The proposed method can be extended in a natural way to modeling of the optical characteristics of fibrous composites as well as to modeling of the effect that structural macrodefects having their own peculiar distributions with respect to size and physical properties have on the physical characteristics.

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# THE THEORY OF THE RHEOLOGICAL PROPERTIES OF DISPERSE SYSTEMS


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The effective rheological characteristics of stacking identical viscoelastic spheres in a matrix of another viscoelastic material are estimated by methods of ensemble averaging theory.


The intensive development of technological processes utilizing stacks of fine particles as working bodies requires the development of physicomathematical models that permit relating the macrorheological properties of such systems to the singularities of their configuration can be given within the framework of the continual approximation, when the disperse mixture is considered as a homogeneous continuum whose behavior is described by the methods of the mechanics of continuous media. However, even in this case the problem that has still not been solved by far arises of calculating the effective charcteristics of a heterogeneous material as a function of the properties of its phases or components and the singularities of their arrangement.

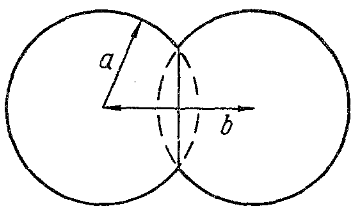


Fig. 1. Model of the contacts between spheres. Dashes are the geometric surfaces of continuation of the spheres.


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An extensive literature, whose surveys can be found in [1-3], is devoted to methods of qualitative estimates of the effective rheological properties of composites with isolated inclusions. The system characteristics, whose particles form a connected skeleton are investigated much less. Meanwhile precisely such systems play a very large part in applications [4-8]. Certain formulas for the determination of effective elastic moduli and viscosity of porous materials, that are extensively utilized in powder metallurgy, are obtained in [4, 8] and discussed briefly in [7]. However, these papers are based on heuristic models and although their results are completely suitable for simple engineering computations, the further development of the theory and practice of disperse systems requires the development of more sequential methods of determining their rheological properties.

Stacks of identical viscoelastic particles in a matrix of a different viscoelastic material are investigated below. Such a consideration complicates the calculation somewhat as compared with the stacks ordinarily investigated in air but then possesses great generality. For simplification we consider the particles mutually penetrating spheres without deformation (Fig. 1). The investigation is performed on the basis of a rigorous theory of ensemble averaging developed in [9] and applied in [10] to describe the viscoelastic properties of a composite with isolated inclusions. The general ideas of [9] were utilized earlier in [11] to analyze the thermophysical properties of finely dispersed stacks.

## MACROSCOPIC EQUATIONS

The behavior of viscoelastic matrices and particles can be described by using their bulk $k_j$ and shear $\mu_j$ elastic moduli as well as the bulk $\xi_j$ and shear $\eta_j$ viscosities ($j = 0, 1$) [12]. The problem is to determine the effective coefficients k, $\mu$, $\eta$, and $\xi$, referring to the material as a whole.

For the sequel it is convenient to apply the Fourier time transformation to the matrix and particle deformation equations. The relationships obtained agree formally with the deformation equaitons for elastic materials [1, 10], but in place of the elastic moduli $k_j$ and $\mu_j$ the equations

$$\beta_j = k_j + i\omega\xi_j,\ \gamma_j = \mu_j + i\omega\eta_j,\ i = \sqrt{-1} \tag{1}$$

are therein. Taking the average in conformity with the general theory [9, 10], of the Fourier representation of the particle and material deformation equaitons over the whole physically realizable particle positions in the mixture, we arrive at the macroscopic equations of deformation of a composite that relates the mean (measurable) values of the material displacement vector $\mathbf{u}$, the viscoelastic stress $\sigma$ and the bulk particle

$$-d\omega^2\mathbf{u} = \nabla\sigma,\ \sigma = \mathbf{I}\beta\,\mathrm{div}\,\mathbf{u} + 2\gamma\mathbf{e},\ d = d_0 + \rho(d_1 - d_0), \tag{2}$$

where the components of the pure shear tensor $\mathbf{e}$ are expressed in the usual way [12] in terms of the derivatives of $\mathbf{u}$ with respect to the coordinates.

