PREDICTION OF NONLINEAR FLOW PARAMETERS FOR MULTICOMPONENT MIXTURES

V. A. Buryachenko and A. M. Lipanov UDC 541.24:539.4

A study of the flow mechanism for nonlinearly viscous microinhomogeneous media makes it possible to predict, according to component properties, the effective flow parameters for emulsified liquids, suspensions, lubricant additives, and other dispersed systems. A multicomponent viscous medium is considered consisting of a uniform matrix and a random mass of ellipsoidal inclusions. Computation of macroscopic rheological constants for the medium is accomplished by a variant of the effective field method suggested in [1, 2] for solving a broad class of elasticity problems in structural mechanics. The method is based on asymptotically accurate solution of binary interaction of inclusions which are in an effective field assuming uniformity of stresses within each inclusion. Solution of the problem was obtained assuming uniformity within each tensor component characterizing the nonlinear properties of the material.

1. General Relationships. We consider an unbounded inhomogeneous medium whose components are described by rheological equations local at point x for the relationship between stress σ and strain rate ε tensors:

$$\sigma = L(\varepsilon)\varepsilon \tag{1.1}$$

[$L(\varepsilon)$ is viscosity tensor of the fourth rank governing the linear and nonlinear rheological properties of the medium]. It is possible, by means of tensor L, to describe the mechanism of bulk viscosity and component anisotropy.

Let matrix v_0, with characteristic function V_0 and tensor L_0, contain a random assembly $X = (V_k, x_k, \omega_k)$ of ellipsoids v_k with characteristic functions V_k, centers x_k, forming a Poisson point field with semiaxes a_k^i (i = 1, 2, 3; $a_k^1 \geq a_k^2 \geq a_k^3$), a set of Euler angles ω_k, and tensors $L_0 + L_1^{(k)}$. Here and below, it is assumed that all of the random values are statistically uniform and are ergodic fields, and their mathematical expectation coincides with components X_α averaged for the volume:

$$\langle(\cdot)\rangle_\alpha = (\operatorname{mes} v_\alpha)^{-1} \int (\cdot)\, V_\alpha(x)\, dx, \quad \langle(\cdot)\rangle = (\operatorname{mes} w)^{-1} \int (\cdot)\, W(x)\, dx \quad (\alpha = 0, 1, \ldots),$$

$w = \bigcup_{\alpha=0} v_\alpha$, $W = \sum_{\alpha=0} V_\alpha$, $\langle(\cdot)|x_1\rangle$ is conditional average for the assembly of field X, assuming that at point x_1 there is an inclusion v_1.

It is assumed that the hydrodynamics of components are described by equations of viscous flow and only hydrodynamic interaction exists between inclusions. Brownian movement of inclusions is not considered. It is assumed that L is determined by the first invariant of the strain tensor $I_1 = \varepsilon_{ii}$ and by the second invariant of the strain rate deviator $I_2 = e_{ij}e_{ij}$, $e_{ij} = \varepsilon_{ij} - \varepsilon_0\delta_{ij}$, $\varepsilon_0 = \varepsilon_{ii}/3$. In particular, for isotropic components we shall use $L = (3L^1, 2L^2) = 3L^1N_1 + 2L^2N_2$, $N_1 = \delta_{ij}\delta_{k\ell}/3$, $N_2 = (\delta_{i\ell}\delta_{jk}\delta_{j\ell} - 2\delta_{ij}\delta_{k\ell}/3)/2$. With $L^1 = \infty$, L^2 = const there is an incompressible Newtonian fluid, and with $2L^2 = 2\mu_0{}^0(I_2)^{(n-1)/2}$ there is a power fluid.

In the general case system (1.1) is not linear, and in order to use known linear methods of elastic theory [1, 2] for obtaining effective rheological rules it is necessary to linearize the nonlinear equation by making additional assumptions.

