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CALCULATING THE EVOLUTION OF COAGULATING SYSTEMS

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We substantiate a quantitative analysis of the granulometric composition of coagulating particles.

The collision and subsequent merging (cohering) of particles in a stream with the formation of aggregates are determined by a wide range of effects – random (thermal or Brownian) motion and convergence under the action of electrical, gravitational, hydrodynamic, and other forces. Strictly speaking, study of the evolution of the granulometric composition of the particles coagulating in a stream should be made on the basis of mathematical analysis of the laws of mass and momentum conservation for each phase of the mixture, and also on the basis of a balance relationship with respect to the probability density of the coagulating particles in the form, for example, of the Smolukhovskii kinetic equation [1]. The difficulties arising in this approach are so great that it is, as a rule, possible to obtain results with respect to quantitative modeling of the kinetics of the coagulating particles only for problems of the simplest type.

Assuming that with regard to the conditions of realization of the particle interaction process all the assumptions adopted in deriving the Smolukhovskii equation are met, we have [2]

$$\partial n(m)/\partial t = 0{,}5\int_0^m \beta_1(m-\mu,\mu)n(m-\mu)n(\mu)d\mu - n(m)\int_0^\infty \beta_1(m,\mu)n(\mu)d\mu, \tag{1}$$

where n(m) is the probability density (PD) of the particles with respect to mass; t is time; $\beta_1(m, \mu)$ is the kernel of the integrodifferential equation (1), which is on the basis of its physical meaning a non-negative symmetric function of its arguments. With the introduction of the Dirac delta function, Eq. (1) takes the form [2, 3]

$$\partial n/\partial t = \int_0^\infty\int_0^\infty K(m, m', m'')n(m')n(m'')dm'dm''. \tag{2}$$

Here

$$K(m, m', m'') = 0{,}5\beta_1(m', m'')\Delta(m, m', m'');$$
$$\Delta(m, m', m'') = \delta(m - m' - m'') - \delta(m - m') - \delta(m - m'').$$

We take as the initial condition for the PD

$$n(m, 0) = n^0(m). \tag{3}$$

To simplify the analysis of the problem (2), (3), we convert to dimensionless quantities using the formulas

$$n = M^{-1}L^{-3}\bar{n},\ t = T\bar{t},\ m = M\bar{m}, \tag{4}$$

Where M, L, T are the characteristic magnitudes of the mass, length, and time, which are specified for each specific problem; $\bar{n}$, $\bar{t}$, $\bar{m}$ are the dimensionless PD, time, and mass. Then, substituting Eq.(4) into Eq. (2) and dropping for simplicity the bars over the dimensionless quantities, we have

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$$\partial n / \partial t = \varepsilon \int_0^\infty \int_0^\infty K(m, m', m'')n(m')n(m'')dm'dm'' \tag{5}$$

(ε is a dimensionless parameter). The initial condition (3) with respect to the dimensionles PD retains its form.

We find the solution of the integrodifferential equation (5), agreeing with the initial condition (3), in the form of the series

$$n(m, t; \varepsilon) = n_0(m, t) + \varepsilon n_1(m, t) + \dots \tag{6}$$

Then as a consequence of Eqs. (3), (6) we will have the initial conditions with respect to the successive approximations n_0, n_1, ...,:

$$n_0(m, 0) = n^0(m); \tag{7}$$

$$n_1(m, 0) = 0,\ n_2(m, 0) = 0, \dots, \tag{8}$$

and in accordance with Eqs. (2), (6) we will have the reduced system of differential equations

$$\partial n_0 / \partial t = 0; \tag{9}$$

$$\partial n_1 / \partial t = \int_0^\infty \int_0^\infty K(m, m', m'')n_0(m')n_0(m'')dm'dm''; \tag{10}$$

$$\partial n_2 / \partial t = \int_0^\infty \int_0^\infty K(m, m', m'')[n_0(m')n_1(m'') + n_1(m')n_0(m'')]dm'dm'', \tag{11}$$

....