The effective bulk $\beta$ and shear $\gamma$ moduli are determined from the relationships:

$$(\beta - \beta_0)\,\mathrm{div}\,\mathbf{u} = (\beta_1 - \beta_0)\frac{\rho}{v}\int\limits_{r\leqslant a}\mathrm{div}\,\mathbf{u}^* d\mathbf{r},$$
$$(\gamma - \gamma_0)\mathbf{e} = (\gamma_1 - \gamma_0)\frac{\rho}{v}\int\limits_{r\leqslant a}\mathbf{e}^* d\mathbf{r}, \tag{3}$$

obtained in [10] and equivalent to those presented in [1-3]. Here integration is over the volume of a certain arbitrarily separated test particle, whose center is at a point to which div $\mathbf{u}$ and $\mathbf{e}$ are referred. Within the framework of [9, 10] the deformations div $\mathbf{u}^*$ and $\mathbf{e}^*$ within the test sphere are determined from an auxiliary problem formed by taking the average of the particle and matrix deformation equiatons over all possible particle positions under the condition that the position of the test sphere is fixed.

## TEST PARTICLE PROBLEM

The apparatus of conditional averaging is developed in [9] and used [10] in application to mixture with isolated particles. Omitting the details, let us mention that in systems with particles making contact the problem of a test inclusion agrees formally with that obtained in [10] and has the form

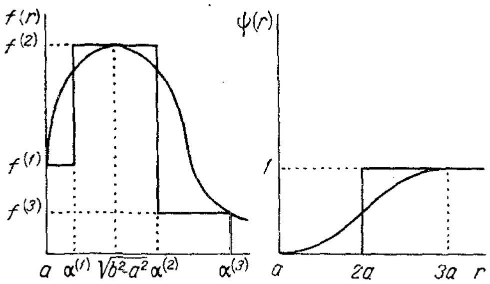


Fig. 2

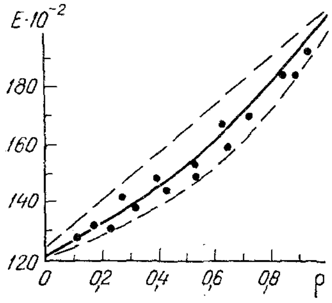


Fig. 3

Fig. 2. Functions f(r) and ψ(r) that are in (A.1) (smooth lines) and their step approximations utilized in (8) and in later computations. The values of $\alpha^{(i)}$ and $f^{(i)}$ are presented in (A.2).

Fig. 3. Comparison of the results of computations of the Young's modulus E of a composite material consisting of a skeleton of iron particles placed in a copper matrix carried out according to the proposed theory (solid line) with experiments [14] (points and computations [14] by different methods (dashed lines). E in kg/mm².

$$-d'\omega^2\hat{\mathbf{u}} = \nabla\hat{\sigma},\ r \geqslant a;\ -d_1\omega^2\mathbf{u}^* = \nabla\sigma^*,\ r \leqslant a;$$
$$\mathbf{u}^* = \mathbf{u} + \hat{\mathbf{u}},\ \mathbf{n}\sigma^* = \mathbf{n}\sigma + \mathbf{n}\hat{\sigma},\ r = a;$$
$$\hat{\mathbf{u}} \to 0,\ r \to \infty;$$
$$\hat{\sigma} = \mathbf{I}\beta' \operatorname{div}\hat{\mathbf{u}} + 2\gamma'\hat{\mathbf{e}},\ d' = d_0 + (d - d_0)\frac{\rho'(r)}{\rho}, \tag{4}$$
$$\beta' = \beta_0 + (\beta - \beta_0)\frac{\rho'(r)}{\rho},\ \gamma' = \gamma_0 + (\gamma - \gamma_0)\frac{\rho'(r)}{\rho}.$$

Here $\mathbf{u}$, $\mathbf{e}$, and σ are the mean values occurring in (2) and (3) that are referred to the site of the test sphere position. They are considered as given in (4). The quantities with carets above them are perturbations introduced by the test particle into the appropriate mean fields, ρ'(r) is the conditional bulk particle concentration near the fixed test particle (the probability of finding r within one of the particles surrounding the test sphere). It does not equal the mean concentration ρ of the disperse phase in the material since the particles cannot penetrate freely within each other and a fixed particle "pushes apart" the neighbors. The properties ρ'(r) for mixtures with isolated spheres are discussed in [13]. In particular, as $r \to \infty$ $\rho' \to \rho$, which corresponds to weak correlation between the positions of remote particles. Solving (4) by some method of a specific function ρ'(r), we find $\mathbf{u}^*$, dependent on β and γ as parameters. Substituting this value into the integrals of (3) we obtain a system of equations to determine β and γ, which permits finding the values of these characteristics of the medium by a self-consistent method.

