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Hill condition and overall properties of composites

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Summary We discuss the Hill principle's role and applications in modern micromechanics of industrial composite materials. Uniform boundary conditions, fundamental in micromechanics, are introduced as a class of Hill solutions. Mixed uniform conditions, basic for experimental testing, are analysed. Domains of application of the Hill principle are reviewed, like homogeneization of heterogeneous media, definition of effective properties and size effect in heterogeneous materials. Generalization of the Hill condition is realized for arbitrary materials, in particular for nonlinear inelastic composites with imperfect interfaces.

Key words Hill principle, micromechanics, composites, effective properties, size effect

1 Introduction

Under the term of the Hill principle, the engineering science usually understands the condition for equivalence (compatibility) of energetically and mechanically defined effective properties of heterogeneous elastic materials.

For a linear elastic material, this principle can be expressed analytically through the stress and strain tensors $\boldsymbol{\sigma}$ and $\boldsymbol{\varepsilon}$ as

$$\langle \boldsymbol{\sigma} : \boldsymbol{\varepsilon} \rangle = \langle \boldsymbol{\sigma} \rangle : \langle \boldsymbol{\varepsilon} \rangle \quad . \tag{1}$$

In (1) and elsewhere forthwith $\langle \mathbf{c} \rangle$ means spatial average of the variable $\mathbf{c}$ in the domain D and the symbol ":" means a twice contracted tensor product. Thus, according to (1), the average of the products equals the product of the averages.

Relationship (1) was obtained by Hill and named after him in [8].

Nonetheless, the significance of this principle turns out to be much more than the mentioned above energetical-mechanical equivalence.

For example, it is useful when considering the size effect and the boundary data influence, that is especially important for specimens smaller than the representative volume. Thus, paper [6] uses the Hill condition for developing the theory of apparent properties for small specimens. In [12], [3] and [4], the Hill condition was used as a basic relation for obtaining the uniform boundary conditions cases and to develop the theory of overall apparent properties for mixed boundary data.

The most fundamental domain of its application is the homogeneization of statistically homogeneous and ergodic random materials. This direction was recently analysed and developed in [16], where the Hill macro- homogeneity principle has been established, and its application to several important classes of heterogeneous materials has been realized.

Together with the concepts of statistical uniformity and representative volume, the Hill principle can be considered as an essential ingredient of modern micromechanics (see [6], [16]).

The aim of the present paper is to generalize the Hill condition for arbitrary constitutive laws, in particular, for nonlinear inelastic materials, and to discuss the role of this principle in

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micromechanics. As the importance of the Hill principle in homogeneization of random and periodic materials has been sufficiently shown in [16], we will mainly concentrate on the first problem.

2
Generalization

The physical meaning of the left-hand side of (1) is nothing else than the average of the free Helmholtz energy. Suppose that for a nonlinear inelastic medium the free energy can be expressed by a functional F

$$F = \boldsymbol{\sigma} * \boldsymbol{\varepsilon} \ . \tag{2}$$

The physical meaning of linear operator $*$ in (2) will be illustrated on particular examples in Sec. 3 and 4. Let the constitutive expressions be

$$\boldsymbol{\sigma} = \boldsymbol{\varphi}(\boldsymbol{\varepsilon}), \boldsymbol{\varepsilon} = \boldsymbol{\psi}(\boldsymbol{\sigma}) \ . \tag{3}$$

In (3), $\boldsymbol{\varphi}$ and $\boldsymbol{\psi}$ are reciprocal time-space functionals.

Energetic and mechanical effective properties, respectively e and m, will be introduced as

$$\langle F \rangle = \boldsymbol{\varphi}^e(\langle \boldsymbol{\varepsilon} \rangle) * \langle \boldsymbol{\varepsilon} \rangle \ , \tag{4}$$

$$\langle \boldsymbol{\sigma} \rangle = \boldsymbol{\varphi}^m(\langle \boldsymbol{\varepsilon} \rangle); \langle \boldsymbol{\varepsilon} \rangle = \boldsymbol{\psi}^m(\langle \boldsymbol{\sigma} \rangle) \ . \tag{5}$$

Superscripts e and m in (4) and (5) mean that the space material functions in (3) are replaced by the constants representing effective material properties.

