Equation for Brownian coagulation of aerosol particles in a stochastic medium

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The Brownian coagulation of highly dispersed aerosol particles in a stochastic medium with small velocity fluctuations is analyzed. The fluctuations of the velocity field are assumed to be Gaussian with a uniform pair correlation function. Exact equations are found for the mean field of the nonuniform size distribution function of the particles. An effective Brownian-coagulation equation is constructed for a velocity correlation function of a specific type. The relationship between the turbulent diffusion coefficient and the effective coagulation kernel is determined.

1. INTRODUCTION

Coagulation is the predominant mechanism for the transformation of the size distribution of highly dispersed aerosol particles in many natural and industrial disperse systems.[1-3] In general, the theoretical work on this mechanism has been carried out in the approximation of a coagulation kinetic equation, which is written for the case of a homogeneous medium as

$$\partial f/\partial t=\{f,f\}. \tag{1}$$

Here

$$\{f,f\}=\frac{1}{2}\int_0^V S(V-V',V')f(t,V-V')f(t,V')dV' - f(t,V)\int_0^\infty S(V,V')f(t,V')dV'$$

is the particle collision integral, and $f(t, V)$ is the size (or volume) distribution function of the particles, which is related to the number of particles in the system by

$$n(t)=\int_0^\infty f(t,V)dV.$$

The symmetric function $S(V_1,V_2)$ is the "coagulation kernel." It characterizes the probability for the coagulation of two particles and is determined by the particular features of the interaction of these particles in the medium. In the case of a Brownian coagulation, the following expression is ordinarily used for $S(V_1,V_2)$:

$$S(V_1,V_2)=4\pi(R_1+R_2)[D(V_1)+D(V_2)], \tag{2}$$

where R_α and $D(V_\alpha)$ are respectively the radius and Brownian diffusion coefficient of particle α.

An important question in coagulation theory is that of dealing with the effect of turbulent fluctuations of the dispersion medium on the evolution of the particle size distribution. A turbulence can evidently change the nature of the relative motion of particles, so most studies have been aimed at a description of the microscopic physics of the closing of two particles on each other in a turbulent medium with the goal of determining the corresponding coagulation kernel.[3] It has usually been assumed that the medium is homogeneous.

Examining possible mechanisms for the effect of turbulence on coagulation, Voloshchuk and Sedunov[1] singled out in particular phenomena associated with a nonuniformity of the distribution function. They mentioned that fluctuations of the distribution function should in general increase the rate of coagulation and lead to a more rapid appearance of large particles.

The equation for the coagulation of aerosol particles in a stochastic medium was first derived, under several simplifying assumptions, in Ref. 4. However, the corresponding stochastic equation was not solved there, even in that simplified formulation of the problem. Consequently, corrections describing turbulent diffusion and coagulation were expressed in terms of the Green's function of the fluctuating component of the distribution function in a formal way, so it was not possible to derive an explicit expression for the turbulent diffusion coefficient or for the effective coagulation kernel. It is also important to note that the term describing the Brownian diffusion of aerosol particles was omitted from the outset in that paper.

In the present paper we carry out a systematic averaging of the equation for nonuniform Brownian coagulation over the ensemble of realizations of the stochastic velocity field of the medium, using a generating-functional method. This approach makes it possible to derive an equation for the stochastic coagulation of highly disperse aerosol particles in the approximation of a weak turbulence under the most general initial assumptions. We treat the aerosol particles as a passive impurity; this is a good approximation for Brownian particles.[5] In addition, we assume, as in Ref. 4, that the coagulation kernel $S(V_1,V_2)$ is independent of the turbulence fluctuations. Note that the calculations below impose no restriction on the function $S(V_1,V_2)$, but for definiteness we will restrict the discussion to the case of Brownian coagulation.

2. GENERATING FUNCTIONAL OF THE DISTRIBUTION-FUNCTION FIELD

Let us consider the kinetic equation for the nonuniform Brownian coagulation of particles in a stochastic medium:

$$\partial f/\partial t-D(V)\nabla_r^2 f+\mathbf{u}\nabla_r f-\{f,f\}=0. \tag{3}$$

The function $f(t, \mathbf{r}, V)$ in (3) is a nonuniform size distribution function of the particles, and $\mathbf{u}(t,\mathbf{r})$ is the hydrodynamic velocity of the medium. This velocity can be written as the additive sum of a regular component and a fluctuating component:

$$\mathbf{u}(t,\mathbf{r})=\langle\mathbf{u}(t,\mathbf{r})\rangle+\delta\mathbf{u}(t,\mathbf{r}). \tag{4}$$

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For simplicity we assume $\langle \mathbf{u} \rangle = \text{const}$. Here and below, the angle brackets ($\langle ... \rangle$) mean an average over an ensemble of realizations of the stochastic field **u**. For the analysis below it is convenient to formally introduce a deterministic source density $\hat{\eta}(t, \mathbf{r}, V)$ and to transform to an equivalent form of the collision integral:

$$\{f, f\} = -\int dV_1' dV_2' P(V, V_1', V_2') f(V_1') f(V_2'), \qquad (5)$$

where

$$P(V_1, V_2, V_3) = {}^1\!/_2[\delta(V_1 - V_2) + \delta(V_1 - V_3) - \delta(V_1 - V_2 - V_3)] S(V_2, V_3).$$

