SECTION 10.3

Nonlinear Composites: Secant Methods and Variational Bounds

PIERRE M. SUQUET
LMA/CNRS 31 chemin Joseph Aiguier, 13402, Marseille, Cedex 20, France

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10.3.1 INTRODUCTION

The problem addressed here is that of the effective behavior of *nonlinear composites*. By composites we understand not only man-made materials but also all types of inhomogeneous materials which are ubiquitous in nature (polycrystals, wood, rocks, bone, porous materials).

Consider a finite volume element V of such a composite material, large enough to be representative of the composite microstructure and nevertheless small enough for the different phases (grains or different mechanical phases) to be clearly distinguished. This representative volume element (RVE) V is composed of N distinct homogeneous phases V_r, $r = 1, \ldots, N$, the behavior of which is characterized by a nonlinear relation between the (infinitesimal) strain and stress fields, $\boldsymbol{\varepsilon}$ and $\boldsymbol{\sigma}$,

$$\boldsymbol{\varepsilon}(x) = \mathscr{G}^{(r)}(\boldsymbol{\sigma}(x)) \quad \text{when } \boldsymbol{x} \text{ is in phase } r \tag{1}$$

This constitutive relation corresponds either to *nonlinear elastic behavior* within the context of small strains, or to *finite viscous deformations* when $\boldsymbol{\varepsilon}$ and $\boldsymbol{\sigma}$ are interpreted as the Eulerian strain rate and Cauchy stress, respectively.

When the RVE V is subjected to an average strain $\bar{\boldsymbol{\varepsilon}}$, it reacts to this strain by an average stress $\bar{\boldsymbol{\sigma}}$. The relation between $\bar{\boldsymbol{\sigma}}$ and $\bar{\boldsymbol{\varepsilon}}$ is the effective constitutive relation of the composite.

The question addressed here is: *Can predictions be made regarding this effective constitutive relation, given the constitutive relations of the phases and some (often limited) information about the composite microstructure?*

When the phases are linear, there exists a large body of literature partially answering this question. However, when the phases are nonlinear, there are very few *really* nonlinear schemes to analyze the global, as well as the local, response of nonlinear composites. Most methods are heuristic and are extensions or modifications of the secant method (which we will discuss in Section 10.3.2) and of the incremental method (which we will not discuss here). Most of these schemes proceed in three successive steps:

1. First, the constitutive relations of each individual phase are linearized in an appropriate manner. This is done pointwisely and serves to define a *linear comparison solid* with local elastic moduli which, in general, vary from point to point.
2. Then, the problem is reduced to that of estimating the effective properties of a *linear comparison solid* with a *finite* number of phases. To this aim, an approximation is introduced by assuming that the local moduli are piecewise uniform. In most cases (but not all), the regions

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where the moduli are uniform are precisely the domains occupied by the material phases.

3. Finally, the effective linear properties of the linear comparison solid are estimated or bounded by a scheme which is relevant for the type of microstructure exhibited by the linear comparison solid. These linear effective properties are used to estimate the nonlinear effective properties of the actual nonlinear composite.

Following the seminal work of J. Willis [1], more rigorous results, namely, bounds for the nonlinear effective properties of composites, have been developed in the past ten years by Ponte Castañeda [2], Willis [3], and Suquet [4], among others. Ponte Castañeda's variational procedure [5], which will be briefly recalled in Section 10.3.3, is probably the most rigorous bounding theory available to date. Other methods, less rigorous but sometimes more accurate — for instance, the second-order procedure of Ponte Castañeda [6] or the affine procedure of Masson and Zaoui [7] — will not be discussed here. Some connections do exist between the heuristic secant methods and the more elaborate bounding techniques, and we will briefly outline them in Section 10.3.3.3, following Suquet [8, 9]. More details can be found in the review papers by Ponte Castañeda and Suquet [10] and Willis [11] (see also several contributions in Reference [12]).

10.3.2 SECANT METHODS

10.3.2.1 NONLINEAR LOCAL PROBLEM

The local stress and strain fields within V are solutions of the *local problem* consisting of the constitutive equations (Eq. 1), the compatibility conditions satisfied by $\boldsymbol{\varepsilon}$, and the equilibrium equations satisfied by $\boldsymbol{\sigma}$:

$$\boldsymbol{\varepsilon}(\boldsymbol{x}) = \mathscr{G}^{(r)}(\boldsymbol{\sigma}(\boldsymbol{x})) \text{ in phase } r, \quad \boldsymbol{\varepsilon} = \tfrac{1}{2}\left(\nabla \boldsymbol{u} + \nabla \boldsymbol{u}^t\right), \quad \mathrm{div}(\boldsymbol{\sigma}) = 0 \quad \text{in } V \qquad (2)$$

Several classes of boundary conditions can be considered on ∂V (all conditions being equivalent in the limit of a large RVE under appropriate growth conditions on the functions $\mathscr{G}^{(r)}$). For definiteness, we will assume uniform tractions on the boundary $\boldsymbol{\sigma}(\boldsymbol{x}).\boldsymbol{n}(\boldsymbol{x}) = \bar{\boldsymbol{\sigma}}.\boldsymbol{n}(\boldsymbol{x})$ on ∂V. Once Eq. 2 is solved (at least theoretically), the spatial average $\bar{\boldsymbol{\varepsilon}}$ of the strain field $\boldsymbol{\varepsilon}$ can be taken:

$$\bar{\boldsymbol{\varepsilon}} = \langle \boldsymbol{\varepsilon} \rangle, \quad \text{where} \quad \langle . \rangle = \frac{1}{|V|}\int_V . \, dx$$