Then, because of the commutativity of space and time operators, one can write

$$\langle F \rangle = \boldsymbol{\varphi}^e(\langle \boldsymbol{\varepsilon} \rangle) * \langle \boldsymbol{\varepsilon} \rangle = \boldsymbol{\varphi}^e[\boldsymbol{\psi}^m(\langle \boldsymbol{\sigma} \rangle)] * \langle \boldsymbol{\varepsilon} \rangle \ . \tag{6}$$

The condition for the coincidence of energetic and mechanical effective properties leads to

$$\langle F \rangle = \langle \boldsymbol{\sigma} \rangle * \langle \boldsymbol{\varepsilon} \rangle \ . \tag{7}$$

Thus, we obtain a generalization of (1)

$$\langle \boldsymbol{\sigma} * \boldsymbol{\varepsilon} \rangle = \langle \boldsymbol{\sigma} \rangle * \langle \boldsymbol{\varepsilon} \rangle \ . \tag{8}$$

Similar to (1), both sides of Eq. (8) are scalars, because of the tensor operation of double contraction throughout (8). Thus, for nonlinear and inelastic materials in supposition (2), the Hill principle will have a simple form (8).

Still, practical needs demand to present (8) in terms of the boundary data; this is important in micromechanical problems, especially when a specimen under study is smaller than the representative volume of the material, [7].

For materials with perfect interfaces, an identity is known, e.g. [5], [12]

$$\langle \boldsymbol{\sigma} : \boldsymbol{\varepsilon} \rangle - \langle \boldsymbol{\sigma} \rangle : \langle \boldsymbol{\varepsilon} \rangle = \frac{1}{V} \int_{\partial D} (\mathbf{P} - \langle \boldsymbol{\sigma} \rangle \cdot \mathbf{n}) \cdot (\boldsymbol{\xi} - \langle \boldsymbol{\varepsilon} \rangle \cdot \mathbf{x}) \, \mathrm{d}\Sigma \ . \tag{9}$$

In (9), $\mathbf{P}$ and $\boldsymbol{\xi}$ are the traction vector density on the specimen boundary ∂D and the displacement vector, respectively, and $\mathbf{n}$ is the exterior unit normal, V the body volume and $\mathbf{x}$ the radius vector.

For the case of imperfect interfaces, the most frequently used is a model with displacement jumps proportional to their associated traction components, [2]

$$\mathbf{P} = \mathbf{D} \cdot [\boldsymbol{\xi}]; \ \ [\boldsymbol{\xi}] = \mathbf{R}\mathbf{P} \ . \tag{10}$$

In (10), D_{ij} and R_{ij} are the interface parameters. Tensors $\mathbf{D}$ and $\mathbf{R}$ are supposed to be positive-definite, in order to assure the minimum principle's validity. Tensor $\mathbf{R}$ is called in [13] the compliance of the interface.

In [12], the displacement jump $[\xi]$ is decomposed into an opening gap, $\Delta\xi_n$, and a sliding gap, $\Delta\xi_s$, as follows:

$$\Delta\boldsymbol{\xi} = (\mathbf{n}\cdot\Delta\boldsymbol{\xi})\mathbf{n}, \Delta\boldsymbol{\xi}_s \equiv \Delta\boldsymbol{\xi} - \Delta\boldsymbol{\xi}_n \quad . \tag{11}$$

In (11), only nonnegative values of $\mathbf{n}\cdot\Delta\boldsymbol{\xi}$ are admitted, in order to exclude any material penetration.

Then we obtain

$$\begin{aligned}
\langle\boldsymbol{\sigma}:\boldsymbol{\varepsilon}\rangle - \langle\boldsymbol{\sigma}\rangle:\langle\boldsymbol{\varepsilon}\rangle &= \frac{1}{V}\int_D (\boldsymbol{\sigma} - \langle\boldsymbol{\sigma}\rangle):(\boldsymbol{\varepsilon} - \langle\boldsymbol{\varepsilon}\rangle)\,\mathrm{d}V \\
&= \frac{1}{V}\int_{\partial D} (\mathbf{P} - \langle\boldsymbol{\sigma}\rangle\cdot\mathbf{n})\cdot(\boldsymbol{\xi} - \langle\boldsymbol{\varepsilon}\rangle\cdot\mathbf{x})\,\mathrm{d}\Sigma \\
&\quad + \frac{1}{V}\int_{\Gamma} (\mathbf{P} - \langle\boldsymbol{\sigma}\rangle\cdot\mathbf{n})\cdot[\boldsymbol{\xi}]\,\mathrm{d}\Gamma \\
&= \frac{1}{V}\int_{\partial D} (\mathbf{P} - \langle\boldsymbol{\sigma}\rangle\cdot\mathbf{n})\cdot(\boldsymbol{\xi} - \boldsymbol{\varepsilon}_o\cdot\mathbf{x})\,\mathrm{d}\Sigma \quad .
\end{aligned} \tag{12}$$