Using this result, we can rewrite Eq. (3) in the comoving coordinate system as follows:

$$\partial f(V)/\partial t - D(V) \nabla_r^2 f(V) + \delta\mathbf{u} \nabla_r f(V) + \int dV_1' dV_2' P(V, V_1', V_2') f(V_1') f(V_2') = \hat{\eta}(V). \qquad (6)$$

Introducing the simplifying notation

$$(t, \mathbf{r}, V) = (1), \quad \nabla_{r_1} = \nabla_1, \quad \nabla_{r_1}^2 = \nabla_1^2,$$
$$\delta(t_1 - t_2)\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(V_1 - V_2) = \delta(1-2),$$
$$(\partial/\partial t_1 - D(V_1)\nabla_1^2)\delta(1-2) = L_0(1, 2),$$
$$\delta\mathbf{u}(1)\nabla_1\delta(1-2) = u(1, 2),$$
$$\delta(t_1 - t_2)\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(t_1 - t_3)\delta(\mathbf{r}_1 - \mathbf{r}_3) P(V_1, V_2, V_3) = \mathcal{F}(1, 2, 3),$$

we can put Eq. (6) in a compact form which is convenient for the analysis below:

$$L_0(1, 1') f(1') + u(1, 1') f(1') + \mathcal{F}(1, 1', 2') f(1') f(2') = \hat{\eta}(1). \qquad (7)$$

As usual, a repeated index means an integration.

From the symmetry property of the coagulation kernel $S(V_2, V_3)$ and from the definition of the function $P(V_1, V_2, V_3)$ follows a symmetry property of the function $\mathcal{F}$.

$$\mathcal{F}(1, 2, 3) = \mathcal{F}(1, 3, 2).$$

Equation (7) describes the individual realizations of a stochastic field of a distribution function. From the mathematical standpoint, this equation is a differential equation for the scalar field f with a random coefficient u and a nonlocal quadratic nonlinearity. Problems of this type can be reformulated through the use of the concept of a generating functional and its path-integral representations. Formally, this procedure reduces the description of the classical stochastic problem to a quantum field theory, so one can construct a perturbation theory and derive corresponding Dyson and Schwinger equations. In contrast with the well-developed case of an equation with a random source,[6,7] the equation under consideration here contains a random coefficient of the field function. This circumstance should of course lead to a new type of generating functional.

Let us go through this procedure for the random field f, whose individual realizations are described by Eq. (7). By definition the generating functional is

$$W[\eta, \hat{\eta}] = \langle \exp(i\eta(1') f(1')) \rangle.$$

The cumulative mean fields f are determined in terms of $\ln W$ with the help of the operation of variational differentiation:

$$\langle f(1) \ldots f(N) \rangle = \left(\frac{\delta}{i\delta\eta(1)} \cdots \frac{\delta}{i\delta\eta(N)} \ln W[\eta, \hat{\eta}] \right)_{\eta=0}.$$

Writing the definition of the generating functional, we have

$$W[\eta, \hat{\eta}] = \langle \exp(i\eta f) \rangle = \int Df \mathcal{P}[f, \hat{\eta}] \exp(i\eta f),$$

where $\mathcal{P}[f, \hat{\eta}]$ is the probability density of state f in the presence of an external field $\hat{\eta}$, and Df means a functional integration over the field f. For brevity here and below, we omit the arguments from the functions and operators where it is possible to do so without causing any confusion. The probability density $\mathcal{P}$ is expressed in terms of the mean of the δ-functional over individual realizations of the stochastic field f:

$$\mathcal{P}[f, \hat{\eta}] = \langle \delta(f - \tilde{f}[u, \hat{\eta}]) \rangle,$$

where the individual realization $\tilde{f}[u, \hat{\eta}]$ is a solution of Eq. (7) with the corresponding u and $\hat{\eta}$.

Using the formal relation

$$\delta(f - \tilde{f}) = \det\left[\frac{\delta}{\delta f}(L_0 f + uf + \mathcal{F}ff - \hat{\eta})\right] \delta(L_0 f + uf + \mathcal{F}ff - \hat{\eta})$$

for the δ-functional and its representation as a Fourier functional integral,

$$\delta(L_0 f + uf + \mathcal{F}ff - \hat{\eta}) = \int D\hat{f} \exp[i\hat{f}(L_0 f + uf + \mathcal{F}ff - \hat{\eta})]$$

we find the following expression for W:

$$W[\eta, \hat{\eta}] = \left\langle \int D\hat{f}\, Df \det\left[\frac{\delta}{\delta f}(L_0 f + uf + \mathcal{F}ff - \hat{\eta})\right] \times \exp[i\hat{f}(L_0 f + uf + \mathcal{F}ff - \hat{\eta})] \right\rangle. \qquad (8)$$