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Then the relation between this average strain and the imposed average stress defines the *effective constitutive relation* of the composite:

$$\bar{\boldsymbol{\varepsilon}} = \tilde{\mathscr{G}}(\bar{\boldsymbol{\sigma}}) \tag{3}$$

10.3.2.2 LINEARIZATION

It is in general impossible to solve the nonlinear local problem (Eq. 2) exactly. Therefore, a first step in most nonlinear schemes is to write the constitutive relations (Eq. 1) in the form (the reference to the phase is implicit here):

$$\boldsymbol{\varepsilon}(\boldsymbol{x}) = \boldsymbol{M}(\boldsymbol{\sigma}(\boldsymbol{x})) : \boldsymbol{\sigma}(\boldsymbol{x}) + \boldsymbol{\eta}(\boldsymbol{x}) \tag{4}$$

where $\boldsymbol{M}$ and $\boldsymbol{\eta}$ have to be specified. Two particular choices, giving rise to two broad classes of models referred to as *secant* and *tangent* models and schematically depicted in Figure 10.3.1 (several different choices are reviewed in Gilormini [13]), correspond respectively to the following linearlizations:

$$secant : \boldsymbol{M}(\boldsymbol{\sigma}(\boldsymbol{x})) = \boldsymbol{M}_{sct}(\boldsymbol{\sigma}(\boldsymbol{x})), \quad \boldsymbol{\eta} = 0 \tag{5}$$

and

$$tangent : \boldsymbol{M}(\boldsymbol{\sigma}(\boldsymbol{x})) = \boldsymbol{M}_{tgt}(\boldsymbol{\sigma}(\boldsymbol{x})), \quad \boldsymbol{\eta} = \boldsymbol{e}_0 \tag{6}$$

Note that there are several choices for $\boldsymbol{M}_{sct}$ in (Eq. 5) for which $\boldsymbol{M}_{sct}(\boldsymbol{\sigma}) : \boldsymbol{\sigma} = \mathscr{G}(\boldsymbol{\sigma})$. Consequently, the secant moduli are not uniquely defined

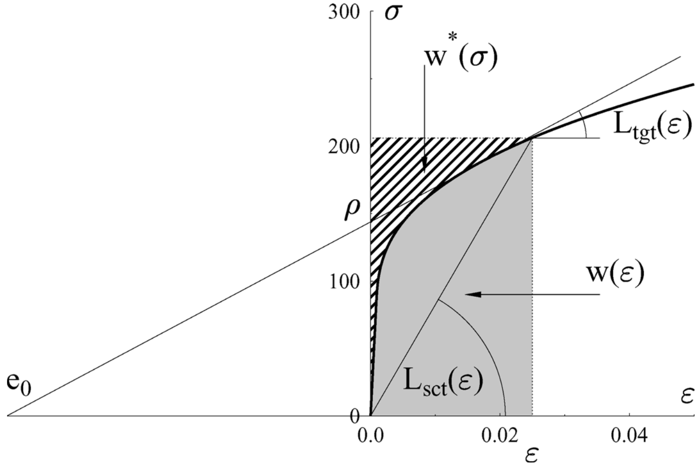

FIGURE 10.3.1 Secant and tangent moduli in a tensile uniaxial test. The secant and the tangent compliance are the inverse of the secant and tangent stiffness $\boldsymbol{L}_{sct}$ and $\boldsymbol{L}_{tgt}$, respectively.

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(this observation was made by Gilormini [13]). The tangent moduli are defined (uniquely in general) as $\boldsymbol{M}_{tgt}(\boldsymbol{\sigma}) = d\mathcal{G}(\boldsymbol{\sigma})/d\boldsymbol{\sigma}$.

We will not discuss the second choice (Eq. 6), namely, the class of tangent methods (the interested reader is referred to References [6, 13, 14] for additional details).

10.3.2.3 Secant Methods in General

Using the equivalent writing (Eq. 5) of the constitutive relations (Eq. 1), the local problem (Eq. 2) can be reformulated as

$$\boldsymbol{\varepsilon}(\boldsymbol{x}) = \boldsymbol{M}^{(r)}_{sct}(\boldsymbol{\sigma}(\boldsymbol{x})) : \boldsymbol{\sigma}(\boldsymbol{x}) \text{ in phase } r, \quad \boldsymbol{\varepsilon} = \tfrac{1}{2}\left(\nabla \mathbf{u} + \nabla \mathbf{u}^t\right), \quad \operatorname{div}(\boldsymbol{\sigma}) = 0 \text{ in } V \tag{7}$$

This problem can be considered a linear elasticity problem for a composite with infinitely many phases, since the moduli $\boldsymbol{M}^{(r)}_{sct}(\boldsymbol{\sigma}(\boldsymbol{x}))$ may vary from point to point. Equation 7 is therefore not simpler than the original (Eq. 2). A simplifying assumption is introduced by assuming that the secant moduli take piecewise uniform values $\boldsymbol{M}^{(r)}$ on subdomains $W_r, r = 1, \ldots, M$, which coincide with, or are contained in, the physical domains V_r. For simplicity we will assume in the following that the domains W_r and V_r coincide.

