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EXTREMUM PRINCIPLES IN NON-LINEAR ELASTICITY AND THEIR APPLICATION TO COMPOSITES—I

THEORY

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Abstract—This is the first of two papers concerned with the overall properties of inhomogeneous elastic solids which can support finite elastic strain. Part I deals with the theoretical development. The first objective is to obtain extremum principles which are direct generalizations of those available in the linear theory. For such principles to be valid at finite strain it is necessary to restrict attention to situations where the energy density of the elastic material is convex (in a sense to be described). An immediate consequence of the extremum principles is that the overall energy of the composite may be bounded from above and below, bounds thereby being placed on the overall moduli occurring in the macroscopic constitutive law of the overall material. This is analogous to the linear theory, but there are certain difficulties which arise when non-linearity is considered and these are discussed in detail.

It is important to be able to analyse the effects of introducing inclusions into a material, having regard to strengthening and weakening. This is particularly relevant in the case of rubberlike solids which are strengthened by fillers such as carbon black. Some strengthening and weakening theorems analogous to those known in the linear theory are therefore discussed. Part II is concerned with the application of the theoretical results.

1. INTRODUCTION

It is common practice to use fillers consisting of particles of, for example, carbon black and silicone in order to improve the mechanical properties of rubberlike solids. In particular, the tensile strength of rubber can be increased if appropriate volume concentrations of filler are used. It is therefore important to be able to analyse the overall mechanical properties of the composite material in terms of the properties of its constituents (the filler particles being effectively rigid). Since rubberlike solids can undergo large elastic deformations the non-linear theory of elasticity is required in the analysis.

Unfortunately, the existing body of theory which deals with the overall properties of elastic composites is set almost exclusively in the context of infinitesimal deformations, and is not appropriate for the discussion of rubberlike composites. The objective of this paper (Part I) therefore is to develop various aspects of the non-linear theory of elastic composites, generalizing results of the linear theory where possible. Applications of the theory are discussed in the companion paper (Part II).

As in the linear theory very little can be done by way of exact analysis towards the calculation of "overall" quantities, given the constitution and geometry of the constituent materials. In the linear theory there is a considerable literature concerned with the derivation of bounds on the overall elastic constants (see, e.g. [1–10]).

The methods used rely on the extremum properties of the solutions of the boundary-value problems of place and traction, and, more particularly, on the assumption that the elastic strain energy is positive definite. *No corresponding assumption is appropriate for all deformations in the non-linear theory*, as pointed out by Hill[11]. However, certain *stationary* principles are available in finite elasticity (see, e.g. [12, 13]; [14], Section 88), but, as observed by Sewell[15], the variational principles (of energy and complementary energy) cannot in general be strengthened to extremum principles. Ogden[16] has reviewed the various variational principles and emphasised particular functionals which will be useful in the present paper. The extent of the validity of the principles has been discussed in detail by Ogden[17], and the relevant results are summarized in Section 2 of the present paper. See also [37].

This work provides a basis for strengthening the variational principles to extremum principles and an understanding of the range of validity of the extremum principles at large deformations. Essentially it is necessary to restrict attention to configurations in which the material is stable *in a restricted sense.* This requires consideration of certain convexity properties of the strain-energy function of the elastic material in question, and for this purpose we draw on some results given in [18]. In Section 3 the extremum principles and their limitations are discussed.

In Section 4, following Hill[19], Hill and Rice[20] and Ogden[21], we define an "overall constitutive law" for the composite material in terms of certain macroscopic measures of stress and deformation. This enables us, with the help of the extremum principles, to obtain upper and lower bounds on the *macroscopic* energy and complementary energy of the composite. In particular, we obtain immediate generalizations to non-linear elasticity of the Reuss–Voigt–Hill estimates for the moduli of composite materials.

The *strengthening theorem* of Hill ([5], p. 370; see also Walpole[22], Section 3) is generalized to the case of finite deformations in Section 5, and its local and overall ramifications discussed.

2. STATIONARY CHARACTER OF THE ENERGY AND COMPLEMENTARY ENERGY

Let X_μ and x_i ($\mu, i = 1, 2, 3$) be the initial and final coordinates of a typical material point. The deformation gradient $\partial x_i/\partial X_\mu \equiv x_{i,\mu}$ is denoted by $\alpha_{i\mu}$, or $\boldsymbol{\alpha}$ in symbolic notation. Cartesian coordinates are used throughout. We consider an elastic material which has strain-energy function W per unit initial volume. The material is assumed to have a stress-free ground state (taken as the initial and reference configuration here) in which it has volume V, bounded by surface Σ.

From the polar decomposition theorem we note that $\boldsymbol{\alpha}$ can be decomposed as the product $\mathbf{r}\times\mathbf{u}$ of the proper orthogonal $\mathbf{r}$ and right stretch $\mathbf{u}$ (positive definite and symmetric).

The nominal stress $\mathbf{s} \equiv s_{\mu i}$ is given by

$$\mathbf{s} = \partial W/\partial\boldsymbol{\alpha}, \qquad s_{\mu i} = \partial W/\partial\alpha_{i\mu} \tag{1}$$

and satisfies the equilibrium equations

$$s_{\mu i,\mu} = 0 \tag{2}$$

when there are no body forces (these can easily be incorporated into the analysis if required). W depends on $\boldsymbol{\alpha}$ only through a frame-indifferent quantity such as $\mathbf{u}^2 \equiv \boldsymbol{\alpha}^{\mathrm{T}}\times\boldsymbol{\alpha}$, where $\boldsymbol{\alpha}^{\mathrm{T}}$ is the transpose of $\boldsymbol{\alpha}$.

The stress-deformation relation may also be written

$$\boldsymbol{\tau} = \frac{\partial W}{\partial \mathbf{u}}, \tag{3}$$

where $\boldsymbol{\tau}$ is the symmetric stress (conjugate to $\mathbf{u}$) given by

$$\boldsymbol{\tau} = \frac{1}{2}(\mathbf{s}\times\mathbf{r} + \mathbf{r}^{\mathrm{T}}\times\mathbf{s}^{\mathrm{T}}). \tag{4}$$

It is important to note that in view of the symmetry of $\mathbf{u}$ the identity

$$\mathbf{s}\boldsymbol{\alpha} = \boldsymbol{\tau}\mathbf{u} \tag{5}$$

holds, where $\mathbf{s}\boldsymbol{\alpha} \equiv s_{\mu i}\alpha_{i\mu}$ and similarly for $\boldsymbol{\tau}\mathbf{u}$. *The variables* $\boldsymbol{\tau}$ *and* $\mathbf{u}$ *play a central role in this paper.* (If $\boldsymbol{\alpha}$ is replaced by the displacement gradient the simplicity of (5) is lost, as is the frame-indifference.)

The complementary-energy function W_c is defined by means of the equation

$$W + W_c = \mathbf{s}\boldsymbol{\alpha} \equiv \boldsymbol{\tau}\mathbf{u}, \tag{6}$$

and it may be interpreted as a function of s or of $\boldsymbol{\tau}$ at deformations for which $\mathbf{s}(\boldsymbol{\alpha})$ and $\boldsymbol{\tau}(\mathbf{u})$ respectively are *locally invertible.* A full discussion of the existence of these inversions has been given in [17].