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It is assumed that L(x) within the limits of each component depends on average values according to the component of the function of invariants I_1 and I_2. A similar assumption is made in the majority of works for nonlinear problems of structural mechanics [3, 4]. Then tensor L(x) is piecewise-constant in region w, and in analyzing a microinhomogeneous material it is possible to use the effective field method developed in linear elastic theory. With the assumptions adopted, the expression for effective viscosity is found by averaging local Eq. (1.1) [1, 2]:

$$\langle\sigma\rangle = L^* \langle\varepsilon\rangle, \quad L^* = L_0 + \sum_k \langle L_1^{(k)} \bar{B}_k V_k \rangle, \tag{1.2}$$

where constant tensor $\bar{B}_k$ describes the average concentration of strain rates in the k-th inclusion $\langle\varepsilon V_k\rangle_k = \bar{B}_k\langle\varepsilon\rangle$. Since with the assumptions adopted, L_1 is a piecewise-constant function of the coordinates, then in estimating $\bar{B}_k$ it is possible to use known methods for transforming the equilibrium equation $\sigma_{ij,j} = 0$ to an integral equation [1, 2]:

$$\varepsilon(x) = \langle\varepsilon\rangle + \int G(x-y)\{L_1(y)\,\varepsilon(y)\,V(y) - \langle L_1\varepsilon(y)\,V(y)\rangle\}\,dy \tag{1.3}$$

[$G(x - y) = \nabla\nabla U(x - y)$ is expressed in terms of Green tensor U of the equilibrium equation for a uniform medium with parameters L_0, $V(y) = W - V_0$]. Relationship (1.3) is formally similar to that obtained in [1, 2] for the problem of linearly-elastic composite materials with the same differences in which tensors U, L_1, and L_0 are functions of previously unknown strain rates for the components. Therefore, in what follows, details of deriving relationships similar to [1, 2] are omitted.

We fix arbitrary realization of assembly X and we consider an inclusion with number i. We designate in terms of $\bar{\varepsilon}_i$ the local external fields in which the i-th inclusion is found; from (1.3) we find

$$\bar{\varepsilon}_i = \langle\varepsilon\rangle + \int G(x-y)\{L_1(y)\,\varepsilon(y)\,V(y;\,x) - \langle L_1\varepsilon V\rangle\}\,dy \tag{1.4}$$

[$V(y;\,x) = V(y) - V_i(x)$]. In order to calculate the average $\langle\bar{\varepsilon}_i\rangle$ it is necessary to prescribe the structure of the composite which is written by a binary function of distribution $\varphi(x_k,\,\omega_k|x_i,\,\omega_i)$, i.e., the probable location of the k-th inclusion in assembly X with fixed i-th inclusion. Since inclusions do not intersect, then $\varphi(x_k,\,\omega_k|x_i,\,\omega_i) = 0$ in the vicinity of v_{ik}' (with characteristic function V_{ik}') of region v_i. We shall not consider close ordering in inclusion location [5] and percolation effects typical for small inclusions connected with formation of the skeletal structure of a suspension and a sharp increase in viscosity [6]. For simplicity we assume that v_{ik}' is a sphere of radius $a_{ik} = a_k^1 + a_k^3$, and φ is centrally symmetrical:

$$\varphi(x_k,\omega_k|x_i,\omega_i) = \psi(\omega_k)\psi_1(|r|)(\mathrm{mes}\ w)^{-1}. \tag{1.5}$$

Here $|r| = |x_i - x_k|$, $\psi_1(|r|) = 0$ with $r \in v_i'$ and $\psi_1(|r|) \to n_k$ with $|r| \to \infty$; n_k is a countable concentration of inclusions; $c_k \equiv \langle V_k\rangle = (4\pi/3)\langle a_k^1 a_k^2 a_k^3\rangle n_k$. For definiteness we assume that $x_i = 0$, and we average (1.4) for assembly $X(\cdot|x_i,\,\omega_i)$:

$$\langle\bar{\varepsilon}_i\rangle = \langle\varepsilon\rangle + \int G(x-y)\{\langle L_1(y)\,\varepsilon(y)\,V(y;\,0)\,|\,0\rangle - \langle L_1\varepsilon V\rangle\}\,dy. \tag{1.6}$$

2. Solution of the Problem for One and Two Inclusions. In order to determine $\langle\bar{\varepsilon}_i\rangle$ in (1.6) we first consider the particular case of an isolated inclusion v_i in an infinite matrix with a uniform field $\varepsilon^0 = L_0^{-1}\sigma^0 = \mathrm{const}$, prescribed at infinity. Since L_0, $L_1 = \mathrm{const}$ in the matrix and inclusion, then ε^0 according to the theorem of polynomial conservativeness [7], clearly determines the uniform strain rate field within the i-th inclusion:

$$\varepsilon_i(x) = A_i\varepsilon^0, \quad A_i = (I + P_i L_1^{(i)})^{-1}, \quad \sigma_i = \bar{B}_i\sigma^0, \quad \bar{B}_i = (L_0 + L_1^{(i)})\,A_i L_0^{-1}\sigma^0, \tag{2.1}$$

where $x \in v_i$ and constant tensor $P_i = -\int G(x - y)V_i(y)dy$ ($x \in v_i$ for isotropic L_0 considered in this work is known [7]). Since L_0 and L_1 depend on ε, then (2.1) may be solved similarly to [8] by the method of successive approximations. As numerical examples have shown, convergence developed after five to seven iterations.

