## EFFECT OF STRUCTURE OF COMPOSITE MATERIALS ON THEIR ELASTIC PROPERTIES

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A statistical formulation is proposed for determining the bounds of elastic macrocharacteristics of a composite material by using classical variational theorems of the elasticity theory. The calculation scheme provided takes into account the correlation effects of the second and third order; it also provides estimates for elastic macrocharacteristics by taking into account the constant, the latter being that physical characteristic of the material which reflects its structure.

1. A microinhomogeneous elastic body is considered with an elastic moduli tensor given by

$$\lambda_{ij_{\kappa}l} = \gamma \delta_{ij} \delta_{kl} + \mu \left( \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \frac{2}{3} \delta_{ij} \delta_{kl} \right)$$

The bulk and the shear moduli  $\gamma$  and  $\mu$  are considered to be homogeneous and isotropic ergodic functions of the spatial coordinates. The stress-strain state of the medium under consideration can be described by the Hooke's law which relates the stresses  $\sigma_{ii}$  and strains  $\varepsilon_{ij}$  by means of

$$\sigma_{ij} = (\gamma - 2_{3}\mu) \varepsilon_{kk} \delta_{ij} + 2\mu \varepsilon_{ij}$$
(1.1)

together with the equilibrium equations and the Cauchy relations,

(σ

$$\sigma'_{ij,j} = 0 \tag{1.2}$$

$$\varepsilon_{ij}' = \frac{1}{2} (u_{i,j}' + u_{j,i}')$$
(1.3)

In the above and in our further considerations it is assumed that the stress-strain state is macrohomogeneous, that is,

$$\langle \sigma_{ij} \rangle = \text{const}, \quad \langle \varepsilon_{ij} \rangle = \text{const}$$
 (1.4)

The angular brackets denote the mathematical expectation; the dashes refer to deviations of functions from their mathematical expectations.

If the stress and strain tensors are represented in their spherical-cum-deviatory form, the relation (1.1) assumes the form

$$\sigma = 3\gamma\varepsilon \tag{1.5}$$

$$S_{ij} = \Delta \mu e_{ij}$$

$$= \sigma_{kk}, \ \varepsilon = \varepsilon_{kk}, \ S_{ij} = \sigma_{ij} - \frac{1}{3} \mathrm{sd}_{ij}, \ e_{ij} = \varepsilon_{ij} - \frac{1}{3} \mathrm{sd}_{ij})$$

$$(1.6)$$

Our problem consists in determining the bounds  $\gamma^*$  and  $\mu^*$  of the elastic macrocharacteristics which can be introduced by the relations

$$\langle \sigma \rangle = 3\gamma^* \langle \varepsilon \rangle, \quad \langle S_{ij} \rangle = 2\mu^* \langle e_{ij} \rangle \tag{1.7}$$

Let the displacements  $u_i^{\theta}$  be given on the surface S which bounds the volume V in such a way that the relation

$$u_i^{\circ} = \langle \varepsilon_{ij} \rangle x_j \tag{1.8}$$

is valid for their mean values on S.

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Then when (1.7) is used, the elastic potential of the volume V is given by [1]

$$2E = (\gamma^* \langle \varepsilon \rangle^2 + 2\mu^* \langle e_{ij} \rangle \langle e_{ij} \rangle) V$$
(1.9)

In accordance with the principle of least potential energy [2] for any virtual-strain field which satisfies (1.7) the following inequality is valid:

$$\frac{2E}{V} \leqslant \frac{1}{V} \int_{V} (\gamma \epsilon^2 + 2\mu e_{ij} e_{ij}) dV$$
(1.10)

