically possible using the method developed in /5/, is extremely difficult in practice. A method of numerical calculation of T_n^k and S_n^j is also indicated there. Application of the matrix run-through to system (3.1) enables us to determine T_{ij} theoretically with any desired accuracy as the limits of recurrent sequencies /5/. However that method of calculating T_{ij} is complicated by the complexity of determination of T_n^k and S_n^j. In the range of small λ a simpler way of calculating T_{ij} based on the method of reflections proved to be effective and reasonably accurate. Unlike in /7/ and other publications on the hydrodynamic interaction of two spheres, in which the method of reflections was used for obtaining approximate analytic formulas applicable in cases of large distances between spheres, here it is considered to be a computational procedure.

The general recurrent formulas of the method of reflections appear in /7/, they are, however, complicated and unsuitable for computational purposes, owing to the unfortunate choice in that paper two spherical coordinate systems with polar axes normal to the line of centers. Simpler recurrent formulas are obtained below. They make possible the effective calculation of a considerably greater number of reflections than in /7/.

We normalize all distances with respect to distance l between the sphere centers. It is sufficient to consider the case in which a sphere of radius α_1 $(\alpha_i = a_i / l)$ travels at the instantaneous unit velocity $\mathbf{i}_x$ normal to the line of centers, while the second sphere is at rest. In conformity with the general scheme of the reflection method /7,11/, we seek a velocity field definition in the region between the spheres of the form

$$\mathbf{v} = \sum_{k=1}^{\infty} (\mathbf{v}_-^{1,2k-1} + \mathbf{v}_-^{2,2k}) \tag{3.2}$$

Every $\mathbf{v}_-^{i,j}$ field satisfies Stokes equations, is regular everywhere outside the sphere of radius α_i, and vanishes at infinity.

We determine fields $\mathbf{v}_-^{i,j}$ in the usual sequence

$$\mathbf{v}_+^{j,j} \to \mathbf{v}_-^{i,j+1} \to \mathbf{v}_+^{i+1,j+1} \to \mathbf{v}_-^{i+1,j+2} \to \mathbf{v}_+^{i,j+2} \to \ldots \tag{3.3}$$

where the initial field $\mathbf{v}_+^{1,0}$ is equal $-\mathbf{i}_x$, and $\mathbf{v}_+^{i+1,j}$ $(j \geqslant 1)$ denotes the expansion of field $\mathbf{v}_-^{i,j}$ in the neighborhood of the sphere of radius α_{i+1} (indices $i, i+1$ are reduced by module 2).

For the velocity field (3.2) to be a solution of the problem considered here it is necessary and sufficient that transition from $\mathbf{v}_+^{i,j}$ to $\mathbf{v}_-^{i,j+1}$ is determined by boundary conditions that are satisfied at each step on the surface of only one sphere.

1°. Field $\mathbf{v}_+^{i,j} + \mathbf{v}_-^{i,j+1}$ has a zero normal component on the sphere of radius α_i.

2°. A Stokes flow of fluid of viscosity μ_i whose velocities and the tangential stress at the boundary are the same as in field $\mathbf{v}_+^{i,j} + \mathbf{v}_-^{i,j+1}$ exists inside the sphere of radius α_i

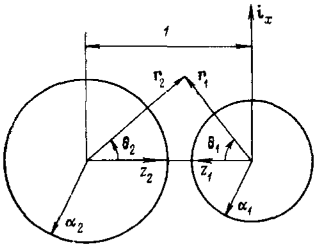

Fig.2

Unlike in /7/, the two systems of spherical coordinates introduced by us $(r_1, \theta_1, \varphi_1)$, $(r_2, \theta_2, \varphi_2)$ are as shown in Fig.2. The angle φ_i corresponds to positive rotation about the z_i axis, and $\varphi_i = 0$ to the half-plane defined by vector $\mathbf{i}_x$ and the line of centers. Using Lamb's general solution /11/ of Stokes equations, we represent the unknown fields in the form