For two inclusions v_i and v_j in an infinite matrix from (1.3) assuming uniformity of field ε^0 in the vicinity of each inclusion we obtain, by the method of successive approximations [1],

$$L_1(y_i)\varepsilon(y_i)\,\mathrm{mes}\,v_i = R_i J_{ij}\varepsilon^0,\quad SL_1(y_i)\varepsilon(y_i)\,\mathrm{mes}\,v_i = (J_{ij}-I)\varepsilon^0; \tag{2.2}$$

$$J_{ij} = \sum_{n=0}^{\infty}\sum_{l=0}^{1}(SR_jSR_i)^n(SR_j)^l,$$
$$S = (\mathrm{mes}\,v_i\,\mathrm{mes}\,v_j)^{-1}\int V_i(y)\,dy\int V_j(x)\,G(x-y)\,dx,\quad R_i = L_1(y_i)A_i\,\mathrm{mes}\,v_i. \tag{2.3}$$

Thus, inclusion v_i is found in a uniform field depending on geometric and rheological properties of the inclusion in question.

In the future it is necessary for us to estimate $\langle\langle J_{ij}\rangle\rangle_{ij}$ for the average value of tensor J_{ij} with respect to ω_i and ω_j in a sphere of radius $|r| = |x_i - x_j|$ with a center at x_i. It is noted that $S(|r|) \to G(|r|)$ with $|r| \to \infty$, and, therefore, we take the point approximation for the inclusion $S(|r|) = G(|r|)$ [1, 2]. For three terms of expansion (2.3), $\langle\langle J_{ij}\rangle\rangle_{ij} = I + J^0(|r|) \equiv I + \langle\langle SR_jSR_i\rangle\rangle_{ij}$. For an isotropic matrix and equally probable orientation of inclusions, tensor J^0 appears to be isotropic:

$$J^0(|r|) = 3J_1^0N_1 + 2J_2^0N_2,\quad 3J_1^0 = 2\xi^2(3\bar{k}_j)(2\bar{\mu}_i)|r|^{-6},$$
$$2J_2^0 = (2/5)\,[\xi^2(3\bar{k}_i)(2\bar{\mu}_j) + (2\bar{\mu}_i)(2\bar{\mu}_j)(7\gamma^2 - \eta^2/4 + 2\xi\eta)]\,|r|^{-6},$$
$$\xi = (3k_0+4\mu_0)^{-1},\ \eta = (3\mu_0)^{-1},\ \gamma = (3k_0+4\mu_0)\,[3\mu_0(3k_0+4\mu_0)]^{-1},$$
$$(3\bar{k}_i, 2\bar{\mu}_i) = \int L_1(y_i)\,A_i\,d\omega_i\prod_{n=1}^{3}a_i^n,\quad L_0 = (3k_0, 2\mu_0).$$

3. <u>Estimation of Effective Rheological Parameters L^*.</u> In obtaining an expression for $\langle\varepsilon_i\rangle$ from (1.6) we take the hypothesis of an effective field [1, 2] according to which the i-th inclusion is in a uniform field $\bar{\varepsilon}_i$ depending on geometrical and rheological properties of v_i, and also each pair of inclusions v_i, v_j exists in a uniform field $\bar{\varepsilon}_{ij} = \bar{\varepsilon} = \mathrm{const}$, independent of the properties of the pair in question. Then, in order to calculate nominal averages in (1.6), we use relationships (2.1) and (2.2) with three terms in the series and substitution in them of ε^0 by $\bar{\varepsilon}(x)$. From (1.2), (1.6), and (2.1) we obtain

$$\langle\bar{\varepsilon}(x_i)\rangle = D\langle\varepsilon\rangle,\quad D = \left\{I(1-c) + \langle AV\rangle - \int\langle\langle J_{ij}(1-V_{ij}^0)\rangle\rangle_{ij}\,dx\right\}^{-1},$$
$$L^* = L_0 + \langle R\rangle D,\ \bar{B}_k = A_kD. \tag{3.1}$$