the right sides of which are the explicit double quadratures with respect to the approximations of lower order. By virtue of the specific nature of the system (9)-(11), the series (6) takes the form

$$n(m, t; \varepsilon) = \sum_{i=0}^{\infty} (\varepsilon t)^i \varphi_i(m) / i!, \tag{12}$$

where $\varphi_0(m) = n_0(m)$, $\varphi_i(m)$ ($i = 1, 2, \dots$) are the explicit expressions for the multiple quadratures on the right in the system (9)-(11). Considering Eqs. (7), (8), on the basis of Eqs. (9)-(11) we obtain

$$n_0 = n^0(m) = \varphi_0(m); \tag{13}$$

$$n_1 = \varphi_1(m)t,\ n_2(m, t) = 0{,}5\, \varphi_2(m)t^2, \dots \tag{14}$$

Here

$$\varphi_1(m) = \int_0^\infty \int_0^\infty K(m, m', m'')\varphi_0(m')\varphi_0(m'')dm'dm''; \tag{15}$$

$$\varphi_2(m) = \int_0^\infty \int_0^\infty K(m, m', m'')[\varphi_0(m')\varphi_1(m'') + \varphi_1(m')\varphi_0(m'')]dm'dm'', \tag{16}$$

....

In the following, without loss of generality of the arguments, we shall examine an initial PD in the form of the superposition of a pair of delta functions [2]:

$$n^0(m) = \nu_0[\alpha_1\delta(m - m_1) + \alpha_2\delta(m - m_2)] \tag{17}$$

($\nu_0 = N_0L^3$, N_0 is the number of particles in unit volume of the starting mixture, $\alpha_1 + \alpha_2 = 1$, $m_1 < m_2$). Moreover, for the sake of some reduction of the volume of the calculations, in the specific quantitative analysis of the problem we shall further assume that the kernel of Eq. (1) is a difference kernel. In this case we have from Eqs. (15), (17), using the properties of the delta function and considering the prohibition on coagulation of particles of identical mass

$$\varphi_1(m) = \alpha_1\alpha_2\nu_0^2 D(m, m_1, m_2), \tag{18}$$

where

$$D(m, m_1, m_2) = \beta(m_1, m_2)\Delta(m, m_1, m_2),\ \beta = \bar{\beta}_1.$$

To find the higher approximations of the expansion (6) we use the obvious formulas

$$\begin{aligned}\int_0^\infty\int_0^\infty D(z, x, y)D(x, a, b)[\alpha_1\delta(y - a) + \alpha_2\delta(y - b)]dxdy = \\ \beta(a, b)\{\alpha_1[D(z, a + b, z_1) - D(z, a, z_1) - D(z, b, z_1)] + \\ \alpha_2[D(z, a + b, z_2) - D(z, a, z_2) - D(z, b, z_2)]\};\end{aligned} \tag{19}$$

$$\begin{aligned}\int_0^\infty\int_0^\infty D(z,x,y)D(x,a,b)D(y,c,d)dxdy = \beta(a,b)\beta(c,d) \times \\ [D(z,a + b,c + d) - D(z,a,c + d) - D(z,b,c + d) + D(z,a + b,c) - \\ D(z,a,c) - D(z,b,c) + D(z,a + b,d) - D(z,a,d) - D(z,b,d)]\end{aligned} \tag{20}$$

(a, b, c, d, z_1, z_2 are positive constants). Then by virtue of Eqs. (13), (16-19), and also on the basis of the difference nature of the kernel, we obtain

$$\begin{aligned}\varphi_2(m) = 2\int_0^\infty\int_0^\infty K(m, m', m'')\varphi_0(m')\varphi_1(m'')dm'dm'' = \\ \int_0^\infty\int_0^\infty D(m, m', m'')\varphi_0(m')\varphi_1(m'')dm'dm'' = \\ \alpha_1\alpha_2\nu_0^3\beta(m_1, m_2)[\alpha_1 D(m, m_1 + m_2, m_1) - \\ (\alpha_1 + \alpha_2)D(m, m_1, m_2) + \alpha_2 D(m, m_1 + m_2, m_2)].\end{aligned} \tag{21}$$