The inverses of (1) and (3), when they exist, are written

$$\boldsymbol{\alpha} = \frac{\partial W_c}{\partial \mathbf{s}} \tag{7}$$

and

$$\mathbf{u} = \frac{\partial W_c}{\partial \boldsymbol{\tau}} \tag{8}$$

respectively with the help of (6). The inversion (8) has a wider range of validity than (7). W_c exists at points where $\partial\boldsymbol{\tau}/\partial\mathbf{u}$ is non-singular, but can only be regarded as a function of s where $\partial\mathbf{s}/\partial\boldsymbol{\alpha}$ is non-singular. Ogden[17] has shown that singularity of $\partial\boldsymbol{\tau}/\partial\mathbf{u}$ implies that of $\partial\mathbf{s}/\partial\boldsymbol{\alpha}$ for isotropic elastic solids, but the converse is not true. Some further remarks about invertibility will be required in Section 3, but it is worth noting here that a given s can be associated with several distinct $\boldsymbol{\alpha}$'s (at least four for isotropic elastic solids) in the global sense[17].

The boundary conditions to be imposed on the complementary parts Σ_x and Σ_s of Σ are

$$\mathbf{x} = \mathbf{x}_0 \quad \text{on } \Sigma_x, \tag{9}$$

$$\mathbf{t} = \mathbf{t}_0 \quad \text{on } \Sigma_s, \tag{10}$$

where $\mathbf{x}_0$ and $\mathbf{t}_0$ are prescribed functions, $t_i = s_{\mu i} n_\mu$, and $\mathbf{n}$ is the unit normal to Σ.

We now introduce the "energy" and complementary "energy" functionals E and E_c defined by

$$E(\mathbf{x}) = \int_V W(\boldsymbol{\alpha})\,\mathrm{d}V - \int_{\Sigma_s} \mathbf{x}\mathbf{t}_0\,\mathrm{d}\Sigma \tag{11}$$

$$E_c(\mathbf{s}) = \int_{\Sigma_x} \mathbf{t}\mathbf{x}_0\,\mathrm{d}\Sigma - \int_V W_c(\mathbf{s})\,\mathrm{d}V. \tag{12}$$

The functional $E(\mathbf{x})$ is defined for all continuous and (piecewise) continuously differentiable functions $\mathbf{x}(\mathbf{X})$ (with $\boldsymbol{\alpha} \equiv x_{i,\mu}$) consistent with (9). $E_c(\mathbf{s})$ is defined for all (piecewise) differentiable nominal "stress" fields s *for which the inversion* (7) *exists,* where $t_i = s_{\mu i} n_\mu$ is consistent with (10) and is continuous across any surface within V which has unit normal $\mathbf{n}$. Because of the continuity of $\mathbf{x}$ and $\mathbf{t}$ there is no need to consider discontinuity surfaces in the variational analysis.

Where (7) does not exist it is necessary to regard E_c as a function of s *and* proper orthogonal $\mathbf{r}$ independently[17]. Then

$$E_c(\mathbf{s}, \mathbf{r}) = \int_{\Sigma_x} \mathbf{t}\mathbf{x}_0\,\mathrm{d}\Sigma - \int_V W_c(\boldsymbol{\tau})\,\mathrm{d}V, \tag{13}$$

where $\boldsymbol{\tau}$ is the symmetric part of $\mathbf{s} \times \mathbf{r}$ and is such that the inversion (8) exists. The values of (12) and (13) are of course equal for a given s and $\mathbf{r}$ in view of (6).

For an actual solution pair $(\mathbf{x}, \mathbf{s})$ of the boundary-value problem defined by eqns (1), (2), (9) and (10), and for a given (frame-indifferent) form of W it is easily verified that

$$E = E_c. \tag{14}$$

It can be shown (see, e.g. [16, 17]) that, within the class of admissible functions $\mathbf{x}$ satisfying (9) and associated through (1) with an s, E is stationary if and only if s satisfies (2) and (10). Dually, within the class of admissible *self-equilibrated* fields s satisfying (10) and associated through (7) with an $\boldsymbol{\alpha}$ (when this is valid), E_c given by (12) is stationary if and only if $\boldsymbol{\alpha}$ is expressible as the gradient of some $\mathbf{x}$ satisfying (9). More generally, within the class of self-equilibrated nominal fields $\mathbf{s}$, satisfying (10), *and* proper orthogonal $\mathbf{r}$, with $\boldsymbol{\tau}$ defined as the symmetric part of $\mathbf{s}\times\mathbf{r}$ associated with a $\mathbf{u}$ through (8), E_c given by (13) is stationary if and only if $\mathbf{r}\times\mathbf{u}$ is expressible as the gradient of some $\mathbf{x}$, satisfying (9), *and* $\boldsymbol{\tau}$ is frame-indifferent.†

Details of the admissible variations appropriate to each case are given in [16, 17] and need not be listed here.

Under certain circumstances, these variational theorems can be strengthened to extremum principles. In Section 3 the extremum principles are first of all stated and there then follows a discussion of the limitations which must be imposed on their range of validity.

3. EXTREMUM PRINCIPLES

3.1 *Some convexity considerations*

We consider the domain, denoted by $\mathscr{D}_\alpha$, in the nine-dimensional Euclidean space of second-order tensors $\boldsymbol{\alpha}$. Let $W(\boldsymbol{\alpha})$ be a scalar function of $\boldsymbol{\alpha}$ and suppose $\mathbf{s}\equiv\mathbf{s}(\boldsymbol{\alpha})$ is given by eqn (1). Then W is a strictly convex function of $\boldsymbol{\alpha}$ on $\mathscr{D}_\alpha$ if

$$W(\boldsymbol{\alpha}^*)-W(\boldsymbol{\alpha})-\mathbf{s}(\boldsymbol{\alpha}^*-\boldsymbol{\alpha})>0 \tag{15}$$

for all $\boldsymbol{\alpha}$ and $\boldsymbol{\alpha}^*\neq\boldsymbol{\alpha}$ in $\mathscr{D}_\alpha$.

A consequence of (15) is that the local inequality

$$\dot{\mathbf{s}}\dot{\boldsymbol{\alpha}}\geq 0 \tag{16}$$

holds for all points $\boldsymbol{\alpha}$ in $\mathscr{D}_\alpha$ and arbitrary variations $\dot{\boldsymbol{\alpha}}$, with $\dot{\mathbf{s}}=\mathscr{L}(\boldsymbol{\alpha})\dot{\boldsymbol{\alpha}}$ and $\mathscr{L}(\boldsymbol{\alpha})=\partial\mathbf{s}/\partial\boldsymbol{\alpha}$. In (16) equality holds for $\dot{\boldsymbol{\alpha}}\neq\mathbf{0}$ only on a nowhere dense set in $\mathscr{D}_\alpha$ [23].