If it is assumed that the scale of heterogeneities is small compared with the macrovolume, then since the homogeneous random fields are ergodic, the integration over the volume V corresponds to the process of averaging. Together with (1.10) one also obtains

$$\begin{array}{l} \gamma^{*} \langle \varepsilon \rangle^{2} + 2\mu^{*} \langle e_{ij} \rangle \langle e_{ij} \rangle \langle \langle e_{ij} \rangle \langle \varepsilon \rangle^{2} + 2 \langle \varepsilon \rangle \langle \gamma^{\prime} \varepsilon^{\prime} \rangle \\ + \langle \gamma \rangle \langle \varepsilon^{\prime} \varepsilon^{\prime} \rangle + \langle \gamma^{\prime} \varepsilon^{\prime} \varepsilon^{\prime} \rangle + 2[\langle \mu \rangle \langle e_{ij} \rangle \langle e_{ij} \rangle + 2 \langle e_{ij} \rangle \langle \mu^{\prime} e_{ij}^{\prime} \rangle + \langle \mu \rangle \langle e_{ij}^{\prime} e_{ij}^{\prime} \rangle \\ + \langle \mu^{\prime} e_{ij}^{\prime} e_{ij}^{\prime} \rangle] \end{array}$$

$$(1.11)$$

The virtual-strain field  $\epsilon_{i\,j}{}^\prime$  is introduced with the aid of the relations

$$\sigma' = \gamma' c + \varepsilon' d \tag{1.12}$$

$$S_{ij}' = \mu' a_{ij} + e_{ij}' b$$
 (1.13)

where  $a_{ij}$ , b, c, d are nonrandom quantities to be determined. In solving the system of equations (1.2), (1.3), (1.12), and (1.13) the Fourier-transform method can be employed; thus the fluctuations  $\varepsilon_{ij}$ ' are determined. With the aid of the latter the correlation moments appearing in (1.11) can now be determined; this inequality provides the upper bounds for the elastic characteristics. To obtain a lower-bound estimate for the bulk and shear moduli, the problem must be solved in stresses by having a system of loads on the entire surface S bending the volume V which would ensure a homogeneous stressed state,

$$P_i = \langle \sigma_{ij} \rangle \ n_j \tag{1.14}$$

In this case the principle of least potential energy for a statically feasible stress field  $\sigma_{ij}$ ' results in the inequality [2]

$$\frac{1}{9} \langle \sigma \rangle \frac{1}{\gamma^{*}} \langle \sigma \rangle + \langle S_{ij} \rangle \frac{1}{2\mu^{*}} \langle S_{ij} \rangle \geq \frac{1}{9} [\langle \kappa \rangle \langle \sigma \rangle^{2} + 2 \langle \sigma \rangle \langle \sigma' \kappa' \rangle + \langle \kappa \rangle \langle \sigma' \sigma' \rangle 
+ \langle \kappa' \sigma' \sigma' \rangle] + \frac{1}{2} [\langle \nu \rangle \langle S_{ij} \rangle \langle S_{ij} \rangle + 2 \langle S_{ij} \rangle \langle \nu' S_{ij}' \rangle 
+ \langle \nu \rangle \langle S_{ij}' S_{ij}' \rangle + \langle \nu' S_{ij}' S_{ij}' \rangle], \quad \kappa = \gamma^{-1}, \quad \nu = \mu^{-1}$$
(1.15)

The statically feasible stress field is introduced by means of the relations

$$\varepsilon' = \varkappa' c^{\circ} + d^{\circ} \sigma' \tag{1.16}$$

$$e_{ij}' = v' a_{ij}^{\circ} + S_{ij}' b^{\circ} \tag{1.17}$$

which contain the nonrandom quantities  $a_{ij}^{0}$ ,  $b^{0}$ ,  $c^{0}$ ,  $d^{0}$ .

Moreover, one should use Fourier transforms together with (1.16) and (1.17) to solve the compatibility equation which, if (1.4) holds, is

$$e_{imk}e_{jnl}\frac{\partial^2 e_{mn'}}{\partial x_k \partial x_l} = 0 \tag{1.18}$$

In the above  $e_{ijk}$  is a Levi-Civita tensor. Subsequently, the correlation moments which appear in the inequality (1.15) can be computed; this provides lower bounds for  $\gamma^*$  and  $\mu^*$ , that is, the determination of the bounds for macrocharacteristics has been obtained.

It is noted that in view of isotropy of general properties of the elastic medium under consideration, it is required that only two elastic constants be determined,  $\gamma^*$  and  $\mu^*$ . There is no need, therefore, to introduce a completely arbitrary general deformation. It is sufficient to consider deformation of only two independent types.