In (12), we use the notation

$$\boldsymbol{\varepsilon} = \langle\boldsymbol{\varepsilon}\rangle - \frac{1}{V}\int_{\Gamma} sym([\boldsymbol{\xi}]\times\mathbf{n})\,\mathrm{d}\Sigma \quad . \tag{13}$$

In (13), the symbol "$\times$" means the dyadic tensor product, while "*sym*" denotes the symmetric operation. Thus, one can finally generalize (9) as

$$\langle\boldsymbol{\sigma}*\boldsymbol{\varepsilon}\rangle - \langle\boldsymbol{\sigma}\rangle*\langle\boldsymbol{\varepsilon}\rangle = \frac{1}{V}\int_{\partial D} (\mathbf{P} - \langle\boldsymbol{\sigma}\rangle\cdot\mathbf{n})*(\boldsymbol{\xi} - \boldsymbol{\varepsilon}_0\cdot\mathbf{x})\,\mathrm{d}\Sigma \quad , \tag{14}$$

denoting by a tensor operator.

When the LHS of (14) is equal to zero, one obtains three classes of particular solutions that are of special importance in micromechanics.

Case (1): on the whole boundary we have

$$\boldsymbol{\xi} = \boldsymbol{\varepsilon}_o\cdot\mathbf{x} \quad . \tag{15}$$

This case is called $\boldsymbol{\varepsilon}_o$ - *KUBC*, or kinematic uniform boundary conditions.

In (15) $\boldsymbol{\varepsilon}_o$ is a given symmetric nondimensional second-rank tensor with values compatible to those of strain tensors for solids.

The relation between the average strain $\langle\boldsymbol{\varepsilon}\rangle$ and $\boldsymbol{\varepsilon}_o$ is given by (13) as in the linear elastic case. If the interfaces in the material are perfect, then we have a classic result, frequently used in applications

$$\langle\boldsymbol{\varepsilon}\rangle = \boldsymbol{\varepsilon}_o \quad , \tag{16}$$

Case (2): on the whole boundary we have

$$\mathbf{P} = \boldsymbol{\sigma}_o\mathbf{n} \quad . \tag{17}$$

This case is called $\boldsymbol{\sigma}_o$ - *SUBC*, static uniform boundary conditions.

In (17) $\boldsymbol{\sigma}_o$ is a given symmetric second-rank tensor of stress dimension with values compatible to those of stress tensors for solids.

Similarly to the linear elastic case, we obtain

$$\langle\boldsymbol{\sigma}\rangle = \boldsymbol{\sigma}_o \quad . \tag{18}$$

Case (3): A combination of the two previous cases, that makes the integral in (14) equal to zero. This is *MUBC*, or mixed uniform boundary conditions.

A great deal of research in modern micromechanics is done in terms of the first two cases, especially when it concerns size and structure effects. In common engineering practice, however, the *KUBC* and *SUBC* loadings are hardly realisable. That is why almost all the laboratory and industrial testing of heterogeneous specimens (e.g. concrete, composites) is usually done in the mixed boundary conditions. The analysis of the mixed case was initiated in [3] and [4].

In the case of uniform mixed boundary conditions, the formal boundary vector, given *a priori* on the body boundary, is of mixed structure, that is it has one or two components defined by the imposed boundary displacements and the lacking components defined by boundary tractions.

In [4], it was demonstrated that in the case of uniform mixed boundary conditions, with $\partial D = \partial D_{\sigma\xi}$, only two principal loading situations are possible: the boundary vectors of the structure (ξ_1, ξ_2, P_3) and (ξ_1, P_2, P_3). All the other cases can be obtained by simple permutation.

Let us recall the definition of mixed uniform boundary conditions following the classical mixed boundary-value problem definition, e.g. [12]. We call mixed uniform boundary conditions (MUBC) a loading when the imposed surface displacements and complementary surface tractions are defined through the given a priori constant components ε^0_{ij} and σ^0_{ij} symmetrical with respect to the indices i, j

$$\xi_i = \varepsilon^0_{ij} x_j;\ P_k = \sigma^0_{kj} n_j \quad \text{on} \quad \partial D \ , \tag{19}$$

Envisaged also are problems where surface displacements are given on a part ∂D_ξ and surface tractions on a part ∂D_σ, but for the sake of clarity we will limit ourselves to the case (19).

Let us now prove the following statement:

Theorem

Mixed uniform boundary conditions must be orthogonal and can be realized only in materials having at least orthotropic elastic symmetry properties.

Proof Indeed, two principal situations are possible on the boundary, when the boundary vector is respectively (ξ_1, ξ_2, P_3) or (ξ_1, P_2, P_3). All the other versions can be received by simple permutations.