$$\mathbf{v}_+^{i,j} = \sum_{n=1}^{\infty} \left[\operatorname{rot}(\mathbf{r}_i \chi_n^{i,j}) + \nabla \Phi_n^{i,j} + \frac{(n+3) r_i^2 \nabla p_n^{i,j}}{2(n+1)(2n+3)} - \frac{n \mathbf{r}_i p_n^{i,j}}{(n+1)(2n+3)} \right] \tag{3.4}$$

$$\mathbf{v}_-^{i,j} = \sum_{n=1}^{\infty} \left[\operatorname{rot}(\mathbf{r}_i \chi_{-(n+1)}^{i,j}) + \nabla \Phi_{-(n+1)}^{i,j} - \frac{(n-2) r_i^2 \nabla p_{-(n+1)}^{i,j}}{2n(2n-1)} + \frac{(n+1) \mathbf{r}_i p_{-(n+1)}^{i,j}}{n(2n-1)} \right]$$

In conformity with the general structure of the exact solution in /5/ the velocity components v_r and v_θ are proportional to $\cos\varphi$, and component v_φ to $\sin\varphi$, hence it is sufficient to consider spherical harmonics of the special form

$$p_{-(n+1)}^{i,j} = \zeta A_{-(n+1)}^{i,j} \cos\varphi_i, \quad \Phi_{-(n+1)}^{i,j} = \zeta B_{-(n+1)}^{i,j} \cos\varphi_i \tag{3.5}$$
$$\chi_{-(n+1)}^{i,j} = \zeta C_{-(n+1)}^{i,j} \sin\varphi_i, \quad \zeta = r_i^{-(n+1)} P_n^1(\cos\theta_i)$$

$$p_n^{i,j}=\eta A_n^{i,j}\cos\varphi_i,\quad \Phi_n^{i,j}=\eta B_n^{i,j}\cos\varphi_i$$
$$\chi_n^{i,j}=\eta C_n^{i,j}\sin\varphi_i,\quad \eta=r_i^{\,n}P_n^{\,1}(\cos\theta_i)$$

where $P_n^{\,1}$ is the associated Legendre function.

The transition from $\mathbf{v}_+^{i,j}$ to $\mathbf{v}_-^{i,j+1}$ is similar to that in /7/, with respective formulas of the form

$$C_{-(n+1)}^{i,j+1}=\frac{(n-1)(1-\lambda_i)}{n+2+\lambda_i(n-1)}C_n^{i,j}a_i^{2n+1} \tag{3.6}$$
$$A_{-(n+1)}^{i,j+1}=-\frac{n(2n-1)}{(n+1)(1+\lambda_i)}\left\{\frac{\lambda_i}{2}A_n^{i,j}a_i^{2n+1}+[2+\lambda_i(2n+1)]B_n^{i,j}a_i^{2n-1}\right\}$$
$$B_{-(n+1)}^{i,j+1}=\frac{n}{2(n+1)(1+\lambda_i)}\left\{\frac{[2-\lambda_i(2n+1)]}{2(2n+3)}A_n^{i,j}a_i^{2n+3}-\lambda_i(2n-1)B_n^{i,j}a_i^{2n+1}\right\}$$

To represent the field $\mathbf{v}_-^{i,j}$ in the neighborhood of the sphere of radius a_{i+1} in form $\mathbf{v}_-^{i,j+1}$ we begin by transforming the spherical harmonics. According to /11/ the following equality applies:

$$\frac{P_m(\cos\theta_i)}{r_i^{m+1}}=\sum_{n=0}^{\infty}\frac{(n+m)!}{n!m!}r_{i+1}^nP_n(\cos\theta_{i+1}) \tag{3.7}$$

Differentiating (3.7) with respect to θ_i we obtain

$$\frac{P_m^{\,1}(\cos\theta_i)}{r_i^{m+1}}=\sum_{n=1}^{\infty}g_n^{\,m}r_{i+1}^nP_n^1(\cos\theta_{i+1}),\quad g_n^{\,m}=\frac{(n+m)!}{(m-1)!(n+1)!} \tag{3.8}$$