In addition, the uniform moduli are assumed to be evaluated at some "effective stress" $\tilde{\boldsymbol{\sigma}}^{(r)}$ for the phase r:

$$\boldsymbol{M}^{(r)} = \boldsymbol{M}^{(r)}_{sct}\left(\tilde{\boldsymbol{\sigma}}^{(r)}\right) \tag{8}$$

The simplified secant problem now consists of the following:

$$\boldsymbol{\varepsilon}(\boldsymbol{x}) = \mathbf{M}^{(r)} : \boldsymbol{\sigma}(\boldsymbol{x}) \text{ in phase } r, \quad \boldsymbol{\varepsilon} = \tfrac{1}{2}\left(\nabla \mathbf{u} + \nabla \mathbf{u}^t\right), \quad \operatorname{div}(\boldsymbol{\sigma}) = 0 \quad \text{in } V \tag{9}$$

Assuming for a moment that the $\boldsymbol{M}^{(r)}$'s are given, Eq. 9 is a problem for a linear N-phase composite called the *linear comparison composite*. The determination of the $\boldsymbol{M}^{(r)}$'s is made possible through Eq. 8. These relations involve the effective stress $\tilde{\boldsymbol{\sigma}}^{(r)}$ (which has not been specified yet, but this will be done in the next two paragraphs), which itself depends on the average stress $\bar{\boldsymbol{\sigma}}$. Therefore, Eq. 8 is a closure condition which renders the problem nonlinear.

In summary, any secant method involves three steps:

1. A linear theory providing an expression for $\tilde{\boldsymbol{M}}$ as a function of the moduli $\boldsymbol{M}^{(r)}$ of the individual phases in the linear comparison solid.

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2. The resolution of N nonlinear tensorial problems for the N unknown tensors $\boldsymbol{M}^{(r)}$:

$$\boldsymbol{M}^{(r)} = \boldsymbol{M}^{(r)}_{sct}\left(\tilde{\boldsymbol{\sigma}}^{(r)}\right), \quad \tilde{\boldsymbol{\sigma}}^{(r)} = \text{function of } \left\{\boldsymbol{M}^{(r)}\right\}_{r=1,\ldots,N} \text{ and } \bar{\boldsymbol{\sigma}} \tag{10}$$

   Note that these equations depend on the particular expression ("function of") chosen for the effective stress $\tilde{\boldsymbol{\sigma}}^{(r)}$. Different choices give rise to different secant methods.
3. Once the N nonlinear tensorial problems (Eq. 10) are solved, the overall stress-strain relation is given by

$$\bar{\boldsymbol{\varepsilon}} = \tilde{\boldsymbol{M}} : \bar{\boldsymbol{\sigma}} \tag{11}$$

   where $\tilde{\boldsymbol{M}}$ is the effective compliance derived by means of the linear theory of step 1 from the individual moduli $\boldsymbol{M}^{(r)}$ as determined in step 2.

10.3.2.4 A Secant Method Based on First-Order Moments (Classical Secant Method)

It remains to define the effective stress $\tilde{\boldsymbol{\sigma}}^{(r)}$. In the classical secant method, this effective stress is set equal to the average stress over phase r (it should be emphasized that the stress field under consideration is now that in the linear comparison solid):

$$\tilde{\boldsymbol{\sigma}}^{(r)} = \bar{\boldsymbol{\sigma}}^{(r)} = \langle \boldsymbol{\sigma} \rangle_r, \quad \text{where} \quad \langle . \rangle_r = \frac{1}{|V_r|}\int_{V_r} . \, d\boldsymbol{x} \tag{12}$$

This "first order moment" of the stress over phase r can be expressed in terms of the overall stress $\bar{\boldsymbol{\sigma}}$ by means of the "stress-localization" tensor $\boldsymbol{B}^{(r)}$:

$$\bar{\boldsymbol{\sigma}}^{(r)} = \langle \boldsymbol{B} \rangle_r : \bar{\boldsymbol{\sigma}} \tag{13}$$

Most linear theories provide (more or less) explicit expressions for the "stress-localization" tensors $\boldsymbol{B}^{(r)}$ as functions of the individual compliances $\boldsymbol{M}^{(r)}$. A typical example will be given in following text. Equation 13 completes Eq. 10.

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10.3.2.5 A Secant Method Based on Second-Order Moments (Modified Secant Method)

The classical secant method has several serious limitations. One of them is illustrated by its unphysical prediction for the response of nonlinear porous materials under hydrostatic loadings. Consider an RVE composed of an incompressible matrix with voids and subject to an hydrostatic stress. The average stress in the matrix is hydrostatic (the average stress in the voids is 0). Since the matrix is incompressible, it is insensitive to hydrostatic stresses. Therefore, the secant compliance associated with a purely hydrostatic stress by Eq. 12 always coincides with the initial compliance (under zero stress) of the material. The secant method applied with Eq. 12 predicts a linear overall response of the porous material. However, the actual response of the porous material is nonlinear, since the local stress state in the matrix is not hydrostatic (analytic calculations can be carried out on the hollow sphere model to prove this point explicitly), even if the average stress is hydrostatic. The occurrence of shear stresses in some regions of the RVE introduces nonlinearities both in the local and overall responses of the RVE which are not taken into account by Eq. 12.