The relation $\mathbf{s}(\boldsymbol{\alpha})$ is invertible locally at all points of $\mathscr{D}_\alpha$ except where

$$\det\mathscr{L}(\boldsymbol{\alpha})=0. \tag{17}$$

In elasticity theory the domain $\mathscr{D}_\alpha$ is bounded, as is well known, and in fact the stress-free configuration is a point where (17) holds [17, 24] independently of the form of W. It is required that W is indifferent to any superposed rotation in the deformed configuration, so that if we set $\boldsymbol{\alpha}^*=\mathbf{q}\times\boldsymbol{\alpha}$, where $\mathbf{q}$ is proper orthogonal, (15) imposes restrictions on the states of stress associated with points in $\mathscr{D}_\alpha$ ([14], p. 163).

If the material possesses symmetry such as isotropy the choice $\boldsymbol{\alpha}^*=\boldsymbol{\alpha}\times\mathbf{q}$ leads to similar restrictions, although this point seems not to have been noticed previously.

When the strict inequality (16) holds for $\dot{\boldsymbol{\alpha}}\neq 0$ the material is locally stable in the sense of Hill [24].

We now consider the domain $\mathscr{D}_\mathbf{u}$ in the six-dimensional Euclidean space of symmetric second-order tensors $\mathbf{u}$, W being regarded as a scalar function of $\mathbf{u}$. Suppose $\boldsymbol{\tau}\equiv\boldsymbol{\tau}(\mathbf{u})$ is given by eqn (3). Then W is a strictly convex function of $\mathbf{u}$ on $\mathscr{D}_\mathbf{u}$ if

$$W(\mathbf{u}^*)-W(\mathbf{u})-\boldsymbol{\tau}(\mathbf{u}^*-\mathbf{u})>0 \tag{18}$$

for all $\mathbf{u}$ and $\mathbf{u}^*\neq\mathbf{u}$ in $\mathscr{D}_\mathbf{u}$.

Analogously to (16), (18) implies

$$\dot{\boldsymbol{\tau}}\dot{\mathbf{u}}\geq 0 \tag{19}$$

for all $\mathbf{u}$ in $\mathscr{D}_\mathbf{u}$ and arbitrary variations $\dot{\mathbf{u}}$, with $\dot{\boldsymbol{\tau}}=\mathscr{L}(\mathbf{u})\dot{\mathbf{u}}$ and $\mathscr{L}(\mathbf{u})=\partial\boldsymbol{\tau}/\partial\mathbf{u}$, equality holding only on a nowhere dense set in $\mathscr{D}_\mathbf{u}$.

The relation $\boldsymbol{\tau}(\mathbf{u})$ is locally invertible at all points of $\mathscr{D}_\mathbf{u}$ except where

$$\det\mathscr{L}(\mathbf{u})=0. \tag{20}$$

†The term *frame-indifferent* is used here to describe a quantity which is unaffected by a superposed rigid-body rotation after deformation.

For actual elastic solids the strict inequality (19) certainly holds in some domain enclosing the undeformed configuration (stress-free), and in the context of infinitesimal deformation theory it corresponds to the customary requirement of positive definite strain energy. *For rubberlike solids all the available experimental evidence indicates that the strict inequality* (19) *holds up to material rupture* [17].

It is therefore reasonable to suppose that (18) also holds for all deformations of practical interest. If the incompressibility constraint is imposed (19) can be violated[25], but when the actual compressibility of real materials is allowed for (even though it may only be small) (19) is entirely consistent with the elastic response of rubberlike solids up to rupture.

The inequality (18) is the basis for a discussion of constitutive inequalities for elastic solids by Krawietz[26], and when $\boldsymbol{\alpha}^* \times \boldsymbol{\alpha}^{-1}$ is restricted to being positive definite, symmetric and not equal to the identity the inequality (15), for *all* $\boldsymbol{\alpha}$ and $\boldsymbol{\alpha}^*$ so restricted, is the Coleman–Noll hypothesis ([14], Sections 52, 87, [27–30], [31] Section 8).

We must emphasise that Krawietz[26] proposed that (18) should hold for all reference configurations and all deformations, whereas we restrict attention to a single reference configuration (the stress-free configuration) and assume that (18) holds at least up to those $\mathbf{u}$ corresponding to rupture.

In view of the above discussion we deduce that the eqn (20) cannot hold for deformations of practical interest, and therefore $\boldsymbol{\tau}(\mathbf{u})$ is locally invertible. It follows that the complementary-energy function W_c exists and the inversion (8) is valid globally in the context of the *elastic* behaviour of actual materials.

Let $\mathscr{D}_\tau$ be the image space of $\mathscr{D}_\mathbf{u}$ under the mapping $\boldsymbol{\tau}(\mathbf{u})$ (which is globally one-to-one on $\mathscr{D}_\mathbf{u}$). Then, by (6) and (18),

$$W_c(\boldsymbol{\tau}^*) - W_c(\boldsymbol{\tau}) - \mathbf{u}(\boldsymbol{\tau}^* - \boldsymbol{\tau}) > 0 \tag{21}$$

for all $\boldsymbol{\tau}$ and $\boldsymbol{\tau}^* \neq \boldsymbol{\tau}$ in $\mathscr{D}_\tau$.

Similarly, by (6) and (15),

$$W_c(\mathrm{s}^*) - W_c(\mathrm{s}) - \boldsymbol{\alpha}(\mathrm{s}^* - \mathrm{s}) > 0 \tag{22}$$

for all s and $\mathrm{s}^* \neq \mathrm{s}$ in $\mathscr{D}_\mathrm{s}$, the image space of $\mathscr{D}_\alpha$ under the mapping $\mathrm{s}(\boldsymbol{\alpha})$ (which is globally one-to-one on $\mathscr{D}_\alpha$).

The convexity conditions (15) *and* (22) *are the inequalities on which the extremum principles are based. However, the domains of validity of* (15) *and* (22) *are limited.* Indeed, those $\mathbf{u}$ for which (15) holds, with $\boldsymbol{\alpha} = \mathbf{r} \times \mathbf{u}$, constitute a subdomain of $\mathscr{D}_\mathbf{u}$ touching the origin (the undeformed configuration). The relationship between the domains $\mathscr{D}_\alpha$ and $\mathscr{D}_\mathbf{u}$ has been discussed in detail by Ogden[17] relative to the local inequalities (16) and (19) with particular reference to isotropic elasticity.

For obvious reasons it is desirable for the extremum principles to be valid on the whole of $\mathscr{D}_\mathbf{u}$ and $\mathscr{D}_\tau$. The extent to which this validity can be achieved will be discussed following an examination of the extremum principles themselves.

In practice it is the *overall* form of the inequality (15) that is required, namely

$$\int_V \{W(\boldsymbol{\alpha}^*) - W(\boldsymbol{\alpha}) - \mathrm{s}(\boldsymbol{\alpha}^* - \boldsymbol{\alpha})\}\,\mathrm{d}V > 0, \tag{23}$$

where $\boldsymbol{\alpha}^* \neq \boldsymbol{\alpha}$ on some subset of V. Clearly, it is possible for (15) to be violated in part of V when (23) holds, and in this sense (23) is more general than (15).