2. To determine the bounds of the shear modulus, the uniform state is considered of the elastic composition in which the phase regions are right continuous cylinders with parallel generating lines and of arbitrary cross section. It is assumed that the composition is macroscopically homogeneous and transversely isotropic. The coordinate system is selected such that the  $x_2$  axis is identical with the generators of the cylinders and with the direction of the shear. The elastic displacements corresponding to this problem are as follows:

u

$$u_1 = u_1(x_i), \quad u_2 = u_3 = 0 \quad (i = 2, 3)$$
 (2.1)

and the stress-strain state in the directions  $x_2$ ,  $x_3$  does not depend on  $x_1$ . In our coordinate system the relation (1.13) becomes

$$S_{1i}' = \mu' a_{1i} + e_{1i}' b \tag{2.2}$$

and, moreover,

$$\sigma_{11} = \sigma_{22} = \sigma_{33} = \sigma_{23} = 0 \tag{2.3}$$

The solution of the system of equations (1.2), (1.3), (2.1), and (2.2) by using Fourier transforms is of the form

$$e_{1i}' = \int_{-\infty}^{+\infty} -g\xi_{1k} \frac{\omega_k \omega_i}{\omega^2} e^{i\omega_n x_n} d\omega \quad (\xi_{1k} = a_{1k} / b, \, \omega^2 = \omega_i \omega_i, \, i = 2, \, 3)$$
(2.4)

where  $\omega_i$  are parameters of the two-variables Fourier transformation, and g is a function of the  $\omega_i$  variables which gives the spectral distribution, namely,

$$\mu' = \int_{-\infty}^{+\infty} g e^{i\omega_n x_n} d\omega$$
 (2.5)

Since the random function  $\mu'$  is isotropic therefore [3]

$$\langle g(\omega_i) g(\omega_i') \rangle = \Lambda(\omega^2) \delta(\omega_i + \omega_i') \langle g(\omega_i) g(\omega_i') g(\omega_i'') \rangle = F(\omega^2, \omega_i, \omega_i', \omega'^2) \delta(\omega_i + \omega_i' + \omega_i'')$$

$$(2.6)$$

where A and F are three-dimensional spectral densities of the function  $\mu$ ', and  $\delta$  is the Dirac's delta function. From (2.4) and (2.5) [4] one obtains by using (2.6)

$$\langle \mu' e_{1i}' \rangle = -\frac{1}{2} D_{\mu} \xi_{1i}, \quad \langle e_{1i}' e_{1i}' \rangle = \frac{1}{2} D_{\mu} \xi_{1i} \xi_{1i}$$

Here  $D_{\mu}$  denotes the variance of random function,

$$D_{\mu} = \int\limits_{-\infty}^{+\infty} \Lambda \left( \omega^2 
ight) e^{i \omega_n x_n} \, d \omega$$

To evaluate the correlation functions of the third order one makes use of the fact that the integral

$$f_{kl} = \int_{-\infty}^{+\infty} F(\omega^2, \omega_i \omega_i', \omega'^2) \frac{\omega_k' \omega_l'}{\omega'^2} d\omega'$$

is an isotropic tensor function of the second rank; consequently, it can be represented in the form [5]

$$f_{kl} = A (\omega^2) \delta_{kl} + B (\omega^2) \omega_k \omega_l / \omega^2$$

One obtains

$$\int_{-\infty}^{+\infty} f_{kl} \frac{\omega_i \omega_j}{\omega^2} d\omega = A^* \delta_{ij} \delta_{kl} + B^* \left( \delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} \right)$$
(2.7)

Hence by convolution with respect to all subscripts an expression follows relating the quantities  $A^*$  and  $B^*$  to the single-point correlation moment of the third order of the function  $\mu$ ', the latter being denoted by

$$\int_{-\infty}^{+\infty} f_{pp} d\omega = \int_{-\infty}^{+\infty} F(\omega^2, \omega_i \omega_i', \omega'^2) d\omega d\omega' = 4A^* + 8B^* = m_{\mu}$$
(2.8)