Similarly, on the basis of Eqs. (13), (17-21) we have

$$\begin{aligned}\varphi_3(m) = 0{,}5\int_0^\infty\int_0^\infty D(m,m',m'')[2\varphi_0(m')\varphi_2(m'') + \varphi_1(m')\varphi_1(m'')]dm'dm'' = \\ \alpha_1\alpha_2\nu_0^4\beta(m_1,m_2)(\alpha_1\beta(m_1 + m_2,m_1)\{\alpha_1[D(m,2m_1 + m_2,m_1) - \\ D(m,m_1 + m_2,m_1)] + \alpha_2[D(m,2m_1 + m_2,m_2) - D(m,m_1 + m_2,m_2) - \\ D(m,m_1,m_2)]\} - (\alpha_1 + \alpha_2)\beta(m_1, m_2)\{\alpha_1[D(m,m_1 + m_2,m_1) - \\ D(m,m_1,m_2)] + \alpha_2[D(m,m_1 + m_2,m_2) - D(m,m_1,m_2)]\} + \\ \alpha_2\beta(m_1 + m_2,m_2)\{\alpha_1[D(m,m_1 + 2m_2,m_2) - D(m,m_1 + m_2,m_1) - \\ D(m,m_2,m_1)] + \alpha_2\beta(m_1 + m_2,m_2)[D(m,2m_2 + m_1,m_2) - \\ D(m,m_1 + m_2,m_2)]\} - \alpha_1\alpha_2\beta(m_1,m_2)D(m,m_1,m_2)).\end{aligned} \tag{22}$$

It follows from the analysis of Eqs. (17), (18), (21), (22) that each of their successive approximations yields an estimate of the contribution to the overall number of particles of increasingly large aggregates, formed as the result of the merging of particles of one size with multiple monomers of another size. However, the finding of approximations of order higher than the third becomes a quite complex problem, although the structure of these approximations is evident. Since in accordance with Eqs. (17), (18), (21), (22) the series (12) is the superposition of delta-type functions, it is difficult to substantiate its convergence. However, for the primitive with respect to m (if it exists) of this expansion, representing with accuracy to a multiplier the distribution function, under the specific limitations imposed on the coagulation kernel β the majorant can be constructed, and thus the convergence of the series

$$\int_0^m n(m, t; \varepsilon)dm = \sum_{i=0}^{\infty} (\varepsilon t)^i \int_0^m \varphi_i(m)dm / i! \tag{23}$$

can be shown.

Considering only the symmetry of the kernel of Eq. (1) and using formulas (15)-(20), we conclude that (for a non-difference kernel!) if the zero approximation contains two terms, then each of the primary terms in the first approximation contains, $2^2 \cdot 3$ terms, in the second approximation – $2^3 \cdot 3^2$ terms, and in the i-th approximation – $2^{i+1} \cdot 3^i$ terms. Therefore the i-th approximation depends on i $2^{i+1} \cdot 3^i$ terms, each of which in accordance with Eqs. (19), (20) is proportional to the product

$$\prod_{j=1}^{i} \alpha_1^{p_j} \alpha_2^{q_j} \beta[m_1 + jm_2, m_2 + (i - j)m_1] < \prod_{j=1}^{i} \beta[m_1 + jm_2, m_2 + (i - j)m_1] \leq \beta_{\max}^i$$
$$(p_j + q_j = 1),$$

where $\beta_{\max} \geq 0$ is the maximal value of the function β in the region (0, m). Thus, for the series (23) there can be constructed the majorant with respect to the variable m:

$$\Omega = \sum_{i=1}^{\infty} t v_0^{i+1} (6\varepsilon t \beta_{\max})^i / i! \tag{24}$$

We see that under the assumption adopted on the existence for the kernel β of a maximum, the series (24) converges in the time interval $0 < t < \infty$, therefore Eq. (24) majorizes the series (23) over this same interval.