Correspondingly, the overall form of (22) is

$$\int_V \{W_c(\mathrm{s}^*) - W_c(\mathrm{s}) - \boldsymbol{\alpha}(\mathrm{s}^* - \mathrm{s})\}\,\mathrm{d}V > 0, \tag{24}$$

where $\mathrm{s}^* \neq \mathrm{s}$ on some subset of V, provided (17) does not hold anywhere in V *in respect of* $\boldsymbol{\alpha}$.

If, in (23), $\boldsymbol{\alpha}$ and $\boldsymbol{\alpha}^*$ are respectively the gradients (with respect to initial coordinates $\mathbf{X}$) of functions $\mathbf{x}$ and $\mathbf{x}^*$, each of which satisfies the boundary condition (9), then, provided s satisfies

(2) and (10) and $\mathbf{x}^* \neq \mathbf{x}$ on some subset of V, we obtain

$$\int_V W(\boldsymbol{\alpha}^*)\,\mathrm{d}V - \int_{\Sigma_s} \mathbf{x}^*\mathbf{t}_0\,\mathrm{d}\Sigma > \int_V W(\boldsymbol{\alpha})\,\mathrm{d}V - \int_{\Sigma_s} \mathbf{x}\mathbf{t}_0\,\mathrm{d}\Sigma \tag{25}$$

by use of the divergence theorem. Thus, within the class of admissible functions $\mathbf{x}^*$ the left-hand functional in (25) is minimised when the associated $\mathbf{s}^* \equiv \mathbf{s}(\boldsymbol{\alpha}^*)$ satisfies (2) and (10).

Dually, from (24), we obtain

$$\int_V W_c(\mathbf{s}^*)\,\mathrm{d}V - \int_{\Sigma_x} \mathbf{t}^*\mathbf{x}_0\,\mathrm{d}\Sigma > \int_V W_c(\mathbf{s})\,\mathrm{d}V - \int_{\Sigma_x} \mathbf{t}\mathbf{x}_0\,\mathrm{d}\Sigma \tag{26}$$

for the class of admissible fields $\mathbf{s}$ and $\mathbf{s}^*$ satisfying (2) and (10), provided $\boldsymbol{\alpha}$ is the gradient of some $\mathbf{x}$ which satisfies (9).

The word *admissible* is used above to indicate that $\boldsymbol{\alpha}^*$ and $\mathbf{s}^*$ in (25) and (26) respectively are in the appropriate domain in $\boldsymbol{\alpha}$-space and $\mathbf{s}$-space respectively for the validity of (23) and (24).

In the literature there is very little concerned with the derivation and application of extremum principles in respect of elastic solids under finite strain, although such principles have been alluded to in passing by various authors (e.g. [15]). Hill[32] has stated extremum principles (essentially (25) and (26) reinterpreted) in a different context and mentioned their applicability to elasticity theory ([32], pp. 68–9). See also Hill's paper[11] on the stability of elastic solids under finite strain.

Lind[33] has derived extremum principles for a special class of elastic constitutive laws. These are such that W is a homogeneous function (of degree greater than one) in the *displacement* gradient. Lind assumes that W is strictly locally convex, that is that strict inequality (16) holds, thereby restricting attention to locally stable configurations. However, the particular constitutive law Lind uses ([33], eqns (32), (34)) in the application of his extremum principles is neither isotropic (as he claims) nor frame indifferent.

3.2 *The extremum principles*

Let $(\mathbf{x}, \mathbf{s})$ be an actual solution (generally unknown) of the boundary-value problem defined in Section 2. The energy and complementary energy functionals $E(\mathbf{x})$ and $E_c(\mathbf{s})$ are given by eqns (11) and (12) (or (13)) respectively.

We also write

$$E(\mathbf{x}^*) = \int_V W(\boldsymbol{\alpha}^*)\,\mathrm{d}V - \int_{\Sigma_s} \mathbf{x}^*\mathbf{t}_0\,\mathrm{d}\Sigma \tag{27}$$

for all continuous and (piecewise) continuously differentiable functions $\mathbf{x}^*(\mathbf{X})$ satisfying the boundary condition (9).

It follows from (25) that

$$E(\mathbf{x}) \le E(\mathbf{x}^*) \tag{28}$$

within the class of admissible $\mathbf{x}^*$, with equality if and only if $\mathbf{x}^* = \mathbf{x}$ in V.

Similarly, we write

$$E_c(\mathbf{s}^*) = \int_{\Sigma_x} \mathbf{t}^*\mathbf{x}_0\,\mathrm{d}\Sigma - \int_V W_c(\mathbf{s}^*)\,\mathrm{d}V \tag{29}$$

for all (piecewise) differentiable self-equilibrated $\mathbf{s}^*$ satisfying the boundary condition (10) and for which W_c is defined.

From (26) we then obtain

$$E_c(\mathbf{s}) \ge E_c(\mathbf{s}^*) \tag{30}$$

within the class of admissible $\mathbf{s}^*$, with equality if and only if $\mathbf{s}^* = \mathbf{s}$ in V.

We note that $\boldsymbol{\alpha}^*$, derived from $\mathbf{x}^*$, in (28) and $\mathbf{s}^*$ in (30) are not related through the constitutive law (1) unless $\mathbf{s}^*=\mathbf{s}$ and $\mathbf{x}^*=\mathbf{x}$. Otherwise $(\mathbf{x}^*, \mathbf{s}^*)$ would represent a second solution to the boundary-value problem and (28) would be contradicted. *This does not rule out the possibility of more than one solution to a given boundary-value problem, but indicates that the solution is unique within the class of admissible fields.*

With the help of (14) we combine (28) and (30) in the form

$$E(\mathbf{x}^*) \geq E(\mathbf{x}) = E_c(\mathbf{s}) \geq E_c(\mathbf{s}^*). \tag{31}$$

The first inequality in (31) bears a close resemblance to that required for stability of the equilibrium solution $(\mathbf{x}, \mathbf{s})$ under dead loading on Σ_s ([14], Section 89; [11], Section 3), and to this extent the domain of validity of (31) is limited. However, in view of the close relation between the variables $(\mathbf{s}, \boldsymbol{\alpha})$ and $(\boldsymbol{\tau}, \mathbf{u})$ and the fact that the inequality (18) has a much larger range of validity than (15) we are able to extend the domain of validity of the extremum principles. Essentially this is done by placing certain taboos on the relation between the $\boldsymbol{\alpha}$'s and $\boldsymbol{\alpha}^*$'s occurring in (15), and correspondingly on the relation between the s's and s*'s in (22). We are then no longer restricted to configurations which are stable under dead loading.

3.3 *Extension of the range of validity*

As already indicated the domain $\mathcal{D}_\alpha$ is not sufficiently extensive to cover all configurations of practical interest. It is, however, worth remarking that its image space $\mathcal{D}_s$ is associated with points outside $\mathcal{D}_\alpha$ because there are several branches of the inversion $\boldsymbol{\alpha}(\mathbf{s})$ in the global sense.