Relations (3.8) enable us to show that the unknown formulas of transition from $\mathbf{v}_-^{i,j}$ to $\mathbf{v}_+^{i+1,j}$ are of the form

$$A_n^{i+1,j}=\sum_{m=1}^{\infty}g_n^{\,m}A_{-(m+1)}^{i,j} \tag{3.9}$$

$$C_n^{i+1,j}=\sum_{m=1}^{\infty}\left[\frac{g_n^{\,m}}{mn(n+1)}A_{-(m+1)}^{i,j}+\frac{m}{n+1}g_n^{\,m}C_{-(m+1)}^{i,j}\right]$$
$$B_n^{i+1,j}=\sum_{m=1}^{\infty}\left\{\frac{A_{-(m+1)}^{i,j}}{m(2m-1)}\left\{\frac{(n-1)[(m-2)(n-1)-(m+1)]}{n(2n-1)}g_{n-1}^m-\frac{m-2}{2}g_n^{\,m}\right\}+g_n^{\,m}B_{-(m+1)}^{i,j}+\frac{g_n^{\,m}}{n}C_{-(m+1)}^{i,j}\right\}$$

The fields $\mathbf{v}_+^{i+1,j}$, $\mathbf{v}_-^{i,j}$ defined by formulas (3.4) satisfy the equations /11/

$$\Delta\mathbf{v}_+^{i+1,j}=\sum_{n=1}^{\infty}\nabla p_n^{i+1,j},\quad \Delta\mathbf{v}_-^{i,j}=\sum_{m=1}^{\infty}\nabla p_{-(m+1)}^{i,j}$$

Since in the neighborhood of sphere a_{i+1} by definition $\mathbf{v}_+^{i+1,j}\equiv\mathbf{v}_-^{i,j}$, hence expanding each harmonic $p_{-(m+1)}^{i,j}$ using (3.5) and (3.8) and summating the results, we obtain the first of relations (3.9).

For the determination of $\chi_n^{i+1,j}$ we consider the identity /11/

$$\mathbf{r}_{i+1}\cdot\operatorname{rot}\mathbf{v}_+^{i+1,j}=\sum_{n=1}^{\infty}n(n+1)\chi_n^{i+1,j} \tag{3.10}$$

On the other hand, from the second of formulas (3.4) we have

$$\mathbf{r}_{i+1}\cdot\operatorname{rot}\mathbf{v}_-^{i,j}=-\sum_{m=1}^{\infty}\frac{1}{m}\frac{\partial p_{-(m+1)}^{i,j}}{\partial\varphi_{i+1}}+\sum_{m=1}^{\infty}r_{i+1}\frac{\partial}{\partial r_{i+1}}\left[\chi_{-(m+1)}^{i,j}+r_{i+1}\frac{\partial\chi_{-(m+1)}^{i,j}}{\partial r_{i+1}}-\mathbf{i}_z\cdot\nabla\chi_{-(m+1)}^{i,j}\right] \tag{3.11}$$

where $\mathbf{i}_z$ is the unit vector of axis z_{i+1} and partial derivations are effected in coordinates $r_{i+1},\theta_{i+1},\varphi_{i+1}$. Harmonics $p_{-(m+1)}^{i,j},\chi_{-(m+1)}^{i,j}$ are defined in these coordinates using (3.5) and (3.8). It is not difficult to calculate

$$\mathbf{i}_z\cdot\nabla\chi_{-(m+1)}^{i,j}=-C_{-(m+1)}^{i,j}\sin\varphi_{i+1}\sum_{n=1}^{\infty}(n+2)g_{n+1}^mr_{i+1}^nP_n^{\,1}(\cos\theta_{i+1}) \tag{3.12}$$

Taking into account (3.10)—(3.12) we obtain the second of relations (3.9).

