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FLUX METHOD IN THE KINETICS OF COAGULATION

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UDC 54-138

Coagulation causes broad fluctuations in the concentration and size distribution of aerosol particles [1]. Measured particle-radius distributions are dome-shaped. The top part of the dome is usually described by the so-called log-normal distribution. The right side may descend significantly slower than the left side in accordance with a power law [2]. A power spectrum was observed for an atmospheric aerosol in [3]. Later on it was explained on the basis of a representation of constant mass flux over the particle spectrum. The form of the spectrum follows from dimensional considerations with the use of the locality hypothesis [4, 5], to within the accuracy of a coefficient. A stationary spectrum was obtained in [6] on the basis of a kinetic equation. Stationary power spectra with thermal and gravitational coagulation in different ranges of particle radius were obtained in [7] along with coefficients. It was shown in [8] that these results follow from a more general analysis of the kinetic equation with the use of the notions of fluxes of particles and mass over the spectrum. However, until now there has been no direct kinetic determination of the flux, which is important in the theory of coagulation and in certain other similar problems.

This article explicitly determines the flux of the number and volume of particles (drops) over the spectrum corresponding to the physical significance of these quantities. This

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approach (in large part similar to that developed earlier in oscillatory kinetics [9]) immediately leads to stationary power distributions and can prove useful in analyzing more complex problems. The other problem examined is the evolution of the log-normal distribution. It is shown that the fluctuation in the number of drops over the spectrum is characterized by a finite effective mean free path, so that it is possible to use equations of the Fokker–Planck type. We introduce a logarithmic particle-radius scale in which the equation takes a form permitting an analogy with the motion of a gas. The rate of propagation of the maximum of the distribution is calculated. Normalization conditions are used to find the time dependence of parameters of the log-normal distribution.

1. We will examine how particles and volume change over a spectrum. The result of the union of drops 1 and 2 of radii $r_1 < r_2$ is conveniently represented as follows: the first drop disappears, while the second is shifted along the radius to point 3 in accordance with the law of volume conservation $r_3^3 = r_1^3 + r_2^3$. The shifting of a large drop makes a contribution to the flux in the number of drops in a space of radii j. The shift is greatest when drops of the same size unite: $r_3 = 2^{1/3} r_2$. The quantity $(1/3) \ln 2$ plays the role of the characteristic path length of the particles in the logarithmic scale of radii $\ln r$. The disappearance of the smaller particle merging with a larger particle represents a contribution to the discharge C.

The above interpretation makes it possible to write the coagulation equation in the form of a conservation law:

$$\partial n/\partial t = -\partial j/\partial r - C, \tag{1.1}$$

where

$$j(r) = \int\int_D K(r_1, r_2)\, n(r_1)\, n(r_2)\, dr_1 dr_2; \tag{1.2}$$

$$C(r) = n(r) \int_r^\infty K(r, r_1)\, n(r_1)\, dr_1. \tag{1.3}$$

Here n(r) is the density of the number of drops in the radius space; t is time; $K(r_1, r_2)$ is the coagulation rate coefficient. The region of integration D in (1.2) is determined by the inequalities $r_1 < r_2 < r$ and $r_1^3 + r_2^3 > r^3$ (Fig. 1). The coefficient of the rate of thermal (Brownian) coagulation for particles with radii greater than 0.1 μm has the form [1]

$$K(r_1, r_2) = (2kT/3\eta)(2 + r_1/r_2 + r_2/r_1), \tag{1.4}$$

where T is temperature; η is the viscosity of the gas; k is the Boltzmann constant.

The integral (1.2) over the region D can be represented as the difference between the integrals over the regions $D'(r_1 < r_2 < r)$ and $D''(r_1 < r_2, r_1^3 + r_2^3 < r^3)$. Then the derivative $\partial j/\partial r$ is written in the form

$$\frac{\partial j}{\partial r} = n(r) \int_0^r K(r_1, r)\, n(r_1)\, dr_1 - \int\int_{r_1 < r_2} K(r_1, r_2)\, n(r_1)\, n(r_2)\, \delta\left[(r_1^3 + r_2^3)^{1/3} - r\right] dr_1 dr_2.$$

Insertion of this expression into (1.1) reduces the coagulation equation to standard form (see [2], for example).

The flux in the number of particles j, generally speaking, is nonlocal. However, in a scale which is large compared to the characteristic path length of the particles in the radius space, the flux j can be considered local. This makes it possible to change Eq. (1.1) to the form of a volume conservation law.