Consider the first case, when one has on the body surface

$$\begin{aligned} \xi_1 &= \varepsilon^0_{1i} x_i \ , \\ \xi_2 &= \varepsilon^0_{2i} x_i \ , \\ P_3 &= \sigma^0_{3j} n_j \ . \end{aligned} \tag{20}$$

Then

$$\begin{aligned} \langle \sigma_{13} \rangle &= \frac{1}{2V} \int_{\partial D} (x_1 P_3 + x_3 P_1) \mathrm{d}\Sigma = \frac{1}{2V} \int_{\partial D} (x_1 \sigma^0_{3i} n_i + x_3 \sigma_{1i} n_i) \mathrm{d}\Sigma \\ &= \frac{\sigma^0_{13}}{2} + \frac{1}{2V} \int_{\partial D} x_3 \sigma_{1i} n_i \mathrm{d}\Sigma = \frac{\sigma^0_{13}}{2} + \frac{1}{2V} \int_D x_3 \sigma_{1i,i} \mathrm{d}V + \frac{1}{2V} \int_D x_{3,i} \sigma_{1i} \mathrm{d}V \\ &= \frac{\sigma^0_{13}}{2} + \frac{1}{2V} \int_{\partial D} x_{3,i} \sigma_{1i} \mathrm{d}\Sigma = \frac{\sigma^0_{13}}{2} + \frac{1}{2V} \int_{\partial D} \sigma_{13} \mathrm{d}V = \frac{1}{2} (\sigma^0_{13} + \langle \sigma_{13} \rangle) \ , \end{aligned} \tag{21}$$

Relation (21) gives then

$$\langle \sigma_{13} \rangle = \sigma^0_{13} \ . \tag{22}$$

In the same fashion one can show that

$$\langle \sigma_{23} \rangle = \sigma^0_{23};\ \langle \sigma_{33} \rangle = \sigma^0_{33} \ . \tag{23}$$

Let us now turn to strains

$$\begin{aligned} \langle \varepsilon_{12} \rangle = \frac{1}{2V} \int_D (\xi_{1,2} + \xi_{2,1}) \mathrm{d}V &= \frac{1}{2V} \int_{\partial D} (\xi_1 n_2 + \xi_2 n_1) \mathrm{d}V \\ &= \frac{1}{2V} \int_D (\varepsilon^0_{1i} x_i n_2 + \varepsilon^0_{2i} x_i n_1) \mathrm{d}\Sigma = \varepsilon^0_{12} \ . \end{aligned} \tag{24}$$

In the same fashion one can show that

$$\langle \varepsilon_{11} \rangle = \varepsilon^0_{11}; \langle \varepsilon_{22} \rangle = \varepsilon^0_{22} \quad . \tag{25}$$

The question rests open only for the components $\langle \varepsilon_{13} \rangle$ and $\langle \varepsilon_{23} \rangle$.

For the sake of clarity, without losing the universality of the results, we give the analysis for the linear elastic case.

Simple transforms show that

$$\begin{aligned}\langle \varepsilon_{13} \rangle &= \frac{1}{2V}\int_D (\xi_{1,3} + \xi_{3,1})\mathrm{d}V = \frac{1}{2V}\int_{\partial D} (\xi_1 n_3 + \xi_3 n_1)\mathrm{d}V \\ &= \frac{1}{2V}\int_D (\varepsilon^0_{1i} x_i n_3 + \xi_3 n_1)\mathrm{d}\Sigma = \frac{1}{2}\varepsilon^0_{13} + \frac{1}{2V}\int_D \xi_{3,1}\mathrm{d}V \quad .\end{aligned} \tag{26}$$

On the other hand, using the effective properties notion, one has

$$\langle \varepsilon_{13} \rangle = S^{\mathrm{eff}}_{13kl} \langle \sigma_{13} \rangle \quad , \tag{27}$$

Relation (27) contains three unknown variables: $\langle \sigma_{11} \rangle$, $\langle \sigma_{12} \rangle$ and $\langle \sigma_{22} \rangle$.

For the averages of strain components, one has through the Hooke's law

$$\begin{aligned}\varepsilon^0_{11} &= S^{\mathrm{eff}}_{11kl} \langle \sigma_{kl} \rangle, \\ \varepsilon^0_{12} &= S^{\mathrm{eff}}_{12kl} \langle \sigma_{kl} \rangle, \\ \varepsilon^0_{22} &= S^{\mathrm{eff}}_{22kl} \langle \sigma_{kl} \rangle \quad .\end{aligned} \tag{28}$$

The system of three linear equations (28) permits to express the unknown components $\langle \sigma_{11} \rangle$, $\langle \sigma_{12} \rangle$ and $\langle \sigma_{22} \rangle$ through the given constants σ^0_{13}, σ^0_{23}, σ^0_{33}, ε^0_{11}, ε^0_{22}, ε^0_{12}. The same is valid through (27) also for $\langle \varepsilon_{13} \rangle$.