This observation has motivated the introduction of theories based on *the second moment of the stress field*, in particular form by Buryachenko [15], in approximate form by Qiu and Weng [16], or in general and rigorous form by Suquet [8] and Hu [17]. It is indeed observed that in many cases of interest the secant compliance $\boldsymbol{M}_{sct}$ depends on the stress through the "quadratic stress"

$$\boldsymbol{M}_{sct}(\boldsymbol{\sigma}) = \boldsymbol{M}_{sct}(\mathcal{S}), \quad \text{where} \quad \mathcal{S} = \frac{1}{2}\,\boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \tag{14}$$

Therefore, rather than expressing $\boldsymbol{M}^{(r)}$ in terms of an "effective stress" $\tilde{\boldsymbol{\sigma}}^{(r)}$, one can express $\boldsymbol{M}^{(r)}$ in terms of an effective "quadratic stress" $\tilde{\mathcal{S}}^{(r)}$. A very natural choice for this effective quadratic stress is

$$\tilde{\mathcal{S}}^{(r)} = \left\langle \tilde{\mathcal{S}} \right\rangle_r = \frac{1}{2}\langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle_r \tag{15}$$

This effective "second-order moment" of the stress over phase r has definite advantages over the first-order moment used in the classical secant method. For instance, it better accounts for local fluctuation of the stress. To see this, note that $\tilde{\mathcal{S}}^{(r)}_{ijij} = \left\langle \sigma_{ij}\sigma_{ij} \right\rangle_r$. Therefore, as soon as $\boldsymbol{\sigma}$ is nonzero in a (non-negligible) region of phase r, the second-moment $\tilde{\mathcal{S}}^{(r)}$ of the stress does not vanish. In particular, the overall response of porous materials under hydrostatic loading, as predicted by the secant method based on the "second-order moment," is nonlinear (as it should be) and close to the exact solution [16]. The modified secant theory consists in solving Eq. 10 together with the definition (Eq. 15) of the effective stress of phase r.

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In practice, one has to compute $\tilde{\mathscr{S}}^{(r)}$ for the linear comparison solid. This can be done analytically by means of a result previously used in different contexts by several authors (see, for instance, Kreher [18]).

Consider a linear composite composed of N homogeneous phases with elastic compliance $\mathbf{M}^{(r)}$. *Let* $\tilde{\mathbf{M}}(\mathbf{M}^{(1)}, \ldots, \mathbf{M}^{(r)}, \ldots, \mathbf{M}^{(N)})$ *be the overall compliance tensor of this composite, and let* $\boldsymbol{\sigma}$ *denote the stress field in this linear composite. Then:*

$$\langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle_r = \frac{1}{c^{(r)}} \bar{\boldsymbol{\sigma}} : \frac{\partial \tilde{\mathbf{M}}}{\partial \mathbf{M}^{(r)}} : \bar{\boldsymbol{\sigma}} \tag{16}$$

A detailed proof of this result can be found in References [9, 18] (among others).

In conclusion, the nonlinear systems of equations to be solved to complete step 2 of the secant method read as:

$$\tilde{\mathscr{S}}^{(r)} = \frac{1}{2c^{(r)}} \bar{\boldsymbol{\sigma}} : \frac{\partial \tilde{\mathbf{M}}}{\partial \mathbf{M}^{(r)}} : \bar{\sigma}, \quad \mathbf{M}^{(r)} = \mathbf{M}_{sct}^{(r)}\left(\tilde{\mathscr{S}}^{(r)}\right) \tag{17}$$

10.3.2.6 Example: Deformation Theory of Plasticity

A rather general form of stress-strain relations for isotropic elastic-plastic materials in the context of a deformation theory is given by the following:

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}^e + \boldsymbol{\varepsilon}^p, \quad \boldsymbol{\varepsilon}^e = \frac{\boldsymbol{\sigma}_m}{k} \, i + \frac{s}{2\mu_0}, \quad \boldsymbol{\varepsilon}^p = \frac{3}{2} \frac{p(\sigma_{eq})}{\sigma_{eq}} s \tag{18}$$

where $\sigma_m = (1/3) \operatorname{tr}(\boldsymbol{\sigma})$ is the hydrostatic stress, $\sigma_{eq} = [(3/2)\boldsymbol{s} : \boldsymbol{s}]^{1/2}$ is the von Mises equivalent stress ($\boldsymbol{s}$ being the stress deviator), and $p(\sigma_{eq})$ is the inverse of the plastic hardening curve $\sigma_{eq}(p)$ of the material. An alternate writing of Eq. 18 is

$$\boldsymbol{\varepsilon} = \boldsymbol{M}_{sct}(\boldsymbol{\sigma}) : \boldsymbol{\sigma}, \quad \text{with } \boldsymbol{M}_{sct}(\boldsymbol{\sigma}) = \frac{m}{3} \boldsymbol{J} + \frac{\theta_{sct}(\sigma_{eq})}{2} \boldsymbol{K} \tag{19}$$

where

$$m = \frac{1}{k}, \quad \theta_{sct}(\sigma_{eq}) = \frac{1}{\mu_{sct}(\sigma_{eq})} = \frac{1}{\mu_0} + \frac{3p(\sigma_{eq})}{\sigma_{eq}} \tag{20}$$

and where $\boldsymbol{J}$ and $\boldsymbol{K}$ are the fourth-order tensors which project any second-order tensor on its hydrostatic and deviatoric parts:

$$\boldsymbol{J} = \tfrac{1}{3} \, i \otimes i, \quad \boldsymbol{K} = \boldsymbol{I} - \boldsymbol{J}$$

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i and $\boldsymbol{I}$ being the identity for symmetric second-order and fourth-order tensors, respectively.