We can show that in this case the determinant in (8) is a constant. For this purpose we note that we have

$$\det\left[\frac{\delta}{\delta f}(L_0 f + uf + \mathcal{F}ff - \hat{\eta})\right]$$
$$= \det\{L_0(1, 1')[\delta(1'; 2) + L_0^{-1}(1'; 2')(u(2'; 2) + 2\mathcal{F}(2', 3', 2) f(3'))]\}$$
$$= \det L_0 \cdot \exp\{\operatorname{tr} \ln[\delta(1; 2) + G_0(1; 1')\delta(t_1' - t_2) M(1'; 2)]\}.$$

The Green's function $G_0 = L_0^{-1}$ here is determined by the equation

$$L_0(1; 1') G_0(1'; 2) = \delta(1-2), \qquad (9)$$

and it satisfies the causality principle, being retarded. The operator M is introduced by means of

$$\delta(t_1 - t_2) M(1, 2) = u(1, 2) + 2\mathcal{F}(1, 1', 2) f(1').$$

It follows that the operator M is t-local and does not contain time derivatives.

The application of the operation tr to a series which is an expansion of a logarithm generates closed cycles of the retarded functions G_0. All such closed cycles which contain more than one function G_0 obviously vanish. The remaining cycle $\mathrm{tr}[G_0(1,1')\delta(t'_1-t_2)M(1',2)]$ requires a separate analysis, since the function G_0 is discontinuous when the times are equal. A cycle of this sort can naturally be redefined as zero.[6,7] To demonstrate the point, we note that a term of the type $G_0(1',2)\mathcal{F}(2',3',1')$ formally contains $\delta(t'_2-t'_1)$ in the vertex $\mathcal{F}$, but actually the physical process of the coalescence of field quanta (the coagulation of particles) occurs over a finite time, and the termination of the interaction event cannot affect its beginning. It is thus necessary, strictly speaking, to introduce infinitely short retardation times in the determination of the vertices describing the interaction. The cycle under consideration then makes a zero contribution.

The determinant in (8) is a constant which (as is easily shown) is insignificant in a determination of correlation functions. It is thus possible to rewrite (8) as

$$W[\eta,\hat\eta]=\int D\hat f\,Df\exp[i\hat f L_0 f+i\hat f\mathcal{F}ff-i\hat f\hat\eta+i\eta f]\langle\exp(i\hat f u f)\rangle.$$

We further assume that the ensemble of realizations of the stochastic velocity field $\delta\mathbf{u}$ is Gaussian with a zero mean and a uniform pair correlation function

$$B_{\alpha\beta}(1,2)=\langle\delta u_\alpha(1)\delta u_\beta(2)\rangle=B_{\alpha\beta}(1-2). \qquad (10)$$

One can then show that

$$\langle\exp(i\hat f u f)\rangle=\exp[-{}^1/_2\hat f(1')\hat f(2')K(1',2',3',4')f(3')f(4')], \qquad (11)$$

where

$$K(1,2,3,4)=B_{\alpha'\beta'}(1,2)\delta(1-3)\delta(2-4)\nabla_3{}^{\alpha'}\nabla_4{}^{\beta'}.$$

Using this relation, we find the representation of the generating functional which we need:

$$W[\eta,\hat\eta]=\int D\hat f\,Df\exp\Big[i\hat f L_0 f+i\hat f\mathcal{F}ff-\frac{1}{2}\hat f\hat f K ff-i\hat f\hat\eta+i\eta f\Big]. \qquad (12)$$

It has thus been shown that stochastic problem (7) with a Gaussian random coefficient of the field function is equivalent to a quantum theory of fields $\hat f, f$ with an action

$$\hat f L_0 f+\hat f\mathcal{F}ff+{}^1/_2 i\hat f\hat f K ff.$$

3. PERTURBATION THEORY AND DIAGRAM TECHNIQUE

To calculate W by perturbation theory, one ordinarily uses the representation[8]

$$W[\eta,\hat\eta]=\exp\Big[i\frac{\delta}{(-i)\delta\hat\eta}\mathcal{F}\frac{\delta}{i\delta\eta}\frac{\delta}{i\delta\eta}-\frac{1}{2}\frac{\delta}{(-i)\delta\hat\eta}\frac{\delta}{(-i)\delta\hat\eta}K\times\frac{\delta}{i\delta\eta}\frac{\delta}{i\delta\eta}\Big]\int D\hat f\,Df\exp[i\hat f L_0 f-i\hat f\hat\eta+i\eta f]. \qquad (13)$$

In the absence of an interaction ($\mathcal{F}=0$, $K=0$) the generating functional is quadratic in the fields and can be evaluated by making use of the property that the path integral is invariant under changes of integration variable of the functional-shift type, i.e., $\hat f\to\hat f+\hat c$, $f\to f+c$, followed by a choice of functions $\hat c$ and c on the basis of the condition that the terms which are linear in $\hat f$ and f in the argument of the exponential function vanish:

$$W_0[\eta,\hat\eta]=\int D\hat f\,Df\exp[i\hat f L_0 f-i\hat f\hat\eta+i\eta f]=\exp[i\eta G_0\hat\eta+i\eta f_0]\times\int D\hat f\,Df\exp[i\hat f L_0 f]=\exp[i\eta G_0\hat\eta+i\eta f_0]W_0[0,0]. \qquad (14)$$