One sees that $\langle \varepsilon_{13} \rangle$ does not depend on ε^0_{13}, and this in its turn contradicts to Eq. (26)!

Identical arguments lead to the conclusion that $\langle \varepsilon_{23} \rangle$ does not depend on ε^0_{23}.

This paradox can be resolved under the hypothesis that the material under study has two planes of elastic symmetry: x_1x_3 and x_2x_3, and that

$$\sigma^0_{13} = \sigma^0_{23} = 0 \quad . \tag{29}$$

This leads to

$$\langle \varepsilon_{13} \rangle = \langle \varepsilon_{23} \rangle = 0 \quad . \tag{30}$$

One can draw the conclusion that uniform boundary conditions of the form (ξ_1, ξ_2, P_3) can be realized in a material with two planes of elastic symmetry x_1x_3 and x_2x_3 and under condition (29).

Then the averages of the stress and strain components can be calculated through (21)–(24) and (30).

On the other hand, uniform boundary conditions of the form (ξ_1, P_2, P_3) leads to the symmetry x_1x_2 and x_1x_3, and to the condition

$$\sigma^0_{12} = \sigma^0_{13} = 0 \quad . \tag{31}$$

To conclude, one can state that the described set of mixed uniform boundary conditions (six possible structures of the boundary vector) can be realized in the specimen when and only when the following holds:

$$\langle \varepsilon_{ij} \rangle = \langle \sigma_{ij} \rangle = 0; \text{ for } i \neq j \quad . \tag{32}$$

On the other hand, recalling the Hooke's law for anisotropic bodies, one can see that in the general case of arbitrary values of $\langle \varepsilon_{ii} \rangle$ or $\langle \sigma_{jj} \rangle$, condition (32) can be satisfied only in a material having at least the orthotropic elastic symmetry properties. Hence, for a mixed uniform loading, the given *a priori* matrix of averages must be a diagonal one; thus, the boundary conditions are orthogonal, excluding shear strains/stresses. This completes the proof.

From the theoretical and practical viewpoints it follows, [4], that at uniform mixed boundary conditions the most preferable situation is when all the boundary is submitted to the mixed loading

$$\partial D = \partial D_{\sigma\xi} \quad . \tag{33}$$

One has for surface displacements and complementary surface tractions

$$\xi_i = \langle \varepsilon_{ij} \rangle x_j ; \;\; P_k = \langle \sigma_{kj} \rangle n_j \quad . \tag{34}$$

The proof is in principle valid for the general case of constitutive equations, in particular for nonlinear elastic and for viscoelastic materials.

Let us illustrate this by the example of a nonlinear elastic material with a constitutive equation of the type

$$\varepsilon_{ij} = c_{ijkl}\sigma_{kl} + \mathrm{d}_{ijklmn}\sigma_{kl}\sigma_{mn} + f_{ijklmnpq}\sigma_{kl}\sigma_{mn}\sigma_{pq} + \cdots \quad . \tag{35}$$

Indeed, following the procedure (27)–(28), one has in the nonlinear case

$$\varepsilon^0_{ij} = c^{\mathrm{eff}}_{ijkl}\langle\sigma_{kl}\rangle + \mathrm{d}^{\mathrm{eff}}_{ijklmn}\langle\sigma_{kl}\rangle\langle\sigma_{mn}\rangle + f^{\mathrm{eff}}_{ijklmnpq}\langle\sigma_{kl}\rangle\langle\sigma_{mn}\rangle\langle\sigma_{pq}\rangle + \cdots \quad . \tag{36}$$

This system of three equations with three unknowns gives a unique solution for $\langle\sigma_{11}\rangle$, $\langle\sigma_{12}\rangle$ and $\langle\sigma_{22}\rangle$. For $\langle\varepsilon_{13}\rangle$ one has

$$\langle\varepsilon_{13}\rangle = c^{\mathrm{eff}}_{ijkl}\langle\sigma_{kl}\rangle + \mathrm{d}^{\mathrm{eff}}_{ijklmn}\langle\sigma_{kl}\rangle\langle\sigma_{mn}\rangle + f^{\mathrm{eff}}_{ijklmnpq}\langle\sigma_{kl}\rangle\langle\sigma_{mn}\rangle\langle\sigma_{pq}\rangle + \cdots \quad . \tag{37}$$

Thus, $\langle\varepsilon_{13}\rangle$ is expressed as a function of σ^0_{13}, σ^0_{23}, σ^0_{33}, ε^0_{11}, ε^0_{22}, ε^0_{12}.

