PMM U.S.S.R.,Vol.44,pp.491-494 0021-8928/81/4 0491 $7.50/0

UDC 532.529

STATIONARY DISTRIBUTIONS OF PARTICLES BY SIZE IN FINITE COAGULATING SYSTEMS WITH DISINTEGRATION*

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A problem of stationary distribution of particles in a disperse phase according to size, where the particles are subjected to coagulation and disintegration processes, is considered. Analytic expressions are obtained for the mean number of particles of arbitrary mass in a stationary distribution. A limiting passage to an infinite system is studied. One of the basic factors in the mechanics of aerosols, which determines the distribution of the particles according to their size is the process of coagulation of the particles in the disperse phase, caused by their collisions with each other. This is often accompanied by a competing process of division or disintegration of the particles caused by some concrete mechanism (e.g. disintegration of particles in the turbulent pulsations of air, or the instability of droplets under surface deformation /1,2/). The kinetic coagulation equation was first generalized to the case of the systems with decomposition in /3/. A model of formation of precipitation from a warm cloud with the drop disintegration process taken into account, was constructed in /4/, and in /5/ an attempt was made to explain the characteristic form of the stationary spectra of the particles in systems with coagulation and disintegration.

A probabilistic approach to the study of coagulation processes in disperse systems was formulated in /6,7/; the coagulation was regarded, within the framework of this approach, as a Markovian process. Numerical modelling of the coagulation processes was carried out in /8,9/ using the Monte-Carlo method. This approach yielded a number of results not accounted for by the Smoluchowski theory, e.g. the appearance of superparticles /10/. Until now, only a small number of analytic solutions for the probabilistic problems of the coagulation theory have been obtained /7/.

1. We use the Smoluchowski theory (see e.g. /1/) to study the kinetics of the coagulation systems with disintegration of the particles taken into account. The state of the coagulating system at every instant of time is described by an averaged mass spectrum $c_g(t)$ where $c_g(t)$ denotes the concentration of the particles of mass g at the instant t (every particle is assumed to be composed of g monomers of unit mass). The evolution of the spectrum caused by coalescence of the particles following their collision and their disintegration, are described by a kinetic equation of the form

$$\frac{dc_g}{dt} = \frac{1}{2}\sum_{n=1}^{g-1} K(g-n, n)\, c_{g-n}c_n - c_g \sum_{n=1}^{\infty} K(g, n)\, c_n + \sum_{n=1}^{\infty} (1+\delta_{g,n})\, L(g, n)\, c_{g+n} - \frac{1}{2} c_g \sum_{n=1}^{g-1} (1+\delta_{n,g-n})\, L(g-n, n) \tag{1.1}$$

Here $K(g, n)$ is the probability of coagulation occurring per unit time, in a unit volume, $L(g, n)$ is the probability of disintegration of a particle and $\delta_{g,n}$ is the Kronecker delta. We shall consider a particular case when L and K are connected by the following relation:

$$(1+\delta_{l,m})\, \varphi_l \varphi_m L(l, m) = \varphi_{l+m} K(l, m) \tag{1.2}$$

where φ_l is an arbitrary positive function of natural argument and $\varphi_1 = 1$.

2. In recent years a more general approach to the problem of describing the coagulating systems has been developed /6,7/, based on the same physical assumptions as the Smoluchowski theory. A homogeneous space of volume V filled with particles is studied. Every state of this system is characterized by a mass spectrum

$$Q = \{n_1 \ldots, n_l \ldots, n_m \ldots, n_g \ldots, n_M\}$$

where n_g is the number of particles composed of g monomers (filling numbers). Every state Q has an associated, time-dependent probability $W(Q, t)$. It is assumed that the particles move chaotically under the action of certain random forces and either disintegrate, or collide and coalesce. A single collision causes a coalescence of two particles and, as a result, the system changes its state to

*Prikl.Matem.Mekhan.,44,No.4,697-701,1980

$$Q^+ = \{n_1\ldots, n_l - 1\ldots, n_m - 1\ldots, n_g + 1\ldots, n_M\},\ g = l + m$$

the mass spectrum of which differs from the previous one by just three filling numbers (two if identical particles collide). The disintegration corresponds to a reverse process $Q^+ \to Q$. If the rates of transitions $Q \to Q^+$, $Q^- \to Q$ $((Q^-)^+ \equiv Q)$ are known, then the equation for $W(Q, t)$ is obtained in the form

$$\frac{dW(Q,t)}{dt} = \sum_{Q^-} A(Q, Q^-) W(Q^-, t) - W(Q, t) \sum_{Q^+} A(Q^+, Q) + \sum_{Q^+} B(Q, Q^+) W(Q^+, t) - W(Q, t) \sum_{Q^-} B(Q^-, Q) \quad (2.1)$$