As a computational example we shall consider the problem of the evolution of the dispersivity of a fine powder that is suspended in an infinite volume of a quiescent viscous incompressible liquid. The fine powder consists particles of two sorts with the masses m_1 and m_2 and the density ρ_s and is subject to the action of gravity forces and Archimedes and Stokes forces (gravitational coagulation). Then the sedimentation rate of the small particles is calculated from the formula [4].

$$v = 2g\Delta\rho R^2 / (9\mu_l)$$

($\Delta\rho = \rho_s - \rho_l > 0$) or, if $R(m) = [3m/(4\pi\rho_s)]^{4/3}$,

$$v(m) = [2g\Delta\rho / (9\mu_l)][3m/(4\pi\rho_s)]^{2/3}. \tag{25}$$

In this case the kernel of Eq. (1) takes the form

$$\beta_1(m, \mu) = \pi[R(m) + R(\mu)]^2 \, |v(m) - v(\mu)|, \tag{26}$$

characteristic for gradient coagulation problems [5-7]. And in accordance with Eqs. (25), (26)

$$\varepsilon_1 = \frac{2\pi g\Delta\rho}{9\mu_l} \left(\frac{3}{4\pi\rho_s}\right)^{4/3}, \beta(m', m'') = (m'^{1/3} + m''^{1/3})^3 \, | \, m'^{1/3} - m''^{1/3} \, |.$$

Since the kernel β(m', m") for the gravitational coagulation problem is an increasing function of its arguments, for the given problem the convergence of the expansion (6) can not be substantiated with the aid of the majorant.

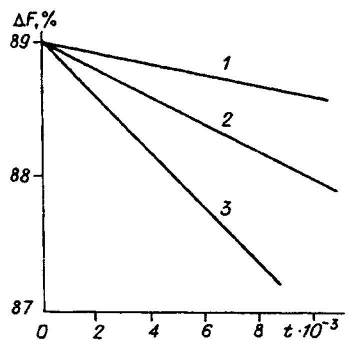

Fig. 1

It we taken as the characteristic magnitudes of the mass, length, and time

$$M = \mu_l^2/(g\Delta\rho),\ L = (\mu_l/\Delta\rho)^{2/3}/g^{1/3},\ T = [\mu_l/(g^2\Delta\rho)]^{1/3},$$

then (in dimensionless variables)

$$\varepsilon = \frac{2\pi}{9}\left(\frac{3}{4\pi}\frac{\Delta\rho}{\rho_S}\right)^{4/3} < 1,\ \nu_0 = N_0\left(\frac{\mu_l}{\Delta\rho}\right)^2\frac{1}{g},$$

$$\beta(m', m'') = (m'^{1/3} + m''^{1/3})^3\,|\,m'^{1/3} - m''^{1/3}\,|.$$

Introducing the total number of particles per unit volume

$$N(t) = \int_0^\infty n(m, t)dm,\ N_0 = \int_0^\infty n^0(m)dm = \nu_0,$$

we obtain the PD of the particles with respect to their masses, normed by N,

$$\Phi(m, t) = n(m, t)/N(t),$$

and also the distribution function

$$F(m, t) = \int_0^m \Phi(\mu, t)d\mu = \frac{1}{N(t)}\int_0^m n(\mu, t)d\mu.$$

Retaining three terms in the expansion (12), we have approximately

$$F(m, t) = \int_0^m [n^0(\mu) + \varepsilon t\varphi_1(\mu) + 0.5(\varepsilon t)^2\varphi_2(\mu)]d\mu.$$

Then we can obtain on the basis of Eq. (26) the change in the relative number of particles of a given size over the time t as a result of their coagulation. For example, the relative change of the number of particles of mass $m < m_2$ is

$$\Delta F = F(m_2)/F(\infty), \tag{27}$$

where

$$F(m_2) = \nu_0\alpha_1(1-\varkappa\alpha_2\beta(m_1,m_2)\{1-0.5\varkappa[(\alpha_1+\alpha_2)\beta(m_1,m_2)-\alpha_1\beta(m_1+m_2,m_1)]\});$$

$$F(\infty) = \nu_0(1 - \varkappa\alpha_1\alpha_2\beta(m_1, m_2)\{1 + 0.5\varkappa[\alpha_1\beta(m_1 + m_2, m_1) -$$
$$(\alpha_1 + \alpha_2)\beta(m_1, m_2) + \alpha_2\beta(m_1 + m_2, m_2)]\}),\ \varkappa = \varepsilon t\nu_0.$$