One sees again that $\langle\varepsilon_{13}\rangle$ does not depend on ε^0_{13}, and this in its turn contradicts the relationship (26).

The same concerns $\langle\varepsilon_{23}\rangle$. To resolve this paradox we come necessarily to conditions (29) and (30).

Usually the plates of the testing machine are stiffer than the material to be tested. It follows that the displacement vector is, at least approximately, imposed on the faces perpendicular to the loading axis, while the stress vector is imposed on the faces parallel to it. These are free boundaries or boundaries loaded by a hydrostatic pressure. Such an experiment can be treated as a pure example of uniform orthogonal boundary conditions; the Hill condition holds here automatically. The tested material possesses realistically at least orthotropic elastic symmetry properties, and the relation $P_2 = P_3 = 0$ holds.

For example, for a sample with perfect interfaces, this means that for the diagonal elements of the given *a priori* matrix of averages we have, respectively,

$$(\langle\varepsilon_{11}\rangle, \langle\sigma_{22}\rangle, \langle\sigma_{33}\rangle) \quad \text{and} \quad (\langle\varepsilon_{11}\rangle, \langle\varepsilon_{22}\rangle, \langle\sigma_{33}\rangle) \quad . \tag{38}$$

According to the idea of Huet, a specific *hierarchy* of the mixed uniform boundary conditions was established in [4].

$$\begin{pmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{33} \end{pmatrix} ; \begin{pmatrix} \varepsilon_{11} \\ \sigma_{22} \\ \sigma_{33} \end{pmatrix} ; \begin{pmatrix} \varepsilon_{11} \\ \varepsilon_{22} \\ \sigma_{33} \end{pmatrix} ; \begin{pmatrix} \varepsilon_{11} \\ \varepsilon_{22} \\ \varepsilon_{33} \end{pmatrix} \quad , \tag{39}$$

where the two mixed versions are enclosed by the static and kinematic ones. The model *problems* 1, 2, 3, 4 are quite useful in establishment of bounds on effective properties.

3
Nonlinear elasticity

Following [9], consider a nonlinear elastic constitutive law of the form

$$\boldsymbol{\sigma} = \boldsymbol{\phi}(\boldsymbol{\varepsilon}) \ . \tag{40}$$

The free Helmholtz energy respectively is

$$F = \int \boldsymbol{\sigma} : \mathrm{d}\boldsymbol{\varepsilon} \ . \tag{41}$$

Thus, the Hill condition in this case will take the form

$$\left\langle \int \boldsymbol{\sigma} : \mathrm{d}\boldsymbol{\varepsilon} \right\rangle = \int \langle \boldsymbol{\sigma} \rangle : \mathrm{d}\langle \boldsymbol{\varepsilon} \rangle \ . \tag{42}$$

Let us illustrate the last result. Following [9] consider a constitutive law of the form

$$\boldsymbol{\sigma} = \sum_{n=1}^{N} \mathbf{C} \ \boldsymbol{\varepsilon}^{n} \ , \tag{43}$$

Equation (43) is another form of nonlinear law (36) used above for the boundary data analysis. The corresponding free energy expression will be

$$F = \sum_{n=1}^{N} \mathbf{C}_n \frac{\boldsymbol{\varepsilon}^{n+1}}{n+1} \ . \tag{44}$$

In (44), tensors $\mathbf{C}_k$ represent the k-th order elastic constants.

Then, the energetic $\mathbf{C}_n^e$ and the mechanical $\mathbf{C}_n^m$ definitions of the effective properties will be, respectively,

$$\langle F \rangle = \sum_{n=1}^{N} \mathbf{C}_n^e \frac{\langle \boldsymbol{\varepsilon} \rangle^{n+1}}{n+1} \ , \tag{45}$$

$$\langle \boldsymbol{\sigma} \rangle = \sum_{n=1}^{N} \mathbf{C}_n^m \langle \boldsymbol{\varepsilon} \rangle^{n} \ . \tag{46}$$

The LHS of condition (42) is, in fact, identical to (45). The RHS of (42) will be

$$\int \langle \boldsymbol{\sigma} \rangle : \mathrm{d}\langle \boldsymbol{\varepsilon} \rangle = \sum_{n=1}^{N} \mathbf{C}_n^m \frac{\langle \boldsymbol{\varepsilon} \rangle^{n+1}}{n+1} \ . \tag{47}$$

Thus, the hypothesis of compatibility of energetic and mechanical effective properties leads to condition (42) that, in its turn, is a consequence of (8). Finally, one writes

$$\sum_{n=1}^{N} \mathbf{C}_n^e \frac{\langle \boldsymbol{\varepsilon} \rangle^{n+1}}{n+1} = \sum_{n=1}^{N} \mathbf{C}_n^m \frac{\langle \boldsymbol{\varepsilon} \rangle^{n+1}}{n+1} \ . \tag{48}$$

The uniform boundary conditions that guarantee the validity of the Hill condition in the nonlinear elastic case, are those defined and discussed above in Sec. 2.