In order to make these points clearer we consider the local inequalities

$$\dot{\mathbf{s}}\dot{\boldsymbol{\alpha}} > 0 \tag{32}$$

and

$$\dot{\boldsymbol{\tau}}\dot{\mathbf{u}} > 0 \tag{33}$$

in more detail, and to be specific we confine attention to isotropic elastic solids. Then $\mathbf{u}$ and $\boldsymbol{\tau}$ are coaxial, their common principal axes being referred to as the Lagrangean axes. Let λ_i and τ_i $(i = 1, 2, 3)$ respectively be the principal values of $\mathbf{u}$ and $\boldsymbol{\tau}$.

Following Ogden[17] we decompose (32) and (33) on the Lagrangean axes in the forms

$$\dot{\boldsymbol{\tau}}\dot{\mathbf{u}} \equiv \sum_{i,j=1}^{3} \frac{\partial \tau_i}{\partial \lambda_j} \dot{\lambda}_i \dot{\lambda}_j + \sum_{i \neq j} (\omega_{ij}^L)^2 (\tau_i - \tau_j)(\lambda_i - \lambda_j) > 0, \tag{34}$$

$$\dot{\mathbf{s}}\dot{\boldsymbol{\alpha}} \equiv \dot{\boldsymbol{\tau}}\dot{\mathbf{u}} + \frac{1}{2} \sum_{i \neq j} (\omega_{ij}^r)^2 (\lambda_i \tau_i + \lambda_j \tau_j) + \sum_{i \neq j} \omega_{ij}^L \omega_{ij}^r (\tau_i - \tau_j)(\lambda_i - \lambda_j) > 0, \tag{35}$$

the latter of which we now write as

$$\dot{\mathbf{s}}\dot{\boldsymbol{\alpha}} \equiv \sum_{i,j=1}^{3} \frac{\partial \tau_i}{\partial \lambda_j} \dot{\lambda}_i \dot{\lambda}_j + \sum_{i \neq j} \left(\omega_{ij}^L + \frac{1}{2}\omega_{ij}^r\right)^2 (\tau_i - \tau_j)(\lambda_i - \lambda_j) + \frac{1}{4} \sum_{i \neq j} (\omega_{ij}^r)^2 (\tau_i + \tau_j)(\lambda_i + \lambda_j) > 0. \tag{36}$$

The ω_{ij}^L are the components of the spin of the Lagrangean axes (on those axes), and

$$\omega_{ij}^r = \omega_{ij} + \omega_{ij}^L \frac{(\lambda_i - \lambda_j)^2}{2\lambda_i \lambda_j} \qquad (\lambda_i \neq \lambda_j),$$

where ω_{ij} is the body spin (the antisymmetric part of $\dot{\boldsymbol{\alpha}} \times \boldsymbol{\alpha}^{-1}$) on the Lagrangean axes. Note also that ω_{ij}^r are the components of $\mathbf{r}^T \times \dot{\mathbf{r}}$ on the Lagrangean axes. Further details can be found in [17]. It is worth remarking that if ω_{ij} is set equal to zero in (35) or (36) the inequality reduces to the local form of the Coleman–Noll inequality ([14], pp. 166–7).

Clearly, necessary and sufficient conditions for (34) to hold for arbitrary, non-zero $\dot{\mathbf{u}}$ are

$$\frac{\partial \tau_i}{\partial \lambda_j} \quad \text{is positive definite} \tag{37}$$

and

$$\frac{\tau_i - \tau_j}{\lambda_i - \lambda_j} > 0 \qquad i \neq j \tag{38}$$

provided λ_i $(i = 1,2,3)$ and ω_{ij}^L $(i \neq j)$ are independent. Similarly, from (36), necessary and sufficient conditions for (36) are (37), (38) *and*

$$\tau_i + \tau_j > 0 \qquad i \neq j, \tag{39}$$

when ω_{ij}^r $(i \neq j)$ also are independent.

From (35) it is also clear that

$$\sigma_i + \sigma_j > 0 \qquad i \neq j \tag{40}$$

are necessary conditions for $\dot{s}\dot{\alpha} > 0$, where $J\sigma_i = \lambda_i \tau_i$ $(i = 1, 2, 3)$ and $J = \lambda_1\lambda_2\lambda_3$, in terms of the principal components σ_i of true stress.

Certainly (36) implies (34) but the converse is not in general true. Similarly, for the global inequalities: (15) implies (18). Moreover, (40) are also necessary for (15)[14], and hence configurations in which one or more of (40) fail are excluded from the domain $\mathscr{D}_\alpha$.

All the available experimental evidence suggests that the inequality (33) holds up to the limit of validity of elasticity theory. In particular, it has been shown by Ogden[17, 18] that for rubberlike solids the inequality holds up to rupture, and certainly does not fail *at* rupture.

For practical purposes we can therefore assume that (33) *holds for all deformations* **u**, *and correspondingly we assume that the global inequality* (18) *holds for all unequal* **u** *and* **u*** *for which elasticity theory is valid. It immediately follows that* W_c *exists and the inverse* (8) *is unique in* $\mathscr{D}_\mathbf{u}$. The boundary of the domain of elastic response is within $\mathscr{D}_\mathbf{u}$ (although, exceptionally, it may coincide with the boundary of $\mathscr{D}_\mathbf{u}$[18].

We now see that $\partial\boldsymbol{\tau}/\partial\mathbf{u}$ is non-singular in $\mathscr{D}_\mathbf{u}$, but there are points within $\mathscr{D}_\mathbf{u}$ where $\partial s/\partial\boldsymbol{\alpha}$ is singular. These points form a nowhere dense set in $\mathscr{D}_\mathbf{u}$ (or more specifically in its analogue in λ_i-space for isotropic solids) distributed according to the equations

$$\tau_i + \tau_j = 0 \qquad i \neq j \tag{41}$$

[17, 24].

Aside from configurations where one or more of (41) hold $\partial s/\partial\boldsymbol{\alpha}$ is non-singular for all **u** in $\mathscr{D}_\mathbf{u}$ (irrespective of **r**) for isotropic elastic solids. Corresponding results can be found for non-isotropic solids.

It follows that except where $\tau_i + \tau_j = 0$, $i \neq j$, the relation $s(\boldsymbol{\alpha})$ is locally invertible and (7) is valid. However, it is important to remark that the inverse is not unique in the global sense on $\mathscr{D}_\mathbf{u}$. This has been discussed in detail by Ogden[17], and we review the salient points here.

For isotropic elastic solid the relation (4), simplifies to

$$s = \boldsymbol{\tau} \times \boldsymbol{r}^T, \tag{42}$$

and we recall that $\boldsymbol{\tau}$ is symmetric. In view of this the polar decomposition theorem can be used to write a given s in the form (42). But since $\boldsymbol{\tau}$ is sign-indefinite there are four such polar decompositions (disregarding the singular situation (41) in which there are infinitely many). The corresponding principal values of $\boldsymbol{\tau}$ are

$$(\tau_1, \tau_2, \tau_3), (\tau_1, -\tau_2, -\tau_3), (-\tau_1, \tau_2, -\tau_3), (-\tau_1, -\tau_2, \tau_3),$$

the signs of the τ_i's being determined by knowledge of the sign of det (s).