Since we will be manipulating isotropic tensors throughout this section, simplified (and classical) notations will be helpful. Any isotropic fourth-order tensor $\boldsymbol{B}$ with minor and major symmetries can be decomposed as $\boldsymbol{B} = b_m \boldsymbol{J} + b_{dev}\boldsymbol{K}$, where $b_m = \boldsymbol{B} :: \boldsymbol{J}$ and $b_{dev} = (1/5)\boldsymbol{B} :: \boldsymbol{K}$. We use the compact notation $\boldsymbol{B} = \{b_m, b_{dev}\}$. The algebra over isotropic (and symmetric) tensors is then very simple: for two such tensors $\boldsymbol{B}$ and $\boldsymbol{C}$, one has $\boldsymbol{B} :: \boldsymbol{C} = \{b_m c_m, b_{dev} c_{dev}\}$ and $(\boldsymbol{B})^{-1} = \{(1/b_m), (1/b_{dev})\}$.

The response of the phases as described by Eq. 19 is linear for purely hydrostatic loadings (characterized by a constant bulk modulus k), and nonlinear in shear (characterized by a stress-dependent secant shear modulus μ_{sct}). Note that $\boldsymbol{M}_{sct}$, which depends on $\boldsymbol{\sigma}$ through σ_{eq} only, is indeed a function of the quadratic stress $\mathscr{S}$ defined by Eq. 14. Indeed, σ_{eq} is itself a function of $\mathscr{S}$: $\sigma_{eq} = (\frac{3}{2}\, \boldsymbol{s} : \boldsymbol{s})^{1/2} = (3\boldsymbol{K} :: \mathscr{S})^{1/2}$.

Consider now a two-phase material, where both phases are elastic-plastic and obey the constitutive relations (Eq. 19) with different material constants. We further consider the case where the phases in the composite are arranged isotropically in a particle-matrix configuration, corresponding, for instance, to the case of spherical particles of phase 1 randomly distributed in phase 2 (which is the matrix). The linear comparison composite is a two-phase linear composite with the same microstructure as the initial nonlinear composite and with isotropic phases characterized by a compliance tensor $\boldsymbol{M}^{(r)} = \{m^{(r)}/3, \theta^{(r)}/2\}$.

Regarding the first step of the three-step procedure outlined previously, namely, a theory for the effective properties of the linear comparison solid, the Hashin-Shtrikman formalism is known to provide, in most cases, an accurate estimate of the effective properties of isotropic linear composites with particle-matrix microstructure. The corresponding estimate reads as

$$\tilde{\boldsymbol{M}} = \boldsymbol{M}^{(2)} + c^{(1)}\left(\boldsymbol{M}^{(1)} - \boldsymbol{M}^{(2)}\right) : \boldsymbol{B}^{(1)},\ \boldsymbol{B}^{(1)} = \left(\boldsymbol{I} + c^{(2)}\boldsymbol{Q} : \left(\boldsymbol{M}^{(1)} - \boldsymbol{M}^{(2)}\right)\right)^{-1} \tag{21}$$

where $c^{(1)}$ and $c^{(2)}$ are the phase volume fractions and where $\boldsymbol{Q} = \{3q_m, 2q_{dev}\}$ with

$$q_m = \frac{1-\alpha}{m^{(2)}}, \quad q_{dev} = \frac{1-\beta}{\theta^{(2)}}, \quad \alpha = \frac{3k^{(2)}}{3k^{(2)} + 4\mu^{(2)}}, \quad \beta = \frac{2}{5}\,\frac{3k^{(2)} + 6\mu^{(2)}}{3k^{(2)} + \left(4/3\right)\mu^{(2)}}$$

Then, the effective compliance $\tilde{\boldsymbol{M}} = \{\tilde{m}/3, \tilde{\theta}/2\}$ and the stress-localization tensors $\boldsymbol{B}^{(r)} = \{b_m^{(r)}, b_{dev}^{(r)}\}$ read, respectively:

$$\tilde{m} = m^{(2)} + c^{(1)}\left(m^{(1)} - m^{(2)}\right)b_m^{(1)}, \quad \tilde{\theta} = \theta^{(2)} + c^{(1)}\left(\theta^{(1)} - \theta^{(2)}\right)b_{dev}^{(1)},$$

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$$b_m^{(1)} = \frac{1}{1 + c^{(2)} q_m (m^{(1)} - m^{(2)})}, \quad b_m^{(1)} = \frac{1}{1 + c^{(2)} q_{dev} \left(\theta^{(1)} - \theta^{(2)}\right)},$$

$$b_m^{(2)} = \frac{1}{c^{(2)}}\left(1 - c^{(1)} b_m^{(1)}\right), \quad b_{dev}^{(2)} = \frac{1}{c^{(2)}}\left(1 - c^{(1)} b_{dev}^{(1)}\right).$$

Regarding step 2 of the procedure, namely, the non-linear equation which stems from the choice of the "effect stress," the classical method makes use of the average stresses $\bar{\boldsymbol{\sigma}}^{(r)} = \boldsymbol{B}^{(r)} : \bar{\boldsymbol{\sigma}}$. However, since $\boldsymbol{M}_{sct}(\boldsymbol{\sigma})$ depends on $\boldsymbol{\sigma}$ only through the von Mises stress, the only useful information in the average stress is the von Mises equivalent stress $\bar{\sigma}_{eq}^{(r)} = b_{dev}^{(r)} \bar{\sigma}_{eq}$. Therefore, the nonlinear equations to be solved in the classical secant method read

$$\theta^{(r)} = \theta_{sct}^{(r)}\left(\bar{\sigma}_{eq}^{(r)}\right) = \frac{1}{\mu_0^{(r)}} + \frac{3p^{(r)}\left(\bar{\sigma}_{eq}^{(r)}\right)}{\bar{\sigma}_{eq}^{(r)}}, \quad \bar{\sigma}_{eq}^{(r)} = b_{dev}^{(r)} \bar{\sigma}_{eq} \tag{22}$$

where the coefficients $b_{dev}^{(r)}$ depend on the $\theta^{(r)}$ (this is where the nonlinearity comes into play).