4 Viscoelasticity

Consider classic constitutive equations of viscoelasticity, the Volterra-Frechet expansion, see e.g. [1]

$$\boldsymbol{\sigma} = \int_{-\infty}^{t} \mathbf{r}_1(t-u_1)\mathrm{d}\boldsymbol{\varepsilon}(u_1) + \int_{-\infty}^{t}\int_{-\infty}^{t} \mathbf{r}_2(t-u_1, t-u_2)\mathrm{d}\boldsymbol{\varepsilon}(u_2)\mathrm{d}\boldsymbol{\varepsilon}(u_1) + \cdots \ . \tag{49}$$

The point now is to define the free energy functional. But the only objective fact known about the free Helmholtz energy functional in viscoelasticity, is that for the case when the instantaneous modulus is not equal to zero, free energy is a stress potential in an isothermal deformation, see e.g. [14], [1].

$$\boldsymbol{\sigma} = \frac{\partial F}{\partial \boldsymbol{\varepsilon}} \quad . \tag{50}$$

Symbol $\partial F/\partial \varepsilon$ in (50) means a partial Frechet derivative, where only actual values of the deformation tensor are counted, and not its history.

The problem of the free energy viscoelastic functional remains still open.

a) Consider at first the linear case, where we have at least some well-known phenomenological free energy expressions.

Consider a linear viscoelastic constitutive law

$$\boldsymbol{\varepsilon} = \mathbf{f} \circ \boldsymbol{\sigma}, \ \ \boldsymbol{\sigma} = \mathbf{r} \circ \boldsymbol{\varepsilon} \ , \tag{51}$$

with $\boldsymbol{\sigma}$ and $\boldsymbol{\varepsilon}$ respectively the stress and the deformation tensors $\mathbf{f}(t)$ and $\mathbf{r}(t)$ the creep and the relaxation functions tensors and o the Stieltjes convolution operator.

Then, the mentioned energy expression, obtained by Staverman & Schwarzl for polymers on the base of the statistical physics concept, has the form, see e.g. [1]

$$F = \frac{1}{2} \int_{0^-}^{t} \int_{0^-}^{t} \mathbf{r}(2t - u - v) \mathrm{d}\boldsymbol{\varepsilon}(v) \mathrm{d}\boldsymbol{\varepsilon}(u) \quad . \tag{52}$$

On the other hand, after algebraic transforms one can present (52) as

$$F = \frac{1}{2} \left(\int_{0^-}^{t} - \int_{t}^{2t} \right) \boldsymbol{\sigma}(2t - u) \mathrm{d}\boldsymbol{\varepsilon}(u) \quad . \tag{53}$$

In an operator form, this can be rewritten as

$$F = 0.5 \boldsymbol{\sigma} \diamondsuit \boldsymbol{\varepsilon} \quad . \tag{54}$$

In [7] the operator $\diamondsuit$ has been named a pseudo-convolution operator.

One can define mechanical and energetical effective properties through, respectively

$$\langle F \rangle = \frac{1}{2} \int_{0^-}^{t} \int_{0^-}^{t} \mathbf{r}_e^{\mathrm{eff}}(2t - u - v) \mathrm{d}\langle \boldsymbol{\varepsilon}(v) \rangle \mathrm{d}\langle \boldsymbol{\varepsilon}(u) \rangle \quad , \tag{55}$$

and

$$\langle \boldsymbol{\sigma} \rangle = \int_{0^-}^{t} \mathbf{r}_m^{\mathrm{eff}}(t - u) \mathrm{d}\langle \boldsymbol{\varepsilon}(u) \rangle \quad . \tag{56}$$

In (55) and (56), $\mathbf{r}_e^{\mathrm{eff}}$ and $\mathbf{r}_m^{\mathrm{eff}}$ are, correspondingly, energetic and mechanical effective relaxation function tensors. Then one obtains through (51)–(56)

$$\langle F \rangle = \frac{1}{2} \langle \boldsymbol{\sigma} \rangle \diamondsuit \langle \boldsymbol{\varepsilon} \rangle = (\mathbf{r}_e^{\mathrm{eff}} \circ \langle \boldsymbol{\varepsilon} \rangle) \diamondsuit \langle \boldsymbol{\varepsilon} \rangle = \frac{1}{2} (\mathbf{r}_e^{\mathrm{eff}} \circ \mathbf{f}_n^{\mathrm{eff}} \circ \langle \boldsymbol{\sigma} \rangle) \diamondsuit \langle \boldsymbol{\varepsilon} \rangle \quad . \tag{57}$$