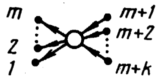
The function f_0 here is the solution of the homogeneous equation

$$L_0(1,1')f_0(1')=0.$$

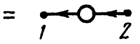
A perturbation theory is constructed by expanding the first exponential function in (13) in a power series in $\mathcal{F}$ and K. It is convenient to associate graphical symbols—Feynman diagrams—with the cumbersome analytic expressions which arise in the perturbation theory. Feynman diagrams make it possible to interpret the various factors and terms as processes by which particles propagate and undergo conversions.

We introduce the following notation:

$$\Big(\frac{\delta}{i\delta\eta(1)}\cdots\frac{\delta}{i\delta\eta(m)}\frac{\delta}{(-i)\delta\hat\eta(m+1)}\cdots\frac{\delta}{(-i)\delta\hat\eta(m+k)}\ln W[\eta,\hat\eta]\Big)_{\eta=0}=\langle f(1)\dots f(m)\hat f(m+1)\dots\hat f(m+k)\rangle$$

$=$ 

where $m=0,1,2,\dots$ and $k=0,1,2,\dots$ except $m=k=1$. We write the diagram for the values $m=k=1$ separately:

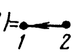
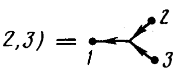
$$\Big(\frac{\delta}{i\delta\eta(1)}\frac{\delta}{\delta\hat\eta(2)}\ln W[\eta,\hat\eta]\Big)_{\eta=0}=\Big(\frac{\delta}{\delta\hat\eta(2)}\langle f(1)\rangle_\eta\Big)_{\eta=0}=(-i)\langle f(1)\hat f(2)\rangle=G(1,2)=$$ 

The case is singled out because this quantity describes the mean linear response of the field f to a unit external agent $\hat\eta$; in the case $\mathcal{F}=0$, $K=0$ it is the Green's function of linear equation (9). To demonstrate the point we use (14) and write

$$(-i)\langle f(1)\hat f(2)\rangle_0=\Big(\frac{\delta}{i\delta\eta(1)}\frac{\delta}{\delta\hat\eta(2)}\ln W_0[\eta,\hat\eta]\Big)_{\eta=0}=G_0(1,2).$$

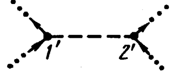
Here $\langle\dots\rangle_0$ means that generating functional W_0 is used in calculating the corresponding mean.

We also assume

$G_0(1,2)=$  , $\mathcal{F}(1,2,3)=$  ,

$$\langle\dots\hat f(1')\rangle\langle\dots\hat f(2')\rangle K(1',2',3',4')\langle f(3')\dots\rangle\langle f(4')\dots\rangle$$

$$\langle\dots\hat f(1')\rangle\langle\dots\hat f(2')\rangle B_{\alpha'\beta'}(1',2')\nabla_{1'}{}^{\alpha'}$$

$\times\langle f(1')\dots\rangle\nabla_{2'}{}^{\beta'}\langle f(2')\dots\rangle=$ 

The dots here can be replaced by any combination of diagrams with a single incoming or outgoing solid line, respectively. An operator vertex with a dashed line implies taking the gradient of the solid line coming into it.

As usual, an integration is carried out over continuous arguments, and a summation is carried out over vector indices, at interior points of diagrams.

An important property of this diagram technique is that all the diagrams which have only incoming lines vanish (correspondingly, all the mean values containing only the auxiliary fields $\hat{f}$ vanish):

$$\langle \hat{f}(1)\rangle_0 = \left(\frac{\delta}{(-i)\delta\hat{\eta}(1)}[i\eta G_0\hat{\eta} + i\eta f_0]\right)_{\eta=0}$$

$$= (-\eta(1')G_0(1',1))_{\eta=0} = 0.$$

Consequently, any diagram which has only incoming lines necessarily contains a closed cycle of retarded propagator lines G_0 and vertices $\mathscr{F}$. It is therefore zero. The properties of diagrams of this type are discussed in Ref. 6; see Ref. 9 regarding the vanishing of the mean values which contain only auxiliary fields.