Multiplying (1.1) by the volume of a drop v(r), we obtain an equation for the density of the volume vn on the radius axis:

$$\frac{\partial (vn)}{\partial t} = -\frac{\partial}{\partial r}(vj) + v'j - vC, \tag{1.5}$$

where the prime denotes a derivative with respect to r. If we regard the flux j as local, then vj is convective flux of the volume connected with the flux in the number of drops; v'j and vC are the volume source and sink. Since drop volume is conserved during coagulation, the difference between the source and sink can be represented in the form of the divergence of the flux of the transferred volume:

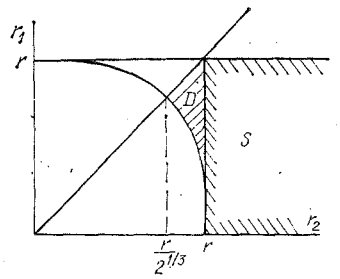

Fig. 1

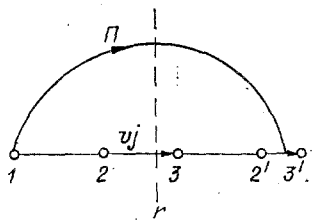

Fig. 2

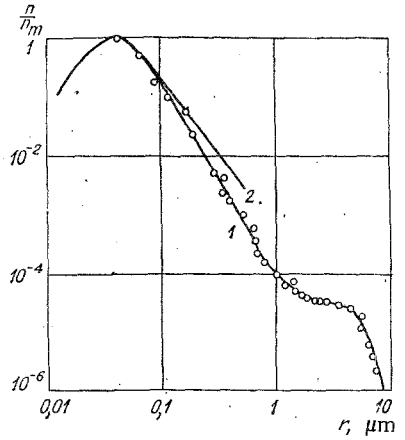

Fig. 3

$$v'j - vC = -\partial\Pi/\partial r. \tag{1.6}$$

The flux of the volume transferred to the section r of the radius axis is determined by the expression

$$\Pi(r) = \iint\limits_{r_1<r<r_2} v(r_1) K(r_1, r_2) n(r_1) n(r_2)\, dr_1 dr_2. \tag{1.7}$$

The region of integration in (1.7) is designated by S in Fig. 1. The flux $\Pi(r)$ corresponds to the transfer of a volume across the section r in the radius space without the intersection of r by the particles, in contrast to convective flow vj (Fig. 2).

With the substitution of Eq. (1.6), Eq. (1.5) takes the form of a continuity equation

$$\partial(vn)/\partial t = -\partial F/\partial r,\ F = vj + \Pi, \tag{1.8}$$

where F is the total flux of volume in the radius space. Thus, the drop volume is transferred in the radius space in the form of a flux F consisting of two parts: convective flow of the volume and flow of the transferred volume.

Continuity equation (1.8) makes sense only in a scale which is large compared to the characteristic path length on the radius axis, equal in the logarithmic scale $\ln r$ to $(1/3) \times \ln 2$. The essence of the matter is that the transferred volume cannot be localized exactly on the radius axis, since it is spread over the path length. In fact, in the process (1) + (2') → (3') the total volume in the interval (r_2', r_3') (but not at point 3') increases by v_1. It should be noted that Eq. (1.6) exists with the same accuracy (if the path length is assumed to be small). We can assure ourselves of this indirectly by calculating the derivative $\partial\Pi/\partial r$ by determining (1.7).

2. In the stationary case, the flux F is equal to the productivity of the source

$$F = vj + \Pi = \text{const}. \tag{2.1}$$

We will seek a power solution to Eq. (2.1) $n \sim 1/r^m$. Using the homogeneity of the kernel (1.4), we represent the flux in the form

$$F = vn^2r^2(Q_m + P_m), \tag{2.2}$$

where Q_m and P_m depend only on the exponent m

$$Q_m = \iint\limits_{\substack{0<x_1<x_2<1 \\ x_1^3+x_2^3>1}} K(x_1, x_2) \frac{dx_1 dx_2}{x_1^m x_2^m}; \tag{2.3}$$

$$P_m = \iint\limits_{0<x_1<1<x_2} K(x_1, x_2) \frac{dx_1 dx_2}{x_1^{m-3} x_2^m}. \tag{2.4}$$

The convective flux and the flux of the transferred volume are, respectively, proportional to Q_m and P_m.