Thus, the condition of equivalence of $\mathbf{r}_e^{\mathrm{eff}}$ and $\mathbf{r}_m^{\mathrm{eff}}$ leads finally to

$$\langle \boldsymbol{\sigma} \diamondsuit \boldsymbol{\varepsilon} \rangle = \langle \boldsymbol{\sigma} \rangle \diamondsuit \langle \boldsymbol{\varepsilon} \rangle \quad . \tag{58}$$

The latter is nothing but a particular case of relationship (8).

b) What concerns nonlinear viscoelasticity, the principal point is to define correctly the free energy functional, that remains an Achilles' heel of viscoelasticity.

That is why let us limit ourselves for the moment to phenomenological models, among which the most well-known are the Leaderman-Rabotnov and Rozovsky models. For the sake of clarity we take them in uniaxial form. The first one, see e.g. [11] and [15], has the constitutive equation

$$\varphi(\varepsilon) = \frac{\sigma}{r_0} + \int_{-\infty}^{t} \Phi(t-u)\sigma(u)\mathrm{d}u \ , \tag{59}$$

where $\varphi(\varepsilon)$ is the so called instantaneous curve, a material characteristic, r_0 denotes the instant modulus and $\Phi(t)$ - the kernel function. An alternative is the Rozovsky model, see [15]

$$\varepsilon = \frac{\sigma}{r_0} + \int_{-\infty}^{t} \Phi(t-u)\Psi[\sigma(u)]\mathrm{d}u \ , \tag{60}$$

with another material characteristic function $\psi(\sigma)$. Let us illustrate the generalized Hill principle on the example of the first model that can be formulated also as

$$\sigma = r \circ \varphi(\varepsilon) = \int_{0^-}^{t} \varphi(\varepsilon)\mathrm{d}r(t-u) \ , \tag{61}$$

where $r(t)$ is a certain material function obtained from (59).

Then, based on the concept (50), one could define the free energy functional as

$$F = \int_{0^-}^{t} \left[\int \varphi(\varepsilon)\mathrm{d}\varepsilon \right] \mathrm{d}r(t-u) = \sigma * \varepsilon \ . \tag{62}$$

Operator $*$ in (62) is defined through

$$\sigma * \varepsilon \equiv \int \sigma \mathrm{d}\varepsilon \ , \tag{63}$$

Then, mechanical and energetical effective properties can be respectively defined through

$$\langle F \rangle = \int_{0}^{t} \left(\int \varphi(\langle \varepsilon \rangle)\mathrm{d}\langle \varepsilon \rangle \right) \mathrm{d}r_e^{\mathrm{eff}}(t-u) \ , \tag{64}$$

$$\langle \sigma \rangle = \int_{0}^{t} \varphi(\langle \varepsilon \rangle)\mathrm{d}r_m^{\mathrm{eff}}(t-u) \ . \tag{65}$$

As a result, through the procedure (61)–(65), the condition of equivalence of r_e^{eff} and r_m^{eff} leads again to the Hill principle (8) with operator $*$ defined through (63).

Conclusion

The Hill condition has been discussed and generalized for nonlinear inelastic materials. The importance of the Hill principle in micromechanics bases on the following points:

1) It represents one of the basical principles in homogeneization of statistically homogeneous and ergodic random and periodic materials. Together with the concepts of statistical uniformity and representative volume, the Hill principle can be considered as an essential ingredient of modern micromechanics.
2) The Hill principle represents the condition for equivalence (compatibility) of the mechanically and energetically defined effective properties.
3) The Hill principle gives as particular solutions the cases of uniform static, kinematic and mixed boundary data, fundamental in micromechanics, especially for the size effect study.

The first point was discussed in detail in [16], while the present paper concentrates mostly on points 2 and 3. Concerning point 3, we have shown that the Hill relationship gives the uniform mixed boundary conditions MUBC as a particular class of solutions.

The definition and the main properties of this important loading case have been discussed, and conditions of its realization in material testing established. It has been shown that the tested material must be at least orthotropic, and that the mixed uniform boundary data can be of diagonal structure, excluding thus shear strains/stresses.

As mixed boundary conditions are fundamental in experimental testing of engineering materials, the obtained results attest the significance of the Hill condition for applications.

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