4. SCHWINGER EQUATIONS OF BROWNIAN COAGULATION IN A STOCHASTIC MEDIUM

It is preferable to construct a perturbation theory with the help of a generating functional rather than to repeatedly iterate the coagulation equation and then take an average, since it is necessary to calculate only the corresponding variational derivatives. In addition, the field formalism makes it possible to construct exact equations which relate various correlation functions without resorting to a perturbation theory. The derivation of these equations in a perturbation theory requires summing infinite diagram series—a laborious problem for a nontrival theory. We will derive some equations which relate the various correlation functions of this problem by making use of the invariance of the path integral under a change in the integration variable of the functional-shift type: $\hat{f}\to\hat{f}+\hat{\varphi}$, where $\hat{\varphi}$ is an arbitrary fixed function:

$$\frac{\delta W[\eta,\hat{\eta}]}{\delta\hat{\varphi}(1)} = \int Df\,D\hat{f}\,\frac{\delta}{\delta\hat{f}(1)}\exp[i\hat{f}L_0 f + i\hat{f}\mathscr{F}ff - \frac{1}{2}\hat{f}\hat{f}Kff - i\hat{f}\hat{\eta} + i\eta f]$$

$$= i\Big\{L_0(1,1')\frac{\delta}{i\delta\eta(1')} + \mathscr{F}(1,1',2')\frac{\delta}{i\delta\eta(1')}\frac{\delta}{i\delta\eta(2')} + i\frac{\delta}{(-i)\delta\hat{\eta}(1')}K(1,1',2',3')\frac{\delta}{i\delta\eta(2')}\frac{\delta}{i\delta\eta(3')} - \hat{\eta}(1)\Big\}W[\eta,\hat{\eta}] = 0. \quad (15)$$

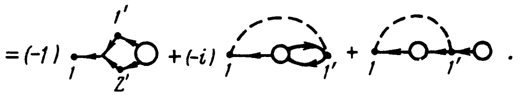
We express the variational derivatives of the generating functional which appear in (15) in terms of the derivatives of the logarithm of this functional, and we set $\eta = 0$. As a result we find the following for our problem, in the absence of deterministic external sources ($\hat{\eta} = 0$), and under the assumption $\langle\hat{f}\rangle = 0$:

$$L_0(1,1')\langle f(1')\rangle + \mathscr{F}(1,1',2')\langle f(1')\rangle\langle f(2')\rangle = (-1)\mathscr{F}(1,1',2')\langle f(1')f(2')\rangle + (-i)K(1,1',2',3')[\langle\hat{f}(1')f(2')\rangle\langle f(3')\rangle + \langle\hat{f}(1')f(3')\rangle\langle f(2')\rangle + \langle\hat{f}(1')f(2')f(3')\rangle]. \quad (16)$$

This equation is an exact variational-derivative equation for the mean field of the distribution function of the aerosol particles. By analogy with quantum field theory, we call it a "Schwinger equation." The left side of (16) corresponds to coagulation equation (3) in the absence of fluctuations ($\delta\mathbf{u} = 0$). Terms describing the effect of fluctuations appear on the right side of (16).

In diagram notation, the Schwinger equation for the mean field is

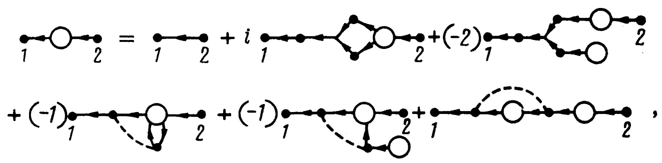
$$L_0(1,1')\langle f(1')\rangle + \mathscr{F}(1,1',2')\langle f(1')\rangle\langle f(2')\rangle$$



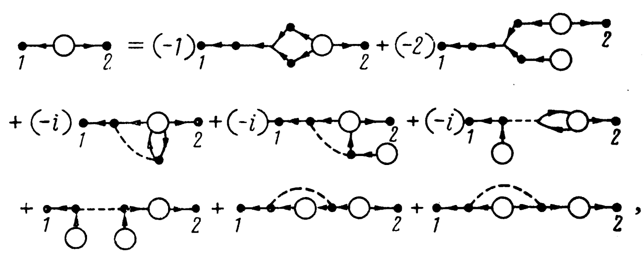
(17)

In (17) and in the expressions to follow, we omit those diagrams which are zero in accordance with properties of our diagram technique. Schwinger equations determining the terms on the right side of (17) are found by analogy with the derivation of (16). In this manner we find a system of exact equations which relate different correlation functions.

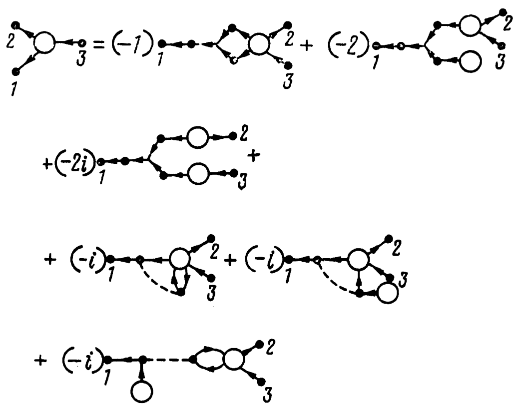
We can write Schwinger equations for the vertices on the right side of Eq. (17):