Inserting (2.2) into (2.1) and considering that $v \sim r^3$, we obtain

$$n(r) \sim 1/r^{5/2}. \tag{2.5}$$

It is easily seen that the integrals (2.3) and (2.4) agree with m = 5/2. The first reduces to a single integration, which is done numerically. The integral (2.4) is calculated as elementary. The numerical values of the coefficients $Q_{5/2} \approx 1.21(2kT/3\eta)$ and $P_{5/2} = 5.05 \times (2kT/3\eta)$. Thus, the main contribution to the volume flux is made by the flux in the transferred volume II. Inserting the quantities $Q_{5/2}$ and $P_{5/2}$ into (2.2) gives us the following equation, in accordance with [7, 8]

$$n(r) = 0.24(F\eta/kTr^5)^{1/2}. \tag{2.6}$$

In reality the distribution (2.6) may exist on a finite interval of radii. If the region of the sources $r < a$ and the region of the sink $r > b$, then the region of existence of distribution (2.6) is limited by the condition $a \ll r \ll b$.

For drops with radii greater than 1 μm, gravitational coagulation plays an important role. The coefficient of the rate of gravitational coagulation is represented by a fourth-order homogeneous function of the radii [1]. Here, the volume flux in the radius space $F \sim r^6 v\eta^2$. From this, we find that the stationary distribution is gravitational coagulation $n \sim r^{-9/2}$. The complete distribution for this case is presented in [7, 8].

3. We will examine the problem of the evolution of a nonstationary dome-shaped drop-radius distribution which quickly falls off for small and large radii. We will examine the neighborhood of the maximum of the distribution, at which most of the drops are distributed. Here, Eq. (1.1) can be simplified by employing the slight change of the distribution in the neighborhood of the maximum.

We will show that Eq. (1.2) for j can be represented in simple algebraic form. In the region of integration r_2 changes within the range from $r/2^{1/3}$ to r, which is equal to the characteristic path length of the particles in the radius space (Fig. 1). Assuming the change in n(r) in this space to be small, we move the multiplier $n(r_2)$ out from under the integral sign. We then note that a large r_1 corresponds to a long path length, so the neighborhood $r_1 \sim r$ makes the main contribution to the integral. Thus, the multiplier $n(r_1)$ can also be moved from under the integral at point r. Using the homogeneity of the kernel (1.4), we obtain

$$j \simeq Q_0 r^2 n^2, \tag{3.1}$$

where Q_0 is determined by Eq. (2.3) with m = 0. Calculation of the integral gives $Q_0 = J \times (2kT/3\eta)$, $J \approx 0.212$.

Inserting (3.1) and (1.3) into (1.1) gives us the equation

$$\frac{\partial n}{\partial t} = -Q_0 \frac{\partial}{\partial r} r^2 n^2 - n \int_r^\infty K(r, r_2)\, n(r_2)\, dr_2. \tag{3.2}$$

We introduce the logarithmic radius scale $y = \ln(r/r_0)$, where r_0 is a certain scale. We designate the density of the drops in this scale as $u(y) = n(r)/(dy/dr) = rn(r)$. We introduce another new time scale $\tau = 2Q_0 t$. A substitution of variables reduces Eq. (3.2) to the form

$$\frac{\partial u}{\partial \tau} + u \frac{\partial u}{\partial y} = -u \int_y^\infty R(y, y_1)\, u(y_1)\, dy_1, \tag{3.3}$$

where

$$R(y, y_1) = (1/J)[1 + \operatorname{ch}(y - y_1)]. \tag{3.4}$$

We should note the evident analogy between Eq. (3.3) and the equations of motion of a gas. The left side of Eq. (3.3) can be represented in the form of a derivative along the characteristic $dy/d\tau = u$, being the trajectory of motion with the velocity u:

$$\frac{d \ln u}{d\tau} = -\int_y^\infty R(y, y_1) u(y_1)\, dy_1. \tag{3.5}$$

Equation (3.5) describes the decrease in velocity u along the characteristic.

The velocity decrease is smallest in the forward part of the moving velocity profile u(y). If the decrease is small, the curvature of the profile will increased. Thus, the faster-moving parts, coming from behind, accelerate the forward parts. As a result, the

analogy with gasdynamic motion is manifest in the possibility of the formation of a steep leading edge on the moving profile.