By employing now (2.3)-(2.5) together with (2.7) and (2.8), one obtains

$$\langle \mu' e_{1i}' e_{1i}' \rangle = \xi_{1k} \xi_{1l} \int_{-\infty}^{+\infty} \frac{\omega_k \omega_i}{\omega^2} \, d\omega \int_{-\infty}^{+\infty} F \frac{\omega_l' \omega_i'}{\omega'^2} \, d\omega' = \left(\frac{m_{\mu}}{2} - 2A^*\right) \xi_{1i} \xi_{1i}$$

By using the obtained correlation moments one now finds the right-hand side of the inequality (1.11) which in this case assumes the form

$$\mu^* \langle e_{1i} \rangle \langle e_{1i} \rangle \leq \langle \mu \rangle \langle e_{1i} \rangle + 2 \langle e_{1i} \rangle \langle \mu' e_{1i'} \rangle + \langle \mu \rangle \langle e_{1i'} e_{1i'} \rangle + \langle \mu' e_{1i'} e_{1i'} \rangle$$
(2.9)

containing an unknown parameter  $\xi_{1i}$ . One minimizes the right-hand side of (2.9) with respect to the parameter and one finds

$$\mathbf{I}^* \langle e_{1i} \rangle \langle e_{1i} \rangle \leq \langle \mu \rangle \langle e_{1i} \rangle \langle e_{1i} \rangle - \frac{1}{2} D_{\mu} \langle e_{1i} \rangle \xi_{1i}$$
(2.10)

It follows from (2.10) that

$$\mu^{(+)} = \langle \mu \rangle - \frac{1}{2} \frac{D_{\mu}^{2}}{\langle \mu \rangle D_{\mu} + m_{\mu} - 2A^{*}}$$
(2.11)

is an upper bound for the shear modulus.

3. A lower bound for the shear modulus is found if the same method is employed to solve the system (1.17) and (1.18), thus

$$S_{ij}' = \int_{-\infty}^{+\infty} \rho \eta_{1l} \left( \frac{\omega_l \omega_i}{\omega^2} - \delta_{li} \right) e^{i\omega_m x_m} d\omega, \quad \eta_{1l} = a_{1l}^\circ / b^\circ$$
(3.1)

In the above  $\rho$  is a function of the variables w<sub>i</sub>, which determines the spectral expansion, namely

$$\mathbf{v}' = \int_{-\infty}^{+\infty} \rho e^{i\omega_n x_n} d\omega$$

Using the isotropy assumption for the random function  $\nu$ ' one obtains

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$$\langle \mathbf{v}' S_{1i}' \rangle = -\frac{1}{2} D_{\mathbf{v}} \eta_{1i}, \qquad \langle S_{1i}' S_{1i}' \rangle = \frac{1}{2} D_{\mathbf{v}} \eta_{1i} \eta_{1i}$$

$$\langle S_{1i}' S_{1i}' \mathbf{v}' \rangle = (m_{\mathbf{v}}^{q_{\mathbf{j}}} - 2A^*) \eta_{1i} \eta_{1i}$$

$$(3.2)$$

where  $D_{\nu}$  is the variance and  $m_{\nu}$  the one-point correlation moment of the third order of the random function  $\nu'$ ,  $A_1^*$  is a constant.

By inserting (3.2) in (1.15) and using (2.2), and having minimized the right-hand side of (1.15) with respect to the parameter  $\eta_{ii}$ , one obtains

$$\frac{1}{\mu^{*}} \langle S_{1i} \rangle \langle S_{1i} \rangle \leqslant \langle S_{1i} \rangle \langle v \rangle \langle S_{1i} \rangle - \frac{1}{2} D_{v} \langle S_{1i} \rangle \eta_{1i}$$

Hence a lower bound is obtained of the shear modulus:

$$\mu^{(-)} = \left[ \langle v \rangle - \frac{1}{2} \frac{D_v^2}{\langle v \rangle D_v + m_v - 2A_1^*} \right]^{-1}$$
(3.3)