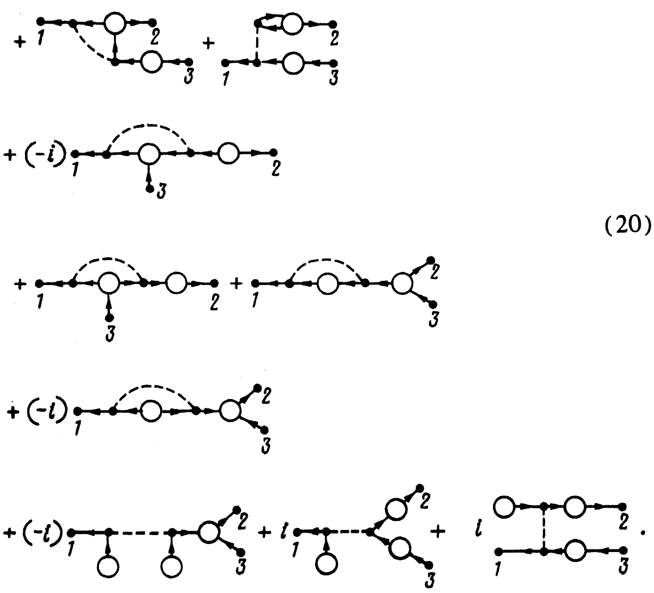


(18)



(19)





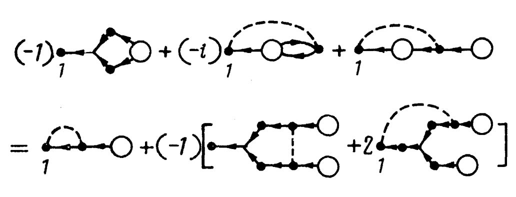
5. EFFECTIVE EQUATION FOR THE MEAN FIELD OF THE DISTRIBUTION FUNCTION

Let us use these results to evaluate the diagrams which describe the effect of fluctuations in Eq. (17) for the mean field under the assumption that the turbulent fluctuations are small, i.e.,

$$v_0^2\tau_0^2/3R_0^2 \ll 1,$$

where $v_0 = (\langle \delta \mathbf{u}^2 \rangle)^{1/2}$ determines the amplitude of the velocity fluctuations, and R_0 and τ_0 are a correlation length and a correlation time of the velocity field of the medium.

To carry out the calculations, we use a perturbation theory in the correlation function B, retaining terms $O(B)$. In addition, allowing for the condition for the applicability of the kinetic equation for Brownian coagulation, $R^3\overline{N} \ll 1$ ($\overline{N}$ is the mean density of the aerosol particles), we naturally include in the description only the terms of first order in the coagulation kernel S and thus of first order in the vertex $\mathcal{F}$. Using Schwinger equations (18)–(20) and the corresponding equations for the other vertices in them, we find the following for the diagrams on the right side of Eq. (17):



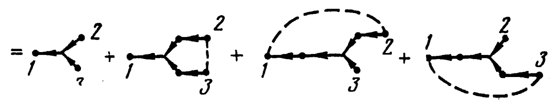
$$+O(B^2)+O(B\mathcal{F}^2). \qquad (21)$$

The expressions in square brackets correspond to corrections $O(B)$ to the effective coagulation vertex Γ, which is defined by

$$\langle f(1) f(2) f(3) \rangle = 2G(1,1')\Gamma(1',2',3')G(2',2)G(3',3). \qquad (22)$$

Specifically, we have

$$\Gamma(1,2,3)$$



$$+O(B^2)+O(B\mathcal{F}^2).$$

The vertex Γ is strongly coupled (one-particle-irreducible) and describes a coalescence (coagulation) of particles without consideration of effects which stem from the propagation of individual particles before and after the coalescence. This approach is in complete accordance with definition (22). It is important to note that within $O(B\mathcal{F}^2)$ the coagulation vertex does not depend on the mean field of the distribution function of the aerosol particles, being determined exclusively by the seed coagulation vertex $\mathcal{F}$ and by the correlation function B of the medium. A dependence of the effective coagulation vertex on the mean field of the distribution function arises in the following orders of perturbation theory.

The diagrams on the right side of (21) correspond to nonlocal operators which are operating on the mean field. In addition to the nonlocality in terms of volumes V (this nonlocality is also a property of the seed coagulation vertex), there is a nonlocality in terms of the spatial and temporal variables in these diagrams. For example,

$$=\mathcal{F}(1,2',3')G_0(2',4')G_0(3',5') \times K(4',5',6',7')\langle f(6')\rangle\langle f(7')\rangle$$
$$=P(V_1,V_2',V_3')\tilde{G}_0(t_1-t_2',\mathbf{r}_1-\mathbf{r}_2',V_2')\tilde{G}_0(t_1-t_3',\mathbf{r}_1-\mathbf{r}_3',V_3')$$
$$\times B_{\alpha'\beta'}(t_2'-t_3',\mathbf{r}_2'-\mathbf{r}_3')\nabla^{\alpha'}\langle f(t_2',\mathbf{r}_2',V_2')\rangle\nabla^{\beta'}\langle f(t_3',\mathbf{r}_3',V_3')\rangle.$$

Here

$$\tilde{G}_0(t_1-t_2,\mathbf{r}_1-\mathbf{r}_2,V_1)=\theta(t_1-t_2)[4\pi D(V_1)(t_1-t_2)]^{-3/2}\cdot \exp[-(\mathbf{r}_1-\mathbf{r}_2)^2/4D(V_1)(t_1-t_2)]$$

is the retarded Green's function of the diffusion equation for an unbounded medium. The Green's functions $\tilde{G}_0$ and G_0 are related by

$$G_0(1,2)=G_0(t_1,\mathbf{r}_1,V_1,t_2,\mathbf{r}_2,V_2) =\delta(V_1-V_2)\tilde{G}_0(t_1-t_2,\mathbf{r}_1-\mathbf{r}_2,V_1).$$

Equation (21) thus gives us an exact expression for the diagrams $O(B)+O(B\mathcal{F})$, whose specific form can be calculated either analytically or numerically, if the pair correlation function of the velocity field of the medium is known.