As for the secant method based on the "second-order moment," using again the fact that the secant compliance depends on the stress only through the von Mises stress, it is sufficient to compute the following quantities $\bar{\bar{\sigma}}_r = \langle \sigma_{eq}^2 \rangle_r^{1/2}$. These quantities can be calculated by means of Eq. 16. The resulting nonlinear systems of equations finally read (see Suquet [9] for more details):

$$\left.\begin{aligned}
&\theta^{(r)} = \theta_{sct}^{(r)}\left(\bar{\bar{\sigma}}_{eq}^{(r)}\right) = \frac{1}{\mu_0^{(r)}} + \frac{3p^{(r)}\left(\bar{\bar{\sigma}}_{eq}^{(r)}\right)}{\bar{\bar{\sigma}}_{eq}^{(r)}}, \quad \bar{\bar{\sigma}}_{eq}^{(r)} = b_{dev}^{(1)} \bar{\sigma}_{eq},\\
&\bar{\bar{\sigma}}_{eq}^{(2)} = \left(a\bar{\sigma}_m^2 + b\bar{\sigma}_{dev}^2\right)^{1/2}\\
&a = \frac{3}{c^{(2)}\theta^{(2)}}\left(\tilde{m} - c^{(1)} m^{(1)} b_m^{(1)^2} - c^{(2)} m^{(2)} b_m^{(2)^2}\right)\\
&b = \frac{1}{c^{(2)}\theta^{(2)}}\left(\tilde{\theta} - c^{(1)}\theta^{(1)} b_{dev}^{(1)^2} - \frac{12}{5} c^{(1)} c^{(2)} m^{(2)} b_{dev}^{(1)^2}\left(\frac{\theta^{(1)} - \theta^{(2)}}{3\theta^{(2)} + 4m^{(2)}}\right)^2\right)
\end{aligned}\right\} \tag{23}$$

Note that the effective stress in phase 2 (matrix) is now sensitive to the overall hydrostatic pressure (which is not the case in the classical secant method).

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10.3.3 VARIATIONAL BOUNDS

10.3.3.1 Effective Potentials

In this section it is further assumed that the constitutive behavior of the individual phases derives from a potential, or strain-energy function $w(\boldsymbol{\varepsilon})$, or equivalently a stress-energy function $w^*(\boldsymbol{\sigma})$, in such a way that the (infinitesimal) strain and stress fields, $\boldsymbol{\varepsilon}$ and $\boldsymbol{\sigma}$, are related by

$$\boldsymbol{\sigma} = \frac{\partial w}{\partial \boldsymbol{\varepsilon}}(\boldsymbol{\varepsilon}), \quad \text{or equivalently} \quad \boldsymbol{\varepsilon} = \frac{\partial w^*}{\partial \boldsymbol{\sigma}}(\boldsymbol{\sigma}),$$

$$\text{with } w(\boldsymbol{\varepsilon}) + w^*(\boldsymbol{\sigma}) = \boldsymbol{\sigma} : \boldsymbol{\varepsilon} \tag{24}$$

In the composite, the potentials w and w^* depend on the phase under consideration (and are denoted by $w^{(r)}$ and $(w^{(r)})^*$, respectively).

The solutions $\boldsymbol{u}$ and $\boldsymbol{\sigma}$ of the local problem (Eq. 2) have variational properties which are essential to deriving bounds on the effective energy of nonlinear composites. These properties, called *minimum energy principles*, also permit us to define the effective potentials of the composite. For simplicity, we shall only consider here the "minimum complementary-energy principle":

$$\tilde{w}^*(\bar{\boldsymbol{\sigma}}) = \inf_{\boldsymbol{\tau} \in \mathscr{K}(\bar{\boldsymbol{\sigma}})} \langle w^*(\boldsymbol{\tau}) \rangle, \quad \mathscr{K}(\bar{\boldsymbol{\sigma}}) = \{\boldsymbol{\tau},\ div(\boldsymbol{\tau}) = 0 \text{ in } V,\ \boldsymbol{\tau}.\boldsymbol{n} = \bar{\boldsymbol{\sigma}}.\boldsymbol{n} \ on\ \partial V\} \tag{25}$$

It can be checked [10] that the effective constitutive relations obtained by averaging the strain field solution of Eq. 2 or equivalently Eq. 25 derives from the potential $\tilde{w}^*$, which is therefore the effective complementary-energy of the composite:

$$\bar{\boldsymbol{\varepsilon}} = \frac{\partial \tilde{w}^*}{\partial \bar{\boldsymbol{\sigma}}}(\bar{\boldsymbol{\sigma}}).$$

10.3.3.2 Bounds

10.3.3.2.1 Ponte Castañeda's General Theory

To obtain bounds on the effective potential $\tilde{w}^*$ which are sharper than the Voigt and Reuss bounds, we introduce an inhomogeneous linear comparison composite with compliance $\boldsymbol{M}(\boldsymbol{x})$ and complementary-energy