Each of the $\boldsymbol{\tau}$'s corresponds to a unique **u** in $\mathscr{D}_\mathbf{u}$ and hence s gives rise to four distinct $\boldsymbol{\alpha}$'s. There are four distinct branches of the inversion $\boldsymbol{\alpha}$(s) and correspondingly four distinct functions W_c(s), one for each branch.

Even where (41) holds the λ_i's are determined from the principal components τ_i. The singularity arises because of the indeterminacy of the orientation of the Lagrangean axes[17].

The important question we now raise is the following: can the inequality (36) be extended to apply on a larger domain than $\mathscr{D}_\alpha$, for configurations where $\tau_i + \tau_j \leq 0$, by placing restrictions on the components of $\dot{\alpha}$? More importantly, can the global inequality (15) hold on a larger domain in some restricted sense, with attendant consequences for the range of applicability of the extremum principles (31)?

Certainly such restrictions can be imposed. Indeed, Krawietz[26] has discussed the local inequality (16) for all $\boldsymbol{\eta} \equiv \dot{\boldsymbol{\alpha}} \times \boldsymbol{\alpha}^{-1}$ having real eigenvalues and subject to the restriction

$$\boldsymbol{\eta}'\boldsymbol{\eta}' \geq 0 \tag{43}$$

(in the present notation), where $\boldsymbol{\eta}'$ is the deviator of $\boldsymbol{\eta}$. Although (43) rules out, in particular, pure rigid rotations there appears to be no physical basis for this inequality in general. For isotropic elastic solids, however, Krawietz does obtain certain sufficient conditions for (16) explicitly in terms of the λ_i's and τ_i's. These allow the domain of validity of (16) to be extended to situations in which $\tau_i + \tau_j < 0$, $i \neq j$.

Krawietz also discusses a restricted form of the global inequality (15) (for a fixed but arbitrary reference configuration) as a constitutive inequality. He proposes that (15) should hold for all $\boldsymbol{\alpha}$ and $\boldsymbol{\alpha}^*$ such that $\boldsymbol{\alpha}^* = \mathbf{p} \times \boldsymbol{\alpha}$ and where $\mathbf{p}$ (not equal to the identity) has *positive* eigenvalues. This generalizes the Coleman–Noll hypothesis in which $\mathbf{p}$ is required to be symmetric and positive definite. The restriction to $\mathbf{p}$ having real and positive eigenvalues ensures that in the polar decomposition of $\mathbf{p}$ the rotational part of $\mathbf{p}$ is limited to a subset of the proper orthogonal group, that subset depending on the magnitudes of the eigenvalues of the positive definite symmetric part of $\mathbf{p}$.

Whatever the mathematical merits of Krawietz's inequality, it has limited value when its possible application to extremum principles is considered. This is because the $\boldsymbol{\alpha}$ corresponding to an actual solution of the boundary-value problem is in general unknown, so that for any chosen $\boldsymbol{\alpha}^*$ it is not in general immediately obvious if $\mathbf{p}$ has the required form. We therefore need to analyse more carefully the global inequality (15) and its connection with (18).

Writing $\boldsymbol{\alpha} = \mathbf{r} \times \mathbf{u}$ and $\boldsymbol{\alpha}^* = \mathbf{r}^* \times \mathbf{u}^*$ and making use of the symmetry of $\mathbf{u}$ and $\mathbf{u}^*$ we obtain

$$W'(\boldsymbol{\alpha}^*) - W'(\boldsymbol{\alpha}) - \mathbf{s}(\boldsymbol{\alpha}^* - \boldsymbol{\alpha}) = W(\mathbf{u}^*) - W(\mathbf{u}) - \boldsymbol{\tau}(\mathbf{u}^* - \mathbf{u}) + (\mathbf{u}^* \times \boldsymbol{\tau})(\boldsymbol{\delta} - \mathbf{r}^T \times \mathbf{r}^*), \tag{44}$$

where $\boldsymbol{\delta}$ is the identity and the notation W' has been introduced temporarily in order to differentiate between W regarded as a function of $\boldsymbol{\alpha}$ and of $\mathbf{u}$.

Clearly (15) follows from (18) if $\mathbf{r}^* = \mathbf{r}$ and the extremum principles are then valid on the whole of $\mathscr{D}_\mathbf{u}$. However, this requires that we know the rotational part $\mathbf{r}$ of the deformation. In general $\mathbf{r}$ will not be known, but there are many situations of practical interest in which it is; for example, in problems with cylindrical or spherical symmetry and for problems in which the deformation is a pure strain ($\mathbf{r} = \boldsymbol{\delta}$). In this sense we have immediate extensions of the domain of validity of the extremum principles (31). And we can go further than this.

Note, first of all, that since $\mathbf{u}^* \neq \mathbf{u}$ is required for (18) it is no longer admissible to have $\boldsymbol{\alpha}^* = \mathbf{q} \times \boldsymbol{\alpha}$ (for orthogonal $\mathbf{q}$) and the restriction (40) is not therefore a consequence of (15).

In view of the fact that for $\mathbf{r}^* = \mathbf{r}$ the inequality (15) is valid over the whole of $\mathscr{D}_\mathbf{u}$ it would seem that when $\mathbf{r}^* \neq \mathbf{r}$ the range of validity of (15) can also be extended beyond $\mathscr{D}_\alpha$ (as has been shown by Krawietz). For this extension to be of practical value one would probably need to know the form of W explicitly in order to determine the relevant domain. It is possible in principle to find the minimum value of the quantity $(\mathbf{u}^* \times \boldsymbol{\tau})(\boldsymbol{\delta} - \mathbf{r}^T \times \mathbf{r}^*)$, from (44), with respect to $\mathbf{r}^T \times \mathbf{r}^*$ at fixed $\mathbf{u}^*$ and $\boldsymbol{\tau}$, and then find restrictions on the relations between $\mathbf{u}$ and $\mathbf{u}^*$ for (44) to be positive. Alternatively, it may be possible to restrict $\mathbf{r}^T \times \mathbf{r}^*$ in some way and then find conditions sufficient for (44). This is reasonable since, although $\mathbf{r}$ is in general unknown, there are many boundary-value problems for which an educated estimate of its value can be given. In many deformations associated with experimental tests this is certainly the case.

It is not our intention at this stage to pursue the discussion of the extended range of validity of the inequality (15). We merely emphasise that with care an extension of the range of validity to the whole elastic domain is possible. Clearly, more can be learned by further analysis, both in general and for specific boundary-value problems. In Part II the range of validity for certain boundary-value problems is examined in detail.