Until now it has been assumed that L_0 and L_1 are known, but according to the assumption they depend on unknown strain rate fields in the compounds. Therefore, we make a number of assumptions. Let $L_i = L_{i0}g(I_1, I_2)$, where g is a scalar function of invariants I_1 and I_2, and similarly to [3] we take the hypothesis of absence of fluctuations in g not only within the limits of component X_α, but also within the whole volume w: $g(I_1, I_2) = \mathrm{const}$. Then GL_1, A, GR_i, and D are constant tensors independent of strain rate, and the problem of estimating effective properties for the material is linear:

$$L^* = L_0^*g(I_1, I_2),\quad L_0^* = L_{00} + \sum_{i=1}^{N}L_{i0}A_iDc_i$$

(as I_1 and I_2 it is natural to take invariants of tensor $\langle\varepsilon\rangle$).

We weaken the assumption of uniformity for functions taking account of nonlinear properties for the material, and, similar to [4], we shall assume that L_0 and L_1 are determined by average values of the invariants for tensor ε within the limits of the component in question. In view of the uniformity of field ε_i within an inclusion for the second invariant we shall assume approximately $I_{2k} = \langle e_{ij}\rangle_k\langle e_{ij}\rangle_k$. The expression for the second invariant in matrix I_{20} is found by means of apparent relationships

$$\langle V_k'\varepsilon'\rangle = c_k(\langle\varepsilon\rangle_k - \langle\varepsilon\rangle),$$
$$(1-c)\langle(L_0\varepsilon)\varepsilon\rangle_0 = \langle L_0\varepsilon\rangle\langle\varepsilon\rangle + \langle L_0\varepsilon'\varepsilon'\rangle - \sum c_k\langle L_0\varepsilon\rangle_k\langle\varepsilon\rangle_k$$

(a prime denotes fluctuations $q' = q - \langle q\rangle$). Then, considering that with transformation of the volumetric integral according to the first Green equation the value of surface integral with respect to ∂w related to mes w tends toward zero with mes $w \to \infty$, and we have

$$\langle L_0\varepsilon'\varepsilon'\rangle = -\sum c_k\langle L_1^{(k)}\varepsilon\rangle_k(\langle\varepsilon\rangle_k - \langle\varepsilon\rangle).$$

Thus, invariants $I_{1\alpha}$ and $I_{2\alpha}$ only depend on uniform fields and they may be expressed by means of the relationships

$$\begin{gathered} I_{2k} = (N_2A_kD\langle\varepsilon\rangle)(N_2A_kD\langle\varepsilon\rangle), \\ I_{20} = (2\mu_0)^{-1}\{[(L^*\langle\varepsilon\rangle)\langle\varepsilon\rangle - (\langle(L_0 + \\ + L_1)ADV\rangle\langle\varepsilon\rangle)(\langle A_kD\rangle\langle\varepsilon\rangle)](1-c)^{-1} - 3^{-1}k_0I_{10}^2\}, \\ I_{1k} = \delta A_kD\langle\varepsilon\rangle,\ I_{10} = \delta(I - \langle AV\rangle D)\langle\varepsilon\rangle/(1-c),\ \delta = \delta_{ij}. \end{gathered} \tag{3.2}$$

In deriving (3.2), an assumption was made: $\langle\varepsilon_{ii}\sigma_{jj}\rangle_0 = 3k_0I_{10}^2$, which is accurate for an incompressible matrix. Since in Eqs. (2.1) and (3.1) A_k and D depend on invariants $I_{1\alpha}$ and $I_{2\alpha}$ ($\alpha = 0, 1, \ldots$), then in the general case $I_{1\alpha}$ and $I_{2\alpha}$ may be found by the method of successive approximations. In fact, in the zero iteration it is assumed that $I_{1\alpha} = \langle\varepsilon_{ii}\rangle$, $I_{2\alpha} = \langle e_{ij}\rangle\langle e_{ij}\rangle$, then successively we find the zero approximation for A_k and D, the first approximation for $I_{1\alpha}$, $I_{2\alpha}$, L_0, L_1, A_k, D, etc.