For the determination of $\Phi_n^{i+1,j}$ we use the identity /11/

$$\mathbf{r}_{i+1}\cdot\mathbf{v}_+^{i+1,j} = \sum_{n=1}^{\infty}\left[\frac{n}{2(2n+3)}r_{i+1}^2 p_n^{i+1,j} + n\Phi_n^{i+1,j}\right] \tag{3.13}$$

On the other hand, from the second of formulas (3.4) we have

$$\mathbf{v}_-^{i,j}\cdot\mathbf{r}_{i+1} = \sum_{m=1}^{\infty}\frac{r_{i+1}}{m(2m-1)}\left\{\cos\theta_{i+1}\left[(m-2)r_{i+1}\frac{\partial p_{-(m+1)}^{i,j}}{\partial r_{i+1}} - (m+1)p_{-(m+1)}^{i,j}\right] + (m+1)r_{i+1}p_{-(m+1)}^{i,j} - \frac{(m-2)}{2}(1+r_{i+1}^2)\frac{\partial p_{-(m+1)}^{i,j}}{\partial r_{i+1}}\right\} + \sum_{m=1}^{\infty}\left(r_{i+1}\frac{\partial\Phi_{-(m+1)}^{i,j}}{\partial r_{i+1}} - \frac{\partial\chi_{-(m+1)}^{i,j}}{\partial\varphi_{i+1}}\right) \tag{3.14}$$

Defining harmonics $p_{-(m+1)}^{i,j}$, $\chi_{-(m+1)}^{i,j}$, $\Phi_{-(m+1)}^{i,j}$ in coordinates r_{i+1}, θ_{i+1}, φ_{i+1} and using the recurrent relations for $\cos\theta\, P_n^1(\cos\theta)$, we can represent the expression (3.14) in the form

$$\sum_{n=1}^{\infty}(E_n r_{i+1}^n + F_n r_{i+1}^{n+2})P_n^1(\cos\theta_{i+1})\cos\varphi_{i+1}$$

with some coefficients E_n, F_n. The expression in (3.13) can be represented in the same form. Comparing coefficients E_n, in both expressions, we obtain the third of relations (3.9).

The initial condition $\mathbf{v}_+^{1,0} = -\mathbf{i}_x$ implies that

$$A_n^{1,0} = C_n^{1,0} = 0 \quad (n \geqslant 1), \quad B_1^{1,0} = -1, \quad B_n^{1,0} = 0 \quad (n \geqslant 2) \tag{3.15}$$

Formulas (3.3), (3.6), (3.9), and (3.15) uniquely define the calculation sequence.

In conformity with data in /11/ the hydrodynamic forces $F_i = -4\pi\mu_e\nabla(r_i^3 p_{-2}^i)$, where p_{-2}^i is the respective harmonic in Lamb's representation of the resultant velocity field (3.2) in the neighborhood of the sphere of radius α_i. Taking into account (3.2), (3.4), (3.5), and (1.1), we obtain

$$T_{11} = \frac{2}{3\alpha_1}\sum_{s=1}^{\infty}A_{-2}^{1,2s-1}, \quad T_{21} = -\frac{2}{3\alpha_2}\sum_{s=1}^{\infty}A_{-2}^{2,2s}$$

If we restrict the investigation to the few first reflections, the above recurrent formulas or formulas in /7/, we obtain for coefficients T_{ij} approximate analytic formulas in the form of polynomials of α_1, α_2, which are asymptotically correct as $\alpha_1, \alpha_2 \to 0$. Such formulas were derived in /7/ and corrected in /5/, where it was shown that these formulas may lead to considerable errors when ε is small. Because of this, the indicated analytical approach was extended to the case of arbitrary numbers of reflections, and realized numerically. Fixing the ratio $k = \alpha_1/\alpha_2$, we represent the coefficients T_{11}, T_{21} in the form of Taylor series

$$T_{11} \simeq \sum_{n=1}^{n_0} a_{11,n}y^{2n-2}, \quad T_{21} \simeq \sum_{n=1}^{n_0} a_{21,n}y^{2n-1}, \quad y = \alpha_1 + \alpha_2 \tag{3.16}$$