It is convenient to calculate the diagrams in the momentum representation, in which the functions $\tilde{G}_0$ correspond to the following Fourier amplitude:

$$\tilde{G}_0(p^0,\mathbf{p},V)=[ip^0+D(V)\mathbf{p}^2]^{-1}.$$

For specific calculations we specify the pair correlation function[10,11]

$$B_{\alpha\beta}(1-2) =B_0\left[\delta_{\alpha\beta}\left(1-\frac{\mathbf{R}^2}{R_0^2}\right)+\frac{R_\alpha R_\beta}{R_0^2}\right]\exp\left[-\frac{|t_1-t_2|}{\tau_0}-\frac{\mathbf{R}^2}{R_0^2}\right], \qquad (23)$$

where $\mathbf{R} = \mathbf{r}_1 - \mathbf{r}_2$ and $B_0 = (1/3)v_0^2$. In other words, B_0 determines the strength of the velocity fluctuations.

It is easy to see that the Fourier amplitude of this correlation function is

$$B_{\alpha\beta}(p^0, \mathbf{p})$$
$$= {}^1/_2\pi^{3/2}B_0R_0^5\tau_0^{-1}[(p^0)^2+\tau_0^{-2}]^{-1}(\delta_{\alpha\beta}\mathbf{p}^2-p_\alpha p_\beta)\exp[-(\mathbf{p}R_0)^2/4].$$

It can be shown that the following conditions hold for many real aerodisperse systems:

1. The correlation radius and time of the turbulent velocity fluctuations of the medium are much smaller than the characteristic length and the characteristic time, respectively, of the variations of the mean field,

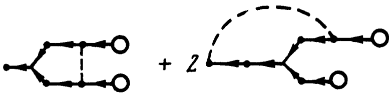
$$|\mathbf{p}|\ll R_0^{-1}, \quad |p^0|\ll\tau_0^{-1}. \tag{24}$$

2. The characteristic spatial-correlation length is much larger than the distance over which the particle densities equalize as a result of Brownian diffusion over the correlation time τ_0; i.e.,

$$R_0\gg(D\tau_0)^{1/2}. \tag{25}$$

These conditions make it possible to simplify the calculations substantially and to derive approximate analytic expressions for the diagrams of interest:


$\approx B_0\tau_0\nabla^2\langle f(1)\rangle,$



$$\approx P(V_1, V_2', V_3')\delta(t_2'-t_3')\delta(\mathbf{r}_2'-\mathbf{r}_3')$$

$$\times\left\{\left[\Delta_1(t_1-t_2'|\tilde{t})\Delta_2(\mathbf{r}_1-\mathbf{r}_2'|\rho)\frac{4}{3}B_0\tau_0\tilde{t}\right.\right.$$
$$\left.+\delta(t_1-t_2')\delta(\mathbf{r}_1-\mathbf{r}_2')B_0\tau_0^2\right]\nabla_{2'}\langle f(2')\rangle\nabla_{3'}\langle f(3')\rangle$$
$$\left.+\delta(t_1-t_2')\delta(\mathbf{r}_1-\mathbf{r}_2')2B_0\tau_0^2\langle f(2')\rangle\nabla_{3'}^2\langle f(3')\rangle\right\}. \tag{26}$$

Here

$$\Delta_1(\tau|\tilde{t})={}^3/_2\theta(\tau)\tilde{t}^{-1}(1+\tau/\tilde{t})^{-5/2},$$
$$\tilde{t}={}^1/_4R_0^2[D(V_2')+D(V_3')]^{-1},$$
$$\Delta_2(\mathbf{R}|\rho)=\frac{\theta(t_1-t_2')}{\pi^{3/2}(2\rho)^3}\exp\left[-\frac{\mathbf{R}^2}{(2\rho)^2}\right],$$
$$\rho^2=\frac{D(V_2')D(V_3')(t_1-t_2')^2}{({}^1/_2R_0)^2+[D(V_2')+D(V_3')](t_1-t_2')}.$$

The second expression in (26) obviously localizes along the spatial and temporal variables if it is assumed that the characteristic spatial-correlation length is much shorter than the distance over which the particle densities are equalized by Brownian diffusion over the characteristic time of the variation of the mean field:

$$R_0\ll(D/|p^0|)^{1/2}. \tag{27}$$

This condition also corresponds to reality in several cases, since the time scale of Brownian coagulation is $(\overline{N}DR)^{-1}$.