4. In the estimates (2.10) and (3.3) there appear the unknowns  $A^*$  and  $A_1^*$  which are of dimensions  $\mu^3$  and  $\mu^{-3}$  respectively; they represent the physical characteristics of the material. For a two-component composition the relation between  $A^*$  and  $A_1^*$  is easily established by expressing  $\nu'$  in terms of  $\mu'$ . To this end, a composition is considered whose shear moduli of the components are  $\mu_1$  and  $\mu_2$ . Suppose that the concentration of the first component is c. Then the fluctuations of  $\mu$  and  $\nu$  are given by

$$\begin{array}{l} \mu_1' = (1-c) \ (\mu_1 - \mu_2), \quad \mu_2' = -c \ (\mu_2 - \mu_1) \\ \nu_1' = (1-c) \ (\mu_2 - \mu_1) \ (\mu_1 \mu_2)^{-1}, \quad \nu_2' = -c \ (\mu_2 - \mu_1) \ (\mu_1 \mu_2)^{-1} \end{array}$$

If one formally considers a two-component medium with fluctuations in its shear modulus,

 $\varkappa_1' = (1 - c), \ \varkappa_2' = -c$ 

then  $\mu_1'$ ,  $\mu_2'$ ,  $\nu_1'$  and  $\nu_2'$  are expressed in terms of  $\kappa_1'$  and  $\kappa_2'$  as follows:

$$\mu_1' = (\mu_1 - \mu_2) \,\varkappa_1', \ \mu_2 = (\mu_1 - \mu_2) \,\varkappa_2' \tag{4.1}$$

By comparing (4.1) and (4.2) one finds that

$$\mathbf{v}' = -\mu'(\mu_1 \mu_2)^{-1} \tag{4.3}$$

From (4.3) one obtains the relation

$$m_{\nu} = -m_{\mu} (\mu_1 \mu_2)^{-3}$$

Thus (3.2) can be rewritten as

$$\langle S_{1i}' \mathbf{v}' S_{1i}' \rangle = -(m_{\mu}^{1/2} - 2A^*) (\mu_1 \mu_2)^{-3} \eta_{1i} \eta_{1i}$$
(4.4)

By comparing (4.4) and (3.2) one finds the sought relation

$$A_1^* = -A^* (\mu_1 \mu_2)^{-3} \tag{4.5}$$

Employing (4.5), one can express the upper and lower bounds of the shear modulus in terms of the concentrations and the shear moduli of the components; in the dimensionless form they become

$$\mu_{0} = cn + (1 - c) - \frac{1}{2} \frac{(n - 1)^{2} (1 - c) c}{cn + (1 - c) + (n - 1) (1 - 2c) - \alpha}$$

$$\mu_{00} = 2n \frac{c + (1 - c) n - (n - 1) (1 - 2c) + \alpha}{2 [c + (1 - c) n] [c + n (1 - c) - (n - 1) (1 - 2c) + \alpha] + (n - 1)^{2} (1 - c) c}$$

$$(\mu_{0} = \mu^{(+)} \mu_{2}^{-1}, \mu_{00} = \mu^{(-)} \mu_{2}^{-1}, n = \mu_{1} \mu_{2}^{-1},$$

$$\alpha = 2A^{*} \mu_{2}^{-3} (n - 1)^{2} [(1 - c) c]^{-1}, n \ge 1)$$

$$(4.6)$$

It is noted that our problem was formulated for mixtures whose phase regions are of arbitrary cross section. It can, therefore, be assumed that the quantity  $\alpha$  which appears in the estimates (4.6) and (4.7) reflects the effect of the composite material structure on its elastic properties.

The magnitude of  $\alpha$  must be found experimentally; nevertheless, some a priori conclusions can be made as regards its limits.