Expressions for the rates of transition $A(Q, Q^-)$ and $B(Q^+, Q)$ are obtained in terms of $K(l, m)$ and $L(l, m)$ from the following combinatorial expressions:

$$A(Q, Q^-) = K(l, m)\, n_l(Q^-)\,[n_m(Q^-) - \delta_{l,m}] / V(1 + \delta_{l,m}), \quad B(Q, Q^+) = n_{l+m}(Q^+)\, L(l, m)$$

We use the expression $n_g(Q)$ to indicate that the number n_g belongs to the state Q. The total mass of all particles

$$\sum_g g n_g = M$$

is preserved in the coagulation and disintegration processes.

The Smoluchowski theory is obtained from the proposed approach as a result of the passage to the thermodynamic limit (volume and mean filling numbers both tending to infinity with their ratio remaining finite). The passage was studied in detail in /7/. A stationary regime is feasible in the systems with disintegration. This can be obtained from the equation (2.1) by putting $dW/dt = 0$. Let us investigate the stationary solutions in the case when the rate of disintegration is connected with the coagulation coefficients by the relation (1.2). This occurs e.g. when a collision between the drops leads to formation of an excited drop with excess energy, the latter causing its subsequent disintegration. Since during the disintegration particles of mass different from l and m may form, this case cannot be reduced to slow coagulation.

If the relation (1.2) holds, then, taking into account (2.1) we obtain a stationary solution of the form

$$W(Q) = \frac{M!\, W_1(M)}{V^M} \prod_{l=1}^{M} \frac{V^{n_l}}{n_l!\, \varphi_l^{n_l}} \quad (2.2)$$

where $W_1(M)$ describes the probability of occurrence of the configuration $\{M, 0, 0 \ldots, 0\}$. The relation (2.2) follows from the equality

$$A(Q, Q^-) W(Q^-) = B(Q^-, Q) W(Q) \quad (2.3)$$

Indeed

$$W(Q^-) A(QQ^-) = \frac{K(l, m)\, n_l(Q^-)\,[n_m(Q^-) - \delta_{l,m}]\, M!\, W_1(M)}{(1 + \delta_{l,m})\, V^{M+1}} \times \prod_{\nu=1}^{M} \frac{V^{n_\nu(Q^-)}}{n_\nu(Q^-)!\, \varphi_\nu^{n_\nu}(Q^-)}$$

Taking into account the fact that $n_l(Q^-) = n_l(Q) + 1$, $n_m(Q^-) = n_m(Q) + 1$, and the remaining $n_\nu(Q^-) = n_\nu(Q)$, we obtain (2.3). We show in the same manner that $B(Q, Q^+) W(Q^+) = A(Q^+, Q) W(Q)$ for any pair of configurations Q, Q^+. Thus the solution (2.2) secures not only the integral balance of the probabilities for an arbitrary configuration Q, but also the equality of the probability fluxes for any pair of configurations Q_1, Q_2. To complete the determination of $W(Q)$ we require to find $W_1(M)$. Taking into account the equation

$$\sum_Q W(Q) = 1, \quad \sum_{Q,\nu} \nu n_\nu(Q) W(Q) = M$$

we obtain

$$M P_M = V\left[\sum_{\nu=1}^{M-1} \frac{\nu P_{M-\nu}}{\varphi_\nu} + \frac{M}{\varphi_M}\right], \quad P_l \equiv \frac{V^l}{l!\, W_1(l)} \quad (2.4)$$

We solve the recurrence relation (2.4) for P_l using the generating function

$$f(z) = \sum_l z^l P_l$$

From (2.4) we obtain the following expression for $f(z)$:

$$f(z) = \exp\left(V \sum_{n=1}^{\infty} \frac{z^n}{\varphi_n}\right) - 1 \quad (2.5)$$