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$w_0^*(\boldsymbol{x}, \boldsymbol{\tau}) = \frac{1}{2}\boldsymbol{\tau} : \boldsymbol{M}_0(\boldsymbol{x}) : \boldsymbol{\tau}$. Writing w^* as $w^* - w_0^* + w_0^*$, one obtains

$$\left.\begin{aligned}\tilde{w}^*(\bar{\boldsymbol{\sigma}}) &= \inf_{\boldsymbol{\tau}\in\mathcal{K}(\bar{\boldsymbol{\sigma}})} \langle w^*(\boldsymbol{\tau})\rangle = \inf_{\boldsymbol{\tau}\in\mathcal{K}(\bar{\boldsymbol{\sigma}})} \left(\left\langle \left(w^* - w_0^*\right)(\boldsymbol{\tau})\right\rangle + \left\langle w_0^*(\boldsymbol{\tau})\right\rangle\right)\\ &\geq \left(\inf_{\boldsymbol{\tau}\in\mathcal{K}(\bar{\boldsymbol{\sigma}})} \left\langle w_0^*(\boldsymbol{\tau})\right\rangle\right) - \langle v(\boldsymbol{x}, \boldsymbol{M}_0(\boldsymbol{x}))\rangle\\ &= \tilde{w}_0^*(\boldsymbol{M}_0, \bar{\boldsymbol{\sigma}}) - V(\boldsymbol{M}_0)\end{aligned}\right\} \tag{26}$$

where $\tilde{w}_0^*$ is the effective complementary energy of the linear comparison solid and

$$v(\boldsymbol{x}, \boldsymbol{M}_0(\boldsymbol{x})) = \sup_{\tau}\left[w_0^*(\boldsymbol{x}, \tau) - w^*(\boldsymbol{x}, \tau)\right], \quad V(\boldsymbol{M}_0) = \langle v(\boldsymbol{x}, \boldsymbol{M}_0(\boldsymbol{x}))\rangle \tag{27}$$

Since Eq. 26 is valid for any choice of $\boldsymbol{M}_0(\boldsymbol{x})$, one has

$$\tilde{w}^*(\bar{\boldsymbol{\sigma}}) \geq \sup_{\boldsymbol{M}_0(\boldsymbol{x})>0}\left[\tilde{w}_0^*(\boldsymbol{M}_0, \bar{\boldsymbol{\sigma}}) - V(\boldsymbol{M}_0)\right] \tag{28}$$

The inequality (Eq. 28), due to Ponte Castañeda, gives a rigorous bound[1] on the nonlinear effective properties of the composite (through the potential $\tilde{w}^*$) in terms of two functions:

- $\tilde{w}_0^*(\boldsymbol{M}_0, \bar{\boldsymbol{\sigma}}) = \frac{1}{2}\bar{\boldsymbol{\sigma}} : \tilde{\boldsymbol{M}}_0 : \bar{\boldsymbol{\sigma}}$ is the elastic energy of a *linear comparison composite* made up of phases with compliance $\boldsymbol{M}_0(\boldsymbol{x})$ at point $\boldsymbol{x}$; the linear comparison solid is chosen among all possible comparison composites by solving the optimization problem (Eq. 28). The difficulty lies in the precise determination of the energy $\tilde{w}_0^*$ for a linear comparison solid consisting of infinitely many different phases.
- The role of $v(\boldsymbol{x}, .)$ is to measure the difference between the nonquadratic potential $w^*(\boldsymbol{x}, .)$ and the quadratic energy $w_0^*(\boldsymbol{x}, .)$ of the linear comparison solid. This function is difficult to compute in general, but a bound can be easily computed for the class of materials considered in following text.

The problem of bounding the effective properties of a linear composite with infinitely many phases being too difficult, we reduce it by minimizing over a smaller set of compliance fields $\boldsymbol{M}_0(\boldsymbol{x})$, namely, those fields which are uniform on each subdomain V_r. With this smaller set, the supremum

[1] For a broad class of material behavior (discussed in Section 10.3.3.3), the inequality (Eq. 28) is in fact an equality and is strictly equivalent to the variational characterization of $\tilde{w}^*$ given in Eq. 25.

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in Eq. 28 even smaller:

$$\tilde{w}^*(\bar{\boldsymbol{\sigma}}) \geq \sup_{M_0^{(r)}>0} \left(\frac{1}{2} \bar{\boldsymbol{\sigma}} : \tilde{\boldsymbol{M}}_0 \left(\left\{ \boldsymbol{M}_0^{(r)} \right\} \right)_{r=1,\dots,N} : \bar{\boldsymbol{\sigma}} - V \left(\left\{ \boldsymbol{M}_0^{(r)} \right\} \right)_{r=1,\dots,N} \right) \tag{29}$$

The linear comparison composite is now an N-phase composite, with compliance $\boldsymbol{M}_0^{(r)}$ uniform throughout phase r. There is a similarity (and even more, as will be discussed in the next paragraph) with the secant methods in that a linear comparison solid is introduced in both approaches. Note, however, that here the elastic moduli in the linear composite are determined by means of an optimization procedure, whereas they were deduced from the (somehow arbitrary) choice of an effective stress in the case of the secant method. This optimization procedure leads to a rigorous bound for the potential $\tilde{w}^*$.

Remark: Most of the nonlinear bounds available to date are bounds on the energy of the composite. Only in specific situations do these bounds give bounds on the stress-strain relations of the composite. This is the case for power-law materials for which, due to the Euler theorem for homogeneous functions of degree $n+1$, one has $\bar{\boldsymbol{\sigma}} : \bar{\boldsymbol{\varepsilon}} = (n+1)\tilde{w}^*(\bar{\boldsymbol{\sigma}})$. Therefore, any bound on $\tilde{w}^*$ gives a bound on the overall strain $\bar{\boldsymbol{\varepsilon}}$ in the direction of the applied stress $\bar{\boldsymbol{\sigma}}$. However, no information on the other components of the strain is provided. Interestingly, a method for bounding directly the stress-strain relation has been recently proposed by Milton and Serkov [19].