Using conditions (27) and (25), we find the following expression for the average collision integral:

$$(-1)\mathscr{F}(1, 1', 2')[1+{}^1/_6N_1(1', 2')\nabla_{1'(\alpha')}\nabla_{2'}^{\alpha'}+N_2\nabla_{2'}^2]$$
$$\times\langle f(1')\rangle\langle f(2')\rangle,$$

where

$$N_1(1',2')=2B_0\tau_0[D(V_1')+D(V_2')]^{-1}R_0^2,$$
$$N_2=2B_0\tau_0^2.$$

The effective equation for the mean field of the distribution function of the aerosol particles thus takes the following form when the turbulence velocity fluctuations are weak:

$$\frac{\partial}{\partial t_1}\langle f(1)\rangle-D_{\text{eff}}\nabla^2\langle f(1)\rangle+\mathscr{F}(1, 1', 2')$$
$$\times\left[1+\frac{1}{3}N_1(1', 2')\nabla_{1'(\alpha')}\nabla_{2'}^{\alpha'}+N_2\nabla_{2'}^2\right]\langle f(1')\rangle\langle f(2')\rangle=0. \tag{28}$$

The expression for the effective diffusion coefficient,

$$D_{\text{eff}}=D(V)+D_T,$$

where $D_T = B_0\tau_0$ is the turbulent diffusion coefficient, is the same as the result found in a corresponding calculation in first order in correlation function (23) for a diffusion equation without coagulation (Ref. 11, for example).

A further simplification can be achieved by assuming that the mean distribution function is a sufficiently smooth function of the spatial coordinates. Using condition (25), we can then ignore the third term in the effective collision integral. As a result we find

$$\frac{\partial}{\partial t}\langle f\rangle-D_{\text{eff}}\nabla^2\langle f\rangle$$
$$=\{\langle f\rangle, \langle f\rangle\}-\frac{1}{12}\int_0^\infty dV_1\int_0^\infty dV_2[\delta(V-V_1)+\delta(V-V_2)$$
$$-\delta(V-V_1-V_2)]S_T(V_1, V_2)R_0^2\nabla\langle f(t, \mathbf{r}, V_1)\rangle\nabla\langle f(t, \mathbf{r}, V_2)\rangle. \tag{29}$$

Here

$$S_T(V_1, V_2)=4\pi(R_1+R_2)\cdot 2D_T$$

is the renormalized kernel of Brownian coagulation in a turbulent medium.

Using $D(V)\ll D_T$, and integrating over volumes in (29), we easily find a corresponding equation for the mean density of the aerosol particles:

$$\frac{\partial}{\partial t}\langle N(t, \mathbf{r})\rangle-D_T\nabla^2\langle N(t, \mathbf{r})\rangle$$
$$=-\frac{1}{2}\int_0^\infty dV_1\int_0^\infty dV_2[S(V_1, V_2)\langle f(t, \mathbf{r}, V_1)\rangle$$
$$\times\langle f(t, \mathbf{r}, V_2)\rangle+\frac{1}{3}S_T(V_1, V_2)R_0^2\nabla\langle f(t, \mathbf{r}, V_1)\rangle\nabla\langle f(t, \mathbf{r}, V_2)\rangle]. \tag{30}$$

6. CONCLUSION

It can be seen from the equation found for the mean distribution function of the aerosol particles that in the approximation of the model of a passive impurity the case of a uniform coagulation ($\nabla\langle f\rangle = 0$) leads to simply a renormalization of the diffusion coefficient, having no effect on the collision integral.

For passive impurities, all the coagulation effects of the turbulence are thus due exclusively to the nonuniformity of the mean distribution function. In addition, the nonnegativity of the turbulence correction in the collision integral confirms the suggestion by Voloshchuk and Sedunov[1] that a coagulation of aerosol particles is accelerated in an inhomogeneous stochastic medium.

It is interesting to examine the structure of this correction. For this purpose, we write it in the form

$$^1/_6 R_0\{4\pi[D(V_1)+D(V_2)]\}^{-1}Q_{R_0}(t,\mathbf{r},V_1)Q_R(t,\mathbf{r},V_1,V_2),$$

where

$$Q_{R_0}(t,\mathbf{r},V_1)=4\pi R_0\cdot 2D_T\nabla\langle f(t,\mathbf{r},V_1)\rangle,$$

$$Q_R(t,\mathbf{r},V_1,V_2)=4\pi(R_1+R_2)[D(V_1)+D(V_2)]\nabla\langle f(t,\mathbf{r},V_2)\rangle.$$

It can be seen that the turbulence correction is proportional to the product of the mean quasisteady integral fluxes of aerosol particles to a correlation sphere (Q_{R0}) and to an absorbing sphere (Q_R).

For Brownian coagulation, the condition

$$S/S_T \ll 1,$$

usually holds, so the turbulence correction in the collision integral may play a governing role if the gradients of the field of the distribution function are sufficiently large. Consequently, the conclusion, reached in Ref. 4, that turbulence has a negligible effect on the coagulation term under conditions (24) is not generally correct, in our opinion.

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