For the determination of coefficients $a_{11,n}$, $a_{21,n}$ $(n \leqslant n_0)$ it is sufficient to perform $2n_0$ reflections (taking each transition from $\mathbf{v}_+$ to $\mathbf{v}_-$ as a reflection), since further reflections do not contribute to the finite sums (3.16). For the same reason it is sufficient to take $n \leqslant n_0 + (1-j)/2$, $m \leqslant n_0 - n + (3-j)/2$ in the conversion formulas (3.9). The quantities $A_n^{i,j}$, $A_{-(n+1)}^{i,j}$ etc. were assumed to be polynomials of y of power not higher than $2n_0$, and transformations were effected on coefficients of these polynomials; in all transformations terms y^s with $s > 2n_0$ were rejected. We stress that the above numerical algorithm provides a strict method for calculating coefficient of Taylor series independent of n_0. The operational memory volume of the available computer imposed the constraint $n_0 \leqslant 115$.

The coefficients of Taylor series for $T_{21} + T_{22}$ and $T_{11} - T_{12}$ can be obtained by exchanging the places of spheres. The numerically calculated data tabulated below show the convergence of the asymptotic series (3.16) to the exact values of T_{ij}. Values of function T with $k = 0.25$, $\lambda = 10$, $\varepsilon = 0.08$ and various n_0 were calculated using approximate values of T_{ij} defined by formulas (3.16). The exact value of T calculated by the method of /5/ for $n_0 = \infty$ are:

n_0	10	30	60	115	∞
T	0.2604	0.2523	0.2508	0.25053	0.25051

Calculations have shown that series (3.16) are convergent even when $y = 1$ (which conforms with data of /5/, according to which coefficients T_{ij} remain finite when the spheres are in contact), but this convergence can be fairly slow. Calculations enable us to assume that for fixed k, λ $(\lambda < \infty)$ and $n \to \infty$ parameters $a_{ij,n}$ approach zero somewhat more rapidly than n^{-2} but this estimate is not uniform with respect to λ as $\lambda \to \infty$. To find the upper bound of the convergence rate we considered the limit case of freely rotating solid spheres $\lambda = \infty$. It was shown in /5/ that in this case

$$T_{ij}(\varepsilon) = T_{ij}(0) + O(|\ln \varepsilon|^{-1}), \quad \varepsilon \to 0 \tag{3.17}$$

Matching the asymptotics of $a_{ij,n}$ with formula (3.17) when $n \to \infty$ enables us to assume that when $\lambda = \infty$

$$a_{ij,n} = O[(n \ln^2 n)^{-1}], \quad n \to \infty \tag{3.18}$$

from which follows (3.17). In any case, when $\lambda = \infty$ and $\varepsilon = 0$ series (3.16) converge extremely slowly.

Table 1

	$\varepsilon=0.0005$				$\varepsilon=0.015$			
k	$\lambda=0$	3	10	30	$\lambda=0$	3	10	30
0.15	814	268	178	145	815	271	182	149
	814	267	169	123	815	270	176	137
0.35	779	329	252	221	782	333	257	227
	779	329	247	206	782	333	255	221
0.75	405	194	159	145	407	196	163	149
	405	194	158	141	407	196	162	148

Table 1 gives an idea of the accuracy of the method of reflections when $n_0 = 47$. It shows for each set of k, λ, ε a column of quantities $T^* \times 10^3$, $T \times 10^3$ with T^* and T denoting the approximate and exact values, respectively and $(T^* \geqslant T)$. The table shows the satisfactory accuracy of the proposed method up to the contact of spheres in the case of small λ and $n_0 = 47$; its accuracy is considerably higher when the spheres are clearly separated. The method is also reasonably effective, since with $n_0 = 47$ the calculation of all coefficients $a_{ij,n}$ $(n \leqslant n_0)$ for each pair of values of k, λ required approximately two minutes of computer time. When the coefficients of Taylor series are known, formulas (3.16) provide a simple dependence of T_{ij} on ε, while the method in /7/ necessitates separate computations for each relative position of spheres.