We now turn briefly to the dual inequalities (21) and (22). The left-hand side of (22) can be rearranged as

$$W_c'(\mathbf{s}^*) - W_c'(\mathbf{s}) - \boldsymbol{\alpha}(\mathbf{s}^* - \mathbf{s}) = W_c(\boldsymbol{\tau}^*) - W_c(\boldsymbol{\tau}) - \mathbf{u}(\boldsymbol{\tau}^* - \boldsymbol{\tau}) + (\mathbf{u} \times \boldsymbol{\tau}^*)(\boldsymbol{\delta} - \mathbf{r}^{*T} \times \mathbf{r}), \tag{45}$$

where $W_c'(\mathbf{s}) = \mathbf{s}\boldsymbol{\alpha} - W'(\boldsymbol{\alpha})$. The form of $W_c'(\mathbf{s})$ depends on which branch of the inversion $\boldsymbol{\alpha}(\mathbf{s})$ is taken. Which branch is the relevant one in a particular situation can be determined by the nature of the problem in question.

When $\mathbf{r}^* = \mathbf{r}$ (22) follows from (21) and remarks similar to those following (44) are relevant in the case of (45), with the proviso that those s's for which $\partial\mathbf{s}/\partial\boldsymbol{\alpha}$ is singular are not considered (for isotropic solids we are therefore restricted to those s's for which $\mathbf{s} \times \mathbf{s}^T$ has distinct principal values). It is not clear at present how the difficulty of the singularity can be overcome in general but, fortunately, situations in which (41) hold (in the case of isotropy) are of limited practical interest (except for homogeneous materials).

In this paper we are concerned with *inhomogeneous* solids. In particular, we are interested in determining bounds on the overall constitutive properties of composites rather than in boundary-value problems *per se*. Thus, with the aim of achieving such bounds over as wide a range of deformations as possible we can choose appropriate boundary conditions and test-specimen shapes, thereby confining attention to situations in which the extremum principles are valid.

In what follows we therefore assume that $\boldsymbol{\alpha}^*$ and $\mathbf{s}^*$ are *admissible*, by which is meant that, for any $\boldsymbol{\alpha}$ or s, $\boldsymbol{\alpha}^*$ and $\mathbf{s}^*$ are such that the extremum principles are valid.

4. APPLICATION TO COMPOSITES

4.1 *The overall constitutive law*

Following the notation of Ogden[21] and the analysis of Hill[19] we introduce the macroscopic (overall) variable $\bar{\boldsymbol{\alpha}}$ defined as the mean value of $\boldsymbol{\alpha}$ over the undeformed volume V of the material. Thus,

$$\bar{\boldsymbol{\alpha}} = V^{-1} \int_V \boldsymbol{\alpha}\, \mathrm{d}V$$

with $\bar{\mathbf{s}}$ and $\bar{W}$ similarly defined. Unless stated otherwise a quantity with a bar over denotes the volume average of that quantity over V.

In component form we have

$$\bar{\alpha}_{i\mu} = V^{-1} \int_\Sigma x_i n_\mu \, \mathrm{d}\Sigma, \qquad \bar{s}_{\mu i} = V^{-1} \int_\Sigma X_\mu t_i \, \mathrm{d}\Sigma \tag{46}$$

when $\boldsymbol{\alpha}$ is a deformation gradient and s satisfies (2) ([19], Section 3).

Amongst other decompositions Hill[19] has established

$$\overline{\mathbf{s}\boldsymbol{\alpha}} = \bar{\mathbf{s}}\bar{\boldsymbol{\alpha}}, \qquad \overline{\mathbf{s}\dot{\boldsymbol{\alpha}}} = \bar{\mathbf{s}}\dot{\bar{\boldsymbol{\alpha}}}, \qquad \overline{\dot{\mathbf{s}}\boldsymbol{\alpha}} = \dot{\bar{\mathbf{s}}}\bar{\boldsymbol{\alpha}}, \qquad \overline{\dot{\mathbf{s}}\dot{\boldsymbol{\alpha}}} = \dot{\bar{\mathbf{s}}}\dot{\bar{\boldsymbol{\alpha}}} \tag{47}$$

under certain loading conditions, but $\boldsymbol{\alpha}$ and s are not necessarily related through the constitutive law for (47) to be valid. It is merely required that $\boldsymbol{\alpha}$ be a deformation gradient and s be self equilibrated. (The superposed dot represents the material time derivative or an increment in the quantity concerned).

There follows the existence of a macropotential, denoted $\hat{W}$, which is the volume average of W[20, 21]. Thus with the help of $(47)_2$

$$\hat{W}(\bar{\boldsymbol{\alpha}}) = \overline{W(\boldsymbol{\alpha})}, \qquad \dot{\hat{W}} = \dot{\bar{W}} = \overline{\mathbf{s}\dot{\boldsymbol{\alpha}}} = \bar{\mathbf{s}}\dot{\bar{\boldsymbol{\alpha}}}$$

and

$$\bar{\mathbf{s}} = \frac{\partial \hat{W}}{\partial \bar{\boldsymbol{\alpha}}}, \tag{48}$$

$\hat{W}$ depending only on $\bar{\boldsymbol{\alpha}}$ (the notation $\hat{W}$ replaces the $\bar{W}$ used by Ogden[21]).

If the boundary data are macroscopically uniform it is reasonable to suppose that the material properties of the "overall material" are independent of this data so that the macroscopic elastic response of the material is characterized by $\hat{W}$ with (48).

Hill ([19], p. 140) has also noted that

$$\overline{\boldsymbol{\alpha} \times \mathbf{s}} = \bar{\boldsymbol{\alpha}} \times \bar{\mathbf{s}} \tag{49}$$

is symmetric so that the local condition for rotational balance is transmitted to the macro-level.

We now write $\bar{\boldsymbol{\alpha}} = \hat{\mathbf{r}} \times \hat{\mathbf{u}}$ where $\hat{\mathbf{r}}$ is proper orthogonal and $\hat{\mathbf{u}}$ is positive definite and symmetric. These are the macro-variables corresponding to $\mathbf{r}$ and $\mathbf{u}$ but, in general, $\hat{\mathbf{r}} \neq \bar{\mathbf{r}}$ and $\hat{\mathbf{u}} \neq \bar{\mathbf{u}}$. In elasticity theory rotational balance is equivalent to frame-indifference of the constitutive law so that, in view of (49), the macro-stress-deformation relation can be written

$$\hat{\boldsymbol{\tau}} = \frac{\partial \hat{W}}{\partial \hat{\mathbf{u}}}, \tag{50}$$

$\hat{W}$ being a function only of $\hat{\mathbf{u}}$ (that is depending on $\bar{\boldsymbol{\alpha}}$ only through $\bar{\boldsymbol{\alpha}}^T \times \bar{\boldsymbol{\alpha}}$) by analogy with (3). Moreover, the symmetric $\hat{\boldsymbol{\tau}}$ is the overall counterpart of $\boldsymbol{\tau}$, but in general $\hat{\boldsymbol{\tau}} \neq \bar{\boldsymbol{\tau}}$.