In Fig. 1,  $\mu_0(\alpha)$  and  $\mu_{00}(\alpha)$  are shown for c=0.7 and n=5. The branches I and II show  $\mu_{00}$  and the branches III and IV  $\mu_0$ . The values  $-\infty < \alpha \le a$  and  $d \le \alpha < \infty$  are, of course, excluded from our considerations since otherwise  $\mu_0$  exceeds  $\langle \mu \rangle \mu_2^{-1}$  and  $\mu_{00}$  is less than  $\langle \mu^{-1} \rangle \mu_2^{-1}$  the latter being inconsistent with the theorems of Feucht and Reuss. We then exclude from our considerations  $a < \alpha < b$ ,  $c < \alpha < d$ , since  $\mu_0 < \mu_{00}$  for these values of  $\alpha$ . Consequently,  $\alpha$  can only assume the values  $b \le \alpha \le c$ , that is, the definition region of  $\alpha$  is an interval between two points at which  $\mu_0 = \mu_{00}$ . It is not difficult to show that these points always exist and that their coordinates are

$$\alpha_1 = \frac{1}{2} (n-1) (1-c), \quad \alpha_2 = -\frac{1}{2} (n-1) c$$
(4.8)

The obtained solution is exact for structures with characteristics given by (4.8). Obviously, in a medium with characteristics  $\alpha_2$ , the first element forms a matrix; but in a medium with characteristics  $\alpha_1$ it is the other element. For media with characteristics  $b < \alpha < c$  the shear modulus is located inside the dashed area. If particular features of the mixture structure are ignored and if the values  $\alpha$  (b) and  $\alpha$  (c) are considered as the effective upper and lower bounds for the shear modulus, then they are identical with the bounds obtained in [1]. It is also noticed that (4.6) yields a quantity below the upper bound for the effective shear modulus which can be obtained from [6] in the case of a lengthwise shear.

5. To find estimates for the bulk modulus of a statically isotropic elastic mixture one has to replace the boundary conditions (1.8) and (1.14) by the following:

$$u_i = \langle \boldsymbol{\varepsilon} \rangle x_i, \quad p_i = \langle \boldsymbol{\sigma} \rangle n_i$$

and Eqs. (1.13) and (1.17) by the Eqs. (1.12) and (1.16). As regards the bulk modulus estimates, the consideration of the general case of a stress-strain state does not present any difficulties if it is assumed that the shear moduli of the components are the same. The analysis is similar to that given previously, the difference lying in that in evaluating the correlation moments of the third order it is now necessary to evaluate the integrals

$$\int_{-\infty}^{+\infty} f_{kl} d\omega = (3A^* + 5B^*)^{\delta}_{kl}$$
(5.1)

Convolution in the subscripts k and l enables one to eliminate both constants

$$\int_{-\infty}^{+\infty} f_{pp} d\omega = \iint_{-\infty}^{+\infty} F(\omega^2, \omega_i \omega_i', \omega'^2) d\omega d\omega' = 3(3A^* + 5B^*) = m_{\gamma}$$

where  $m_{\gamma}$  is the third-order correlation moment of the random function  $\gamma'$ . Consequently, the structure characteristic  $\alpha$  does not appear in the estimates for the bulk modulus. The upper and lower bounds of  $\gamma^*$  are given, respectively, by

$$\begin{split} \gamma^{(+)} &= \langle \gamma \rangle - \frac{D_{\gamma}^{2}}{\sqrt[4]{_{3} \langle \mu \rangle + \langle \gamma \rangle D_{\gamma} + m_{\gamma}}} \\ \gamma^{(-)} &= \langle \theta \rangle - 4 \frac{D_{\theta}^{2}}{3 \langle v \rangle D_{\theta} + 4 \langle \theta \rangle D_{\theta} + 4m_{\theta}} \end{split}$$

In the above  $\theta = \gamma^{-1}$ ,  $D_{\gamma}$ ,  $D_0$  are the variances of the functions  $\gamma'$  and  $\theta'$ ,  $m_{\theta}$  is the third-order correlation moment of the random function  $\theta$ . It can easily be shown that both estimates are equal to one another and also to the value of the bulk modulus obtained in [7, 8].



Thus in estimating the elastic characteristics it has become clear that the bulk modulus of an isotropic mixture depends on the concentrations and on separate moduli; the particular features of the mixture structure have no effect on it if the shear moduli are equal. This was established by Hill in [8]. Moreover, it was also found that the shear modulus of an elastic isotropic mixture contains a parameter characteristic for the structure of the mixture. This confirms Hill's assumption [8] that to

determine the shear modulus of a composite it is not only the concentration and the moduli of the components but also the internal geometry which are of importance.

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