## CONDITIONAL BULK CONCENTRATION OF THE DISPERSE PHASE

The explicit form of ρ'(r) is dictated by the kind of particle stacking in the material. In conformity with the general theory [9, 13], this quantity is defined thus

$$\rho'(r) = \int_{|\mathbf{r}-\mathbf{r}'| \leqslant a} \varphi(r')\, d\mathbf{r}', \tag{5}$$

where φ is a binary particle distribution function that depends on the specific method of arranging the particles in the mixture and should be determined from the solution of an independent statistical problem. Within the framework of the theory being utilized φ is considered known.

As in [11], we consider the stacking structure often encoutered when the mean distance b ($b \leq 2a$) between the centers of the spheres making contact and the coordination number ζ equal to the mean number of contacts per unit surface of the test sphere are known. The simplest form of φ, corresponding to this stacking is

$$\varphi(r) = \zeta\delta(r-b) + \begin{cases} 0, & r \leqslant 2a, \\ \dfrac{\rho}{v}, & r > 2a, \end{cases} \tag{6}$$

where $\delta(x)$ is the delta function. The first component here takes into account that the centers of the spheres making contact with the test sphere are at a distance b from its center, the second is the circumstance that the remaining particles do not penetrate the test particle and consequently their centers cannot be at distances less than 2a from the center of the fixed test sphere. Moreover, the influence of the test sphere on the positions of particles removed at a distance greater than 2a from it.

Substituting (6) into (5) we arrive at a complex dependence of $\rho'$ on r whose explicit form is given in the appendix and is shown by smooth lines in Fig. 2. As is seen, $\rho'(a) \neq 0$, which reflects the finiteness of the contact spots (domains of model sphere intersection (see Fig. 1)). Let us note that within the framework of the problem (4), the contact spots "are spread out" over its surface because of averaging of the deformation equations relative to the positions of particles surrounding the test sphere, and the loaded state of a fictitious continuum containing the test sphere is described by the unit moduli $\mu'$ and $\beta'$ on its whole surface.

## EFFECTIVE VISCOELASTIC MODULI

We shall consider below that the following strong inequality is satisfied

$$\omega^2 \ll \{|\beta_j|, |\gamma_j|\}/\{d_j\}\, a^2, \; j = 0, 1. \tag{7}$$

Physically (7) means that inertial effects are weak in scales on the order of a, on whose basis we shall later neglect the left sides in the first two equations in (4). Estimates show that (7) is satisfied for real systems up to high-frequency sound waves. Meanwhile, the validity of (7) means that the characteristic scale of the variation of $\mathbf{u}$, $\operatorname{div}\mathbf{u}$ and e is much greater than a and near the test sphere $\operatorname{div}\mathbf{u}$, $e=\text{const}$ can be assumed. Let us note that this latter condition is the usual requirement for a continual approximation.

When using $\rho'(r)$ obtained from (5) and (6), the problem (4) allows only numerical solution. For approximate analytic computations it is convenient to approximate $\rho'(r)$ by step functions such as is shown in Fig. 2, say. Exactly the same approximation was used succcessfully in [11] for computations of the heat conduction of the same stack as here.