10.3.3.2.2 Complementary Energies Depending Only on the Quadratic Stress

We consider here a broad class of behaviors corresponding to potentials w^* which depend on the stress tensor $\boldsymbol{\sigma}$ only through the quadratic stress $\mathscr{S}$, $w^*(\boldsymbol{\sigma}) = G(\mathscr{S})$, for some appropriately chosen function G and where $\mathscr{S} = \frac{1}{2}\boldsymbol{\sigma} \otimes \boldsymbol{\sigma}$. G is further assumed to be a *convex* function of $\mathscr{S}$. This class of materials contains in particular all materials with a complementary energy in the form $w^*(\boldsymbol{\sigma}) = \frac{1}{2k}\sigma_m^2 + \psi(\sigma_{eq})$.

For this class of materials, the function $v^{(r)}$ (corresponding to v in phase r) can be bounded from above by

$$v^{(r)}\left(\boldsymbol{M}_0^{(r)}\right) = \sup_{\boldsymbol{\tau}} \left[\boldsymbol{M}_0^{(r)} :: \mathscr{T} - G^{(r)}(\mathscr{T}) \right] \leq \left(G^{(r)}\right)^* \left(\boldsymbol{M}_0^{(r)}\right), \tag{30}$$

where $\mathscr{T} = \frac{1}{2}\boldsymbol{\tau} \otimes \boldsymbol{\tau}$

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$(G^{(r)})^*$ denotes the Legendre transform of the convex function $G^{(r)}$:

$$\left(G^{(r)}\right)^*(\boldsymbol{M}) = \sup_{\mathscr{T}} \left[\boldsymbol{M} :: \mathscr{T} - G^{(r)}(\mathscr{T})\right] \tag{31}$$

Note that in Eq. 31 the supremum is taken over all symmetric fourth-order $\mathscr{T}$, whereas the supremum in Eq. 30 is restricted to rank-one symmetric $\mathscr{T}$. The lower bound (Eq. 29) reduces to

$$\tilde{w}^*(\bar{\boldsymbol{\sigma}}) \geq \sup_{\boldsymbol{M}_0^{(r)}>0} \left(\frac{1}{2}\bar{\boldsymbol{\sigma}} : \tilde{\boldsymbol{M}}_0\left(\left\{\boldsymbol{M}_0^{(r)}\right\}\right)_{r=1,\ldots,N} : \bar{\boldsymbol{\sigma}} - \sum_{r=1}^{N} c^{(r)}\left(G^{(r)}\right)^*\left(\boldsymbol{M}_0^{(r)}\right)\right) \tag{32}$$

10.3.3.3 Connection with the Secant Method Based on Second-Order Moments

When the complementary energy of the constitutive phases depends only on the “quadratic stress” $\mathscr{S}$, the constitutive relation (Eq. 24) can alternatively be written as

$$\boldsymbol{\varepsilon} = \boldsymbol{M}_{sct}(\mathscr{S}) : \boldsymbol{\sigma}, \quad \text{where } \boldsymbol{M}_{sct}(\mathscr{S}) = \frac{\partial G}{\partial \mathscr{S}}(\mathscr{S}), \quad \text{or equivalently } \mathscr{S} = \frac{\partial G^*}{\partial \boldsymbol{M}}(\boldsymbol{M}_{sct}) \tag{33}$$

We are now going to inspect in more detail the optimality conditions for the moduli $\boldsymbol{M}_0^{(r)}$ derived from the optimization problem (Eq. 32). Assuming stationarity with respect to these moduli, the optimality conditions read as

$$\frac{1}{2}\bar{\boldsymbol{\sigma}} : \frac{\partial \tilde{\boldsymbol{M}}_0}{\partial \boldsymbol{M}_0^{(r)}} : \bar{\boldsymbol{\sigma}} = c^{(r)}\frac{\partial\left(G^{(r)}\right)^*}{\partial \boldsymbol{M}}\left(\boldsymbol{M}_0^{(r)}\right)$$

But according to Eq. 16, the first term in this equality is nothing other than the average second-order moment of the stress in the linear comparison solid. Therefore:

$$\tilde{\mathscr{S}}^{(r)} = \frac{1}{2}\langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle_r = \frac{\partial\left(G^{(r)}\right)^*}{\partial \boldsymbol{M}}\left(\boldsymbol{M}_0^{(r)}\right)$$

Making use of Eq. 33, the optimally conditions finally amount to solving the following systems of nonlinear equations:

$$\boldsymbol{M}_0^{(r)} = \boldsymbol{M}_{sct}^{(r)}\left(\tilde{\mathscr{S}}^{(r)}\right), \quad \text{with} \quad \tilde{\mathscr{S}}^{(r)} = \frac{1}{2c^{(r)}}\bar{\boldsymbol{\sigma}} : \frac{\partial \tilde{\boldsymbol{M}}_0}{\partial \boldsymbol{M}_0^{(r)}} : \bar{\boldsymbol{\sigma}}$$

This systems coincides with Eq. 17.

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In conclusion, it has been shown that the optimal moduli $\boldsymbol{M}_0^{(r)}$ in the variational procedure coincide with the secant moduli $\boldsymbol{M}^{(r)}$ determined by the (more heuristic) secant method based on the second-order moment described in Section 10.3.2.5. In other words, the variational procedure can be interpreted as a secant method. It has, however, the definite advantage of delivering a clear rigorous bound on the effective properties of the nonlinear composite.

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