4. Example. We consider cases of practical importance when it is possible to construct L^* in explicit form. Here it relates to a material with absolutely rigid inclusions and pores when I_{11}, $I_{21} = 0$ and $L_0 + L_1 = 0$.

Let hard spherical inclusions of a single size be found in an incompressible matrix. Then

$$I_{20} = I_2(\langle\varepsilon\rangle)(1-c)^{-1}f(c), \tag{4.1}$$

where $f(c) = 1 + 5c(1 - 31c/16)^{-1}/2$ and, for example, for a power matrix with $L_0^1 = \infty$, $2L_0^2 = 2\mu_0^0(I_{20})^{(n-1)/2}$

$$L^* = (\infty,\ 2\mu_0^0 f(c)^{(n+1)/2}(1-c)^{(1-n)/2}(\langle e_{ij}\rangle\langle e_{ij}\rangle)^{(n-1)/2}). \tag{4.2}$$

For a Newtonian fluid n = 1 and (4.2) coincides with a similar relationship in [2]. We compare (4.2) with the results of studies of other authors. In particular, it follows from [3] that

$$L^* = (\infty,\ 2\mu_0^0 f_1(c)(\langle e_{ij}\rangle\langle e_{ij}\rangle)^{(n-1)/2}) \tag{4.3}$$

[$f_1(c) = 1 + 5c(1 - c)^{-1}/2$]. Expression (4.3), in contrast to (4.2), contradicts the relationship $L^* = (\infty,\ 2\mu_0^0 f_2(n, c)(\langle e_{ij}\rangle\langle e_{ij}\rangle)^{(n-1)/2})$ obtained in [9] by the method of analyzing dimensions [$f_2(n, c)$ is a function depending only on n and c]. An expression was found in [10]

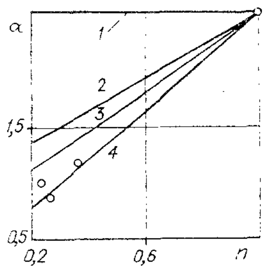

Fig. 1

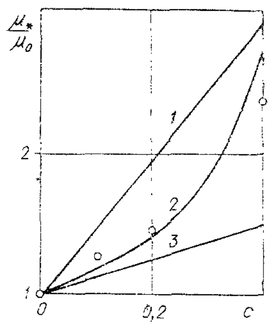

Fig. 2

$$L^* = \left(\infty,\ 2\mu_0^0 \left[\frac{(c/c_{max})^{1/3}}{(1 - c/c_{max})^{1/3}}\right]^{2n-1} (\langle e_{ij}\rangle \langle e_{ij}\rangle)^{(n-1)/2}\right),$$

leading to contradictory results with $n < 0.5$ (viscosity falls with an increase in c); c_{max} is the maximum degree of inclusion packing for a given fractional makeup.

We compare calculated curves $L^* = L^*(c, n)$ for different methods with an experiment [11, 12]. For a Newtonian matrix $n = 1$ in [2] a comparison is provided with an experiment up to $c = 0.43$; it was demonstrated that calculated estimates for shear modulus by the effective field method [1, 2] are more accurate by a factor of two compared with theoretical results of other authors. We consider the opposite case $n \neq 1$, $c \to 0$. Presented in Fig. 1 are experimental [12] and calculated data on coordinates $\beta = [\mu^*/\mu_0 - 1]/c \sim n$. Curves 1-4 were calculated by Eqs. (4.3), [13], (4.2), and [12], respectively; relationships in [12, 13] were obtained assuming infinite smallness for concentration c. Equation (4.2) is valid over a wide range of values of c, and it gives satisfactory agreement with an experiment. Shown in Fig. 2 is comparison of experimental data [12] ($n = 0.41$) with curves 1-3 calculated by methods in [9], (4.2), and in [11].

For spherical pores of a single size in an incompressible power matrix

$$L^* = 2\mu_0^0 [f_3 (1 - c)^{-1} \langle \varepsilon_{ii}\rangle^2 + 2f_4 (1 - c)^{-1} \langle e_{ij}\rangle \langle e_{ij}\rangle]^{(n-1)/2} (3f_3, 2f_4),$$
$$3f_3 = 2(1 - 29/24c)/c,\ 2f_4 = (1 - 35/24c)(1 + 5/24c)^{-1}.$$

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