In this approximation (4) agrees formally with the problem on stationary deformation of a sphere in an infinite medium whose properties agree with the effective properties of the material but separated from this sphere by concentric layers of thicknesses $\alpha_1 - a$, $\alpha_2 - \alpha_1$, and $\alpha_3 - \alpha_2$ (see Fig. 2), where the viscoelastic moduli of these layers are defined, in conformity with the last equality in (4), thus:

$$\beta_k = \beta_0 + (\beta - \beta_0)\frac{\rho_k'}{\rho}, \; \gamma_k = \gamma_0 + (\gamma - \gamma_0)\frac{\rho_k'}{\rho},$$
$$k = 1,\; a < r < \alpha_1; \; k = 2,\; \alpha_1 < r < \alpha_2; \; k = 3,\; \alpha_2 < r < \alpha_3, \tag{8}$$

where the meaning of $\rho_k$ is clarified in Fig. 2 and their explicit form is given in the Appendix.

To determine the effective bulk modulus $\beta$ it is convenient to consider that the medium as a whole experiences multilateral compression even near the test sphere:

$$\mathbf{u} = \frac{1}{3}\varepsilon\mathbf{r}, \; \varepsilon = \operatorname{div}\mathbf{u} = \text{const}. \tag{9}$$

The solutions of (4) in the approximation (7) in each of the partition steps (8) agree formally with those presented in [1] with the elastic characteristics of the matrix replaced by the effective viscoelastic moduli $\beta_k$, $\gamma_k$. In particular, near the test sphere ($r \leq a$)

$$\mathbf{u}^* = \frac{1}{3}A\varepsilon\mathbf{r}, \; A = \text{const}. \tag{10}$$

In order to evaluate the shear modulus $\gamma$, we assume that the medium as a whole experiences pure shear and near the test particle:

$$u_x = ex; \; u_y = -ey; \; u_z = 0; \; e = \text{const}, \tag{11}$$

TABLE 1. Dependences of the Effective Young's Modulus E of Composites on the Bulk Concentration of Spherical Inclusions

| Matrix | Particle | ρ | $E$, kg/mm² | | | |
|---|---|---|---|---|---|---|
| | | | 1 | 2 | 3 | 4 |
| Copper | Iron | 95 | 19500 | 19300 | | 19500 |
| | | 75 | 18340 | 18080 | | 18500 |
| | | 70 | 18130 | 17790 | | 18000 |
| Copper | Molybdenum | 80 | 23800 | 25490 | 24960 | 24000 |
| | | 75 | 21300 | 24400 | 22460 | 21600 |
| | | 68 | 19200 | 22460 | 18240 | 18600 |
| | | 66 | 17400 | 21280 | 13860 | 17500 |

Note. Values of the Young's modulus of two composite materials. Columns: 1) Experiment [14]; 2, 3) Computations [14]; 4) Proposed theory for ζ selected according to recommendations [15] for particles concentration in the free filling state $\rho^0 = 0.6$.

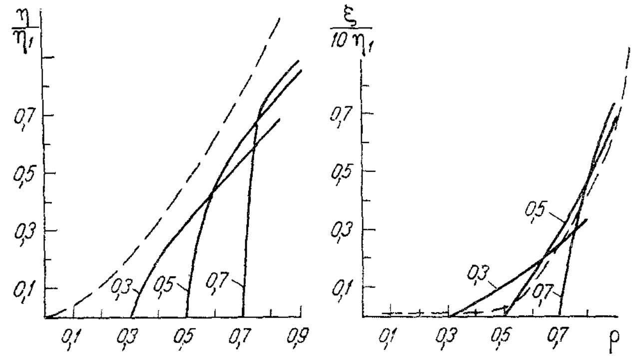


Fig. 4. Computations of the dependences of the effective bulk ξ and shear η viscosities of stacks of viscoelastic particles in air according to the formulas of [4] (dashed lines) and by the proposed theory (solid lines). The numbers at the curves are values of the concentration $\rho^0$ in the free filling.

where x, y, and z are Cartesian coordinates of the radius-vector with origin at the center of the test sphere. In this case the solutions of (4) again agree with those presented in [1] at each step with the replacement noted above taken into account. Within the particle:

$$u_r^* = [B_1 r - (3\beta_1 - 2\gamma_1) r^3 B_2]\, e \sin^2\theta \cos 2\varphi,$$
$$u_\theta^* = \left[B_1 r - \left(5\beta_1 + \frac{11}{3}\gamma_1\right) r^3 B_2\right] e \sin\theta\cos\theta\cos 2\varphi, \tag{12}$$
$$u_\varphi^* = -\left[B_1 r - \left(5\beta_1 + \frac{11}{3}\gamma_1\right) r^3 B_2\right] e \sin\theta \sin 2\varphi,\ B_1,\ B_2 = \mathrm{const},$$

where θ and φ are the polar and azimuthal angles in a spherical coordinate system with origin at the center of the test particle and polar axis directed along z.