Let us consider the stationary spectrum

$$c_g{}^S = \frac{\bar{n}_g}{V} \equiv \frac{1}{V} \sum_Q W(Q)\, n_g(Q)$$

Following the derivation of (2.4), we find

$$c_g{}^S = P_{M-g} / (\varphi_g P_M) \quad (2.6)$$

In order to pass to the infinite systems, we must find the asymptotics of P_M as $M \to \infty$. For P_M given by (2.4), we have

$$P_M = \frac{1}{2\pi i} \oint \frac{dz}{z^{M+1}} \exp\left[V \sum_{n=1}^{\infty} \frac{z^n}{\varphi_n}\right]$$

Here the integration is carried out along the contour encircling the coordinate origin in the complex z-plane, in the anticlockwise direction. Estimating the value of the integral with help of the saddle point method, we obtain

$$P_M \simeq x_0^{-1} (\psi''(x_0) / 2\pi)^{1/2} \exp \psi(x_0), \quad \psi(x) = V \sum_{n=1}^{\infty} \frac{x^n}{\varphi_n} - M \ln x$$

where x_0 denotes the point at which $\psi'(x) = 0$. From the expression for ψ we find, that the saddle point is given by the equation

$$\sum_{n=1}^{\infty} \frac{n x_0^n}{\varphi_n} = \rho, \quad \rho \equiv M / V \tag{2.7}$$

where ρ is regarded as the mass of the particles per unit volume. Then we have

$$P_M = (2\pi M)^{-1/2} x_0^{-M} \exp\left(V \sum_{n=1}^{\infty} \frac{x_0^n}{\varphi_n}\right) \times \left[1 + \frac{1}{\rho} \sum_{n=1}^{\infty} \frac{n(n-1) x_0^n}{\varphi_n}\right]^{1/2}$$

and this yields the mass spectrum in an infinite system

$$c_g^S = x_0^g / \varphi_g \tag{2.8}$$

To illustrate this, we consider some concrete examples of the function φ_g.

1°. $\varphi_g = g$. From (2.5) follows $f(z) = (1-z)^{-V} - 1$, i.e.

$$P_l = V(V+1) \ldots (V+l-1) / l!$$

The quantity x_0 is found from (2.7), which in this case assumes the form

$$\sum_{n=1}^{\infty} x_0^n = x_0 / (1 - x_0) = \rho$$

For the stationary spectrum in an infinite system we have

$$c_g^S = g^{-1} [\rho / (\rho + 1)]^g$$

2°. $\varphi_g = b^{g-1}$. In this case we obtain for $f(z)$ and P_M

$$f(z) = \exp\left(\frac{Vbz}{b-z}\right) - 1, \quad P_M = V^M \sum_{n=0}^{M-1} \binom{M-1}{n} \frac{(Vb)^{-n}}{(M-n)!}$$

From (2.7) we obtain, for x_0

$$b \sum_{n=1}^{\infty} n \left(\frac{x_0}{b}\right)^n = \rho, \quad \frac{x_0}{b} \equiv \gamma = \left(1 + \frac{b}{2\rho}\right) - \left(\frac{b^2}{4\rho^2} + \frac{b}{\rho}\right)^{1/2}$$

The quantity $\gamma < 1$, $\gamma \to 0$ as $\rho \to 0$ and $\gamma \to 1$ when $\rho \to \infty$. The stationary spectrum in an infinite system has the form

$$c_g^S = b\gamma^g$$

3. Let us analyze the connection between the stationary spectrum in an infinite system, and the stationary $(d/dt = 0)$ solutions of the Smoluchowski equation (1.1) for the systems with disintegration. When the relation (1.2) holds, the solution (2.8) is an exact solution of the stationary equation (1.1) for any values of x_0. To choose x_0, we must take into account the law of conservation of mass per unit volume

$$\sum_g g c_g^S = \rho$$

and this makes natural the choice of x_0 from the relation (2.7). We must remember that the stationary regime is realized for any value of the density, provided that the series

$$u(x) = \sum_{n=1}^{\infty} \frac{n x^n}{\varphi_n}$$

has a nonzero radius of convergence. In this case $u(0) = 0$ and $u(x)$ increases faster than x, i.e. the equation $u(x) = \rho$ has a unique positive solution x_0. Since the series is summed at the point x_0 (its sum is equal to ρ), it follows that the quantities $g c_g^S$ are also summable.

In conclusion we indicate that the problem of coagulation with disintegrations in finite systems is equivalent to the problem of dynamics of a cyclic structure of an element of a group of substitutions under the action of random transpositions (see /11/).

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Translated by L.K.