We introduce the rate of propagation of the maximum of the profile

$$u_0 = dy_m/d\tau, \tag{3.6}$$

where y_m is determined by the equation $\partial u/\partial y = 0$. Differentiating the latter equation along the trajectory of the maximum $y_m(\tau)$, we obtain

$$(\partial^2 u/\partial\tau\partial y)_m + u_0 (\partial^2 u/\partial y^2)_m = 0. \tag{3.7}$$

Differentiating with respect to y, we obtain the following from Eq. (3.3) for $\partial u/\partial y = 0$

$$\left(\frac{\partial^2 u}{\partial\tau\partial y}\right)_m + u_m \left(\frac{\partial^2 u}{\partial y^2}\right)_m = \frac{\dot{u}_m}{J}\left(2u_m - \int_{y_m}^{\infty} \operatorname{sh}(y_m - y_1)\, u(y_1)\, dy_1\right). \tag{3.8}$$

Excluding the time derivative, we obtain an expression for u_0 from (3.7) and (3.8)

$$u_0 = u_m\left(1 - \frac{2u_m + \int_{y_m}^{\infty} \operatorname{sh}(y_1 - y_m)\, u(y_1)\, dy_1}{J(\partial^2 u/\partial y^2)_m}\right). \tag{3.9}$$

In accordance with (3.9), the rate of propagation of the velocity profile maximum exceeds the value of the maximum u_m [since $(\partial^2 u/\partial y^2)_m < 0$]. This phenomenon of "advance" propagation of the maximum is due to an increase in the decay of velocity from the front to the rear part of the profile.

Substituting a Gaussian distribution near the maximum u(y)

$$u(y) \simeq u_m \exp\left[-a(y - y_m)^2\right], \tag{3.10}$$

from (3.9) we obtain an estimate of the rate of propagation across the maximum and the parameter a, determining the width of the profile

$$u_0 \simeq u_m\left\{1 + \frac{1}{J}\left[\frac{1}{a} + \frac{\sqrt{\pi}}{4}\frac{e^{1/4a}}{a^{3/2}}\Phi\left(\frac{1}{2\sqrt{a}}\right)\right]\right\}, \tag{3.11}$$

where $\Phi(1/2\sqrt{a})$ is the error integral.

Equations (3.6) and (3.10), together with the law of change in the total number of drops over time

$$N = u_m\sqrt{\Pi/a}$$

and the condition of conservation of the total volume of the drops

$$V/V_0 = N\exp(3y_m + 9/4a), \quad V_0 = 4\pi r_0^3/3,$$

makes it possible to find the time dependence of all of the parameters of the Gaussian distribution (3.10).

With large τ, asymptotically $a \sim$ const, $y_m \sim (1/3)\ln\tau$ + const, and $u_m \sim 1/\tau$.

4. In conclusion, we will briefly discuss the results of the experiment in [10], where a study was made of the radius distribution of drops of Al_2O_3 in a flame during the combustion of a metallized fuel. Samples were taken with a filter, and combustion products that were rising and moving laterally during convection passed through the filter. Solidified spherical particles of aluminum oxide that accumulated in the filter were later analyzed under an electron microscope. Here, the spectrum of the drops (curve 1 in Fig. 3) was averaged over the time of the experiment. Judging from the character of the spectrum, in the region of about 0.1 μm the spectrum may be governed by steady thermal coagulation of drops with an initial radius no less than 0.04 μm. An estimate of the relaxation time $\sim rnv/F$ also follows this probability. Figure 3 (curve 2) shows the power spectrum (2.5), which approximates the experimental spectrum in its middle part well.

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DEPOSITION OF SMALL AEROSOL PARTICLES ON THE SURFACE OF MOVING EVAPORATING CRYSTALS

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UDC 532.73:551.510

The method of augmenting asymptotic expansions is used for the case of low diffusive Peclet numbers to determine the flow of aerosol particles to the surface of an evaporating (or growing) crystal.

1. Formulation of the Problem. The theory of capture of small (moving in the free-molecular regime) aerosol particles by evaporating or condensing drops has by now been developed in quite an amount of detail [1-3]. As concerns processes of the capture of aerosols particles by evaporating or growing crystals, the theory is considerably less well developed.

A characteristic feature of particles of the solid phase (collectors) is their nonspherical form, which is considered in the present study.

We will examine a large (Knudsen number Kn = 0) evaporating particle of a solid phase suspended in a vapor–gas mixture. The theoretical analysis will be made for the case when the Reynolds number and the diffusive and thermal Peclet numbers are small, so that the equations of hydrodynamics and heat and mass transfer near the particle surface have the form

$$\nu\Delta\mathbf{v} = -\nabla p/\rho_e,\ \operatorname{div}\mathbf{v} = 0,\ \Delta T_{e,i} = 0,\ \Delta c_1 = 0, \qquad (1.1)$$

where $\mathbf{v}$, ρ_e, p, and T_e are the velocity, density, pressure, and temperature of the mixture; $c_1 = n_1/n_0$; $n_0 = n_1 + n_2$ (n_1 and n_2 are the concentrations of the vapor and gas); ν is the kinematic viscosity of the mixture; T_i is the particle temperature.

System (1.1) must be solved with allowance for the following conditions on the boundary between the particle (collector) and the medium:

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