The Cartesian coordinates of the tensor $e^*$, determined from (12) that are in the integrals of (3) are

$$e_{xx}^* = -e_{yy}^* = \left[B_1 - \frac{21}{5}\left(\beta_1 + \frac{4}{3}\gamma_1\right) a^2 B_2\right] e, \tag{13}$$

where the remaining $e_{ij}$ equal to zero.

Substituting (9) and (10) into (11), (13) and (3), we obtain after integration

$$\beta = \beta_0 + (\beta_1 - \beta_0) A\rho,$$
$$\gamma = \gamma_0 + (\gamma_1 - \gamma_0) \left[B_1 - \frac{21}{5}\left(\beta_1 + \frac{4}{3}\gamma_1\right) a^2 B_2\right] \rho. \tag{14}$$

The constants A, $B_1$ and $B_2$ can be determined from continuity conditions for the displacements and the normal components of the force density on the particle surface (r = a) and on the approximation step boudnaries (8), i.e., for $r = \alpha^{(1)}$, $\alpha^{(2)}$, and $\alpha^{(3)}$. An analogous "multilayer" problem is examined in detail in [1]. The system of linear algebraic equations coresponding to the mentioned boundary conditions and containing A, $B_1$ and $B_2$ together with other constants of integration (4) can be obtained exactly as was done in [1]. It is quite awkward and consequently not presented.

This system of boundary condidtions contains the effective moduli β and γ as parameters, consequently, it must be solved together with (8) and (14), which is done numerically most conveniently of all. A comparison of the results of computations by the method proposed here with the data of experiments [14] and computations by other methods utilized in [14] is presented in Fig. 3 and Table 1. It is seen that the proposed theory describes experiments better than traditional methods.

The question of the selection of the values of the coordination number ζ and the mean spacing between centers of the spheres making contact b occurs in the execution of practical computations. We used the recommendations in [15] here according to which ζ is the function $\rho^0$ determined in this paper, while $b \approx 2a(\rho^0/\rho)^{1/3}$, where $\rho^0$ is the bulk particle concentration in the free stacking state. As ρ increases the coordination number ζ changes continuously and a continuous line (see Fig. 3) is constructed as the envelope of curves corresponding to these values $\zeta = \zeta(\rho^0)$ [15] of $0 < \rho^0 < 1$.

It follows from (1), (8) and (14) that in the general case the effective moduli β and γ are complex functions of the frequency ω. For slow processes, when $\omega\{\eta_j, \xi_j\} \ll \{k_j, \mu_j\}$, it can be assumed with linear accuracy in ω:

$$\beta = k + i\omega\xi, \quad \gamma = \mu + i\omega\eta, \tag{15}$$

$$\{k, \mu\} = \{\beta, \gamma\}_{\omega=0}, \quad \{\xi, \eta\} = -i\left.\frac{\partial\{\beta, \gamma\}}{\partial\omega}\right|_{\omega=0}.$$

Substituting (15) into (2) and using the inverse Fourier transform in the time, that results in replacement of the factor iω by the operator ∂/∂t, we obtain

$$\sigma = \mathbf{I}\left(k + \xi\frac{\partial}{\partial t}\right) \operatorname{div}\mathbf{u} + 2\left(\mu + \eta\frac{\partial}{\partial t}\right)\mathbf{e}, \tag{16}$$

from which it follows that ξ and η have the meaning of effective bulk and shear viscosities of the material.