Since series (3.16) converge to exact values of T_{ij}, the proposed method should be considered as theoretically exact. It should be, however, pointed out that in the case of large λ and small ε it is extremely difficult to obtain by it reliable values of T_{ij}. This is so because in the case of large λ coefficients $a_{ij,n}$ initially approach zero very slowly (due to the indicated above nonuniform behavior of these coefficients relative to λ as $n \to \infty$), hence a comparatively small increase of n_0 does not markedly improve the accuracy. For example, for $k = 0.15$, $\lambda = 30$, $n_0 = 94$ the proposed method yields $T^* = 0.137$ when $\varepsilon = 0.0005$, and $T^* = 0.143$ when $\varepsilon = 0.015$, which improves only little the value of T^* appearing in Table 1 for $n_0 = 47$. Simultaneously the necessary computer memory volume and the computation time of coefficients $a_{ij,n}$ sharply increases as n_0 is increased. In the proposed here algorithm the required memory volume increases in proportion to n_0^2 and the computation time in proportion to n_0^4, reaching 35 min for each pair of values of k, λ when $n_0 = 94$. Because of this the method of /5/, insensitive to computation accuracy, is the only one reliable scheme for computing T_{ij} in the case of large λ and small ε

Remark. In the general recurrent formulas /7/ it is necessary to use as $p_n, p_{-(n+1)}$, etc. spherical harmonics of the general form, hence the constants that define these harmonics depend on one more additional index. Moreover the formulas transform in /7/ (much more complex than (3.9) involve double summation. A direct application of formulas /7/ in the computation of coefficients $a_{ij,n}$ would result in the necessary memory volume of computer and computation time becoming proportional to n_0^3 and n_0^6, respectively, making that method less efficient that the one proposed here.

4. Results of computation of coagulation effectiveness. Values of $S \times 10^3$ computed for various k and λ are given in Table 2. Since $\lambda \leqslant 10$ was used in computations, the method expounded in Sect.3 for computing T with $n_0 = 47$ ensured the required accuracy of computation of S in the majority of variants. The remaining variants were corrected using the method of /5/.

It is interesting to evaluate the effect of the region of small ε on S. If it is conditionally assumed that coagulation occurs when ε reaches some value ε_0, then the critical parameter d_∞^* is determined by (1.4) with $l = a_1(1 + k^{-1} + \varepsilon_0)$ and $\beta = \pi/2$. Values of $S(\varepsilon_0) \times 10^3$ (where $S(\varepsilon_0) = d_\infty^*(\varepsilon_0) / (a_1 + a_2)$) with $\varepsilon_0 = 10^{-2}, 10^{-3}$ appear in Table 3. In the case of solid spheres coefficients T_{ij} were computed using the method of /5/ by passing to limit with $\lambda \to \infty$.

Table 2

k	$\lambda=0$	0.5	1	2	5	10
0.15	317	242	202	157	103	73
0.25	378	294	247	195	131	95
0.35	416	327	277	221	152	111
0.5	450	357	306	247	173	129
0.75	474	380	327	267	191	145
0.9	478	384	332	271	195	148

Table 3

k	$\lambda=0$	0.5	1	5	10	∞
0.25	384	306	263	157	127	82
	379	297	252	140	106	51
0.5	457	373	327	211	177	126
	451	361	312	186	146	79
0.9	488	404	357	241	206	156
	480	389	339	210	169	98