We shall examine a suspension with the parameters: $\mu_1 = 10^{-3}$ Pa·sec, $\rho_s = 10^3$ kg/m^3, $\Delta\rho = 10^2$ kg/m^3, g = 9.8 m/sec^2, volume concentrations in the starting mixture of each of the solid fractions $c_1 = 5 \cdot 10^4$, $c_2 = 5 \cdot 10^{-4}$, $\alpha_1 = 0.89$, $\alpha_2 = 0.11$ with the relationship between the radii of the particles $R_2 = 2R_1$. Then if $\bar{\varepsilon} = 0.32$ is the coefficient accounting for non-compactness of packing of the spheres [4], the number of particles per unit volume of the starting mixture $N_0 = 3\bar{\varepsilon}(c_1/R_1^3 + c_2/R_2^3)/(4\pi)$. We take as the theoretical process realization time $\tau = 60$ sec (or dimensionless form $\bar{\tau} \approx 10^4$).

It follows from the analysis of the results obtained in accordance with Eq. (27) (see Fig. 1) that the relative content in the mixture as a consequence of the acts of coagulation of the particles of the small fraction, as we would expect, decreases with time, and more rapidly the large the dimension of the particles, although this change over the studied time interval is relatively small (of the order of 2%). In Fig. 1 the lines 1-3 correspond to $R_1 = 5 \cdot 10^{-6}$, 10^{-5}, 25×10^{-6} m and $R_2 = 10^{-5}$, $2 \cdot 10^{-5}$, $5 \cdot 10^{-5}$ m.

The performed quantitative analysis of the coagulation problem was based on the use in deriving the equation (1) of the formula for calculating the geometric probability of collision of the particles, which leads to results that are overstated in comparison with the real results with regard to the number of cohered particles. This is due to neglect in the idealized coagulation kinetics model of phenomenon that is usually present in practice: the flow of the fine particles around the coarse particles [8, 9]. In this connection the kernel of the Smolukhovskii equation is corrected by a capture coefficient in order to refine the calculations [1].

In this case, although we obtain an equation having a more complex structure in comparison with Eq. (1), the quantitative analysis of this equation can in principle be carried out using the same technique used for the relation (1).

REFERENCES

1. V. M. Voloshchuk and Yu. S. Sedunov, Coagulation Processes in Disperse Systems [in Russian], Gidrometeoizdat, Leningrad (1975).
2. V. M. Voloshchuk, Kinetic Theory of Coagulation [in Russian], Gidrometeoizdat, Leningrad (1984).
3. E. Madelung, Mathematical Apparatus of Physics [Russian translation], Nauka, Moscow (1968).
4. J. Happel and H. Brenner, Hydrodynamics at Low Reynolds Numbers [Russian translation], Mir, Moscow (1976).
5. S. D. Grishin, A.-P. Tishin, and R. I. Khairutdinov, "Nonequilibrium two-phase flow in Laval nozzle with coagulation of the polydisperse condensate particles," Izv. Akad. Nauk. SSSR, Mekh. Zhidk. Gaza, No. 2 (1969).
6. A. N. Kraikov and A. A. Shraiber, "On constructing a model describing in the one-dimensional approximation a two-phase flow with coagulation of the polydisperse condensate particles," Prikl. Mekh. Tekh. Fiz., No. 2 (1974).
7. G. L. Babukha and A. A. Shraiber, Interaction of Polydisperse Material Particles in Two-Phase Flows [in Russian], Naukova Dumka, Kiev (1972).
8. N. A. Fuks, Mechanics of Aerosols [in Russian], Izd. Akad. Nauk SSSR, Moscow (1955).
9. L. E. Sternin, B. N. Maslov, A. A. Shraiber, et al., Two-Phase Monodisperse and Polydisperse Gas Flows with Particles [in Russian], Mashinostroenie, Moscow (1980).