A comparison of calculations of ξ and η by the theory proposed here with computations by formulas from [4, 5, 7, 8] for stacks of incompressible viscoelastic particles in air ($\mu_0$, $k_0=0$, $k_1 \gg \mu_1 \gg \omega\eta_1 \sim \omega\xi_1$) is presented in Fig. 4. It is seen that the heuristic formulas of these papers can be utilized to estimate the shear viscosity of powder stacks. The results for ξ that follow from [4, 5, 7, 8] become unrealistically large as $\rho \to 1$ (more accurately, $\xi \to \infty$). We again utilized the recommendations in [15] in the computations to determine ζ and b and constructed an envelope of the curves obtained for different $\rho^0$.

Setting ζ = 0 in (6), which corresponds to no contacts between the particles, we arrive at relationships for β and γ that were obtained earlier in [10] for a composite with isolated inclusions that, as is shown in [10], describe known experiments well. Therefore, the proposed method permits description of materials with isolated particles and those making contact from a single viewpoint, where no constraints are imposed on the relationship between the properties of the particle and the matrix. In particular, pores can be inclusions, then it is necessary to set $\beta_1, \gamma_1 = 0$. This method affords the possibility of successively taking into account the influence of the kind of particle stacking given by the function $\varphi(r')$, on the effective properties of a composite.

The rigor and generality of the proposed theory are achieved at the cost of a loss of computation simplicity inherent in many empirical methods, consequently, it is not convenient for practical engineering usage. However, it can be basic for the analysis of more physically complex situations when intuitive constructions become too unreliable. Consequently, we note that the awkwardness of the rigorous results is a typical situation for the majority of statistical physics domains.

## APPENDIX

Using a distribution function of the form (6) in the integral of (5), we arrive at the following representation for the conditional bulk concentration whose graph is presented in Fig. 2 (smooth lines)

$$\rho'(r) = \zeta\pi f(r) + \rho\psi(r);\ \pi = 3.14\ldots$$
$$f(r) = \left(b - r - \frac{b^2 - a^2 - r^2}{2r}\right)^2 + 2\left[a^2 - \left(\frac{b^2 - a^2 - r^2}{2r}\right)^2\right],$$
$$r \in \Omega = \left[\frac{2b^2 - a^2 - a\sqrt{2b^2 + a^2}}{2b},\ \frac{2b^2 - a^2 + a\sqrt{2b^2 + a^2}}{2b}\right], \tag{A.1}$$
$$f(r) = 0,\ r \notin \Omega;$$
$$\psi(r) = \frac{27 - 56 + 30x^2 - x^4}{16x},\ 1 < x < 3,$$
$$\psi = 0,\ x < 1;\ \psi = 1,\ x > 3;\ x \equiv r/a.$$

The step approximation of f and $\psi$ used for the computations is shown by broken lines in Fig. 2. The parameters there have the following values:

$$\alpha^{(1)} = \frac{a + \sqrt{b^2 - a^2}}{2};\ \alpha^{(2)} = \frac{b + \sqrt{b^2 - a^2}}{2};\ \alpha^{(3)} = 2a;$$
$$f^{(j)} = f(r_j),\ j = 1,\ 2,\ 3; \tag{A.2}$$
$$r_1 = a;\ r_2 = \sqrt{b^2 - a^2};\ r_3 = 2a;$$
$$\rho_k = \zeta\pi f^{(k)} + \psi(r_k).$$

## NOTATION

a is the particle radius; b is the mean spacing between centers of particles making contact; $B_i$ are constants introduced in (12); d is the mean density; e is the pure shear tensor; I is the unit tensor; $k_j$ are bulk elastic moduli; n is the unit normal vector; R, R′ are radius vectors; $r = R - R'$; is time; u is the material displacement vector; $\alpha_i$ are quantities introduced in (A.2); $\beta_i$, $\gamma_i$ are effective bulk and shear elastic moduli; $\varepsilon = \operatorname{div} u$; $\zeta$ is the coordination number; $\eta$ is the shear viscosity; $\mu_i$ are shear elastic moduli; $\xi$ is bulk viscosity; $\rho$, $\rho'$ are mean and conditional bulk concentrations; $\rho^0$ is the particle bulk concentration in the free stacking state; $\sigma$ is the viscoelastic stress; $\varphi(r)$ is a binary distribution function; $\omega$ is the frequency. Subscripts: 0 and 1 refer to the dispersion and dispersed phases, respectively, and the asterisk superscript to quantities determined within the test particle.

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