Let us consider the axisymmetric convergence ($\beta = 0$) on the assumption that the condition $\mu_e V_i^\infty / \sigma \ll 1$ (σ is the surface tension) which ensures the smallness of deformation for $\varepsilon \gtrsim 1$, we determine for which ε the deformation becomes substantial. As shown in /4,10/ regardless of λ size when $\varepsilon \ll 1$, the areas of the sphere surface sections, where considerable lubrication pressure which determine the singularity of Λ_{11}, are of the order of εa_i^2, hence in the region of the small gap pressure $p \sim (\varepsilon a)^{-1} \cdot \mu_e \Lambda_{11} dl / dt$. Taking into account that $\Lambda_{11} \lesssim \Lambda$ and using for dl/dt its expression in (1.3), we find that deformation can only be substantial when

$$\varepsilon \lesssim a_i^2 \mid \rho - \rho_e \mid g / \sigma \tag{4.1}$$

For example, for $a_i \sim 30$ μm, $|\rho - \rho_e| \sim 0.2$ g/cm^3, and $\sigma \sim 0.05$ N/m the extimate yields $\varepsilon \lesssim 4 \cdot 10^{-5}$. But condition (4.1) does not take into account any allowance for additional nonhydrodynamic forces. Using the conventional definition of molecular forces

$$F \approx \frac{A}{6(1+k) a_1 \varepsilon^2} \quad (\varepsilon \ll 1), \quad A = \text{const}$$

we find these forces comparable with gravitational forces when

$$\varepsilon \lesssim (24 a_i^4 \mid \rho - \rho_e \mid g / A)^{-1/3} \quad (k, \ 1 - k \sim 1)$$

For $A \sim 10^{-20}$ J, $|\rho - \rho_e| \sim 0.2$ g/cm^3, and $a_i \sim 10 \div 30$ μm the estimate yields $\varepsilon \lesssim 10^{-2} \div 10^{-3}$.

The author thanks A.M. Golovin for his interest in this work, and the Reviewer for his remarks.

REFERENCES

1. ENTOV V.M., KAMINSKII V.A. and SINIAVSKII E.G., On the capture of small drops by large ones in an electric field. Izv. Akad. Nauk SSSR, MZhG, No.5, 1973.
2. ENTOV V.M., KAMINSKII V.A. and LAPIGA E.Ia., On the calculation of the rate of emulsion coalescence in a turbulent stream. Izv. Akad. Nauk SSSR, MZhG, No.3, 1976.
3. HABER S.,HETSRONI G. and SOLAN A., On the low Reynolds number motion of two droplets.Intern. J. Multiphase Flow, Vol.1, No.1, 1973.
4. ZINCHENKO A.Z., Calculation of hydrodynamic interaction of drops at low Reynolds numbers. PMM, Vol.42, No.5, 1978.
5. ZINCHENKO A.Z., The slow asymmetric motion of two drops in a viscous medium. PMM, Vol.44, No.1, 1980.
6. WACHOLDER E. and SATHER N.F., The hydrodynamic interaction of two unequal spheres moving under gravity through quiescent viscous fluid. J. Fluid Mech., Vol.65, Pt.3, 1974.
7. HETSTRONI G. and HABER S., Low Reynolds number motion of two drops submerged in an unbounded arbitrary velocity field. Intern. J. Multiphase Flow, Vol.4, No.1, 1978.
8. COOLEY M.D.A. and O'NEILL M.E., On the slow motion generated in a viscous fluid by the approach of a sphere to a plane wall or stationary sphere. Mathematika, Vol.16, No.1,1969.
9. REED L.D. and MORRISON Jr.F.A., The slow motion of two touching fluid spheres along their line of centers. Intern. J. Multiphase Flow, Vol.1, No.4, 1974.
10. ZINCHENKO A.Z., Calculation of close interaction between drops, with internal circulation and slip effect taken into account. PMM, Vol.45, No.4, 1981.
11. HAPPEL J. and BRENNER H., Hydrodynamics at Low Reynolds Numbers /Russian translation/, Moscow, MIR, 1976.

Translated by J.J.D.