The volume average of (6) is now written

$$\hat{W} + \hat{W}_c = \bar{\mathbf{s}}\bar{\boldsymbol{\alpha}} \tag{51}$$

with the help of $(47)_1$, where

$$\hat{W}_c(\bar{\mathbf{s}}) = \overline{W_c(\mathbf{s})}$$

and hence, by analogy with (7),

$$\bar{\boldsymbol{\alpha}} = \frac{\partial \hat{W}_c}{\partial \bar{\mathbf{s}}}. \tag{52}$$

Equation (51) could also be written

$$\hat{W} + \hat{W}_c = \hat{\boldsymbol{\tau}}\hat{\mathbf{u}} \tag{53}$$

and $\hat{W}_c$ regarded as a function of $\hat{\boldsymbol{\tau}}$ only. The existence of $\hat{W}_c$ is guaranteed by the existence of the local complementary energy W_c. The inverse of (50) is

$$\hat{\mathbf{u}} = \frac{\partial \hat{W}_c}{\partial \hat{\boldsymbol{\tau}}}. \tag{54}$$

Returning now to the convexity inequalities we see that on taking the volume averages of (15) and (22) respectively we obtain

$$V^{-1} \int_V W(\boldsymbol{\alpha}^*)\,\mathrm{d}V - \hat{W}(\bar{\boldsymbol{\alpha}}) - \bar{\mathbf{s}}(\bar{\boldsymbol{\alpha}}^* - \bar{\boldsymbol{\alpha}}) > 0 \tag{55}$$

and

$$V^{-1} \int_V W_c(\mathbf{s}^*)\,\mathrm{d}V - \hat{W}_c(\bar{\mathbf{s}}) - \bar{\boldsymbol{\alpha}}(\bar{\mathbf{s}}^* - \bar{\mathbf{s}}) > 0 \tag{56}$$

with the help of $(47)_1$, where $\boldsymbol{\alpha}$ and $\mathbf{s}$ are associated with an actual solution of the boundary-value problem, $\boldsymbol{\alpha}^*$ is a deformation gradient and $\mathbf{s}^*$ is self-equilibrated.

From (55) and (56) one can deduce the overall global convexity of $\hat{W}(\bar{\boldsymbol{\alpha}})$ and $\hat{W}_c(\bar{\mathbf{s}})$. Thus (55) gives

$$\hat{W}(\bar{\boldsymbol{\alpha}}^*) - \hat{W}(\bar{\boldsymbol{\alpha}}) - \bar{\mathbf{s}}(\bar{\boldsymbol{\alpha}}^* - \bar{\boldsymbol{\alpha}}) > 0 \tag{57}$$

when $\bar{\alpha}^*$ ($\neq \bar{\alpha}$) is the average deformation gradient for some set of boundary data. Similarly, (56) gives

$$\hat{W}_c(\bar{s}^*) - \hat{W}_c(\bar{s}) - \bar{\alpha}(\bar{s}^* - \bar{s}) > 0 \tag{58}$$

when $\bar{s}^*$ ($\neq \bar{s}$) is the average nominal stress for some specified boundary data.

4.2 *Bounds on the overall energy and complementary energy*

(a) *The first boundary-value problem.* Suppose $\mathbf{x}$ is given everywhere on Σ so that $\Sigma_s = 0$. Then from (25), (28) or (55) we obtain

$$\hat{W}(\bar{\alpha}) \leq V^{-1} \int_V W(\alpha^*)\,\mathrm{d}V \tag{59}$$

amongst all admissible α^* derivable from an $\mathbf{x}^*(\mathbf{X})$ which satisfies the boundary condition (9) on the whole of Σ, and hence, by $(46)_1$, such that $\bar{\alpha}^* = \bar{\alpha}$. The equality holds at most when $\alpha^* = \alpha$ in V.

Similarly from (26), (30) or (56) with (51)

$$\hat{W}(\bar{\alpha}) \geq V^{-1} \int_\Sigma \mathbf{t}^*\mathbf{x}_0\,\mathrm{d}\Sigma - V^{-1} \int_V W_c(\mathbf{s}^*)\,\mathrm{d}V \tag{60}$$

amongst all admissible self-equilibrated $\mathbf{s}^*$, with equality at most when $\mathbf{s}^* = \mathbf{s}$ in V. Alternatively (60) can be put as

$$\hat{W}(\bar{\alpha}) \geq V^{-1} \int_V \{\mathbf{s}^*\alpha_0 - W_c(\mathbf{s}^*)\}\,\mathrm{d}V, \tag{61}$$

where α_0 is any deformation gradient compatible with the boundary data on Σ.

It is important to recognize that α^* in (59) and $\mathbf{s}^*$ in (60) and (61) are not in general related through the constitutive law but can be chosen independently subject to the prescribed conditions.

The difference between the upper and lower bounds on $\hat{W}(\bar{\alpha})$ is

$$V^{-1} \int_V \{W(\alpha^*) + W_c(\mathbf{s}^*) - \mathbf{s}^*\alpha_0\}\,\mathrm{d}V. \tag{62}$$

(b) *The second boundary-value problem.* Suppose now that $\mathbf{t}$ is prescribed on the whole of Σ. Then (26), (30) or (56) gives

$$\hat{W}_c(\bar{s}) \leq V^{-1} \int_V W_c(\mathbf{s}^*)\,\mathrm{d}V \tag{63}$$

within the class of admissible self-equilibrated $\mathbf{s}^*$ consistent with the boundary condition (10) on Σ, and therefore, by $(46)_2$, such that $\bar{s}^* = \bar{s}$.

Equation (25), (28) or (55), with the help of (51), yields

$$\hat{W}_c(\bar{s}) \geq V^{-1} \int_\Sigma \mathbf{x}^*\mathbf{t}_0\,\mathrm{d}\Sigma - V^{-1} \int_V W(\alpha^*)\,\mathrm{d}V \tag{64}$$

in the class of admissible functions $\mathbf{x}^*(\mathbf{X})$, with equality at most when $\alpha^* = \alpha$ in V.

The inequality (64) can also be written as

$$\hat{W}_c(\bar{s}) \geq V^{-1} \int_V \{\alpha^*\mathbf{s}_0 - W(\alpha^*)\}\,\mathrm{d}V \tag{65}$$

for any self-equilibrated $\mathbf{s}_0$ consistent with the boundary condition on Σ.

The difference between the bounds in (63) and (65) is

$$V^{-1}\int_V \{W(\boldsymbol{\alpha}^*)+W_c(\mathrm{s}^*)-\boldsymbol{\alpha}^*\mathrm{s}_0\}\,\mathrm{d}V \tag{66}$$

and again we emphasise that $\boldsymbol{\alpha}^*$ and s* are not in general related through the constitutive law.

In general the bounds on $\hat{W}(\bar{\boldsymbol{\alpha}})$ and $\hat{W}_c(\bar{\mathrm{s}})$ obtained from the two boundary-value problems are different, as are the expressions (62) and (66). Indeed, the admissible functions **x*** and s* in the two cases are different. For example, in (59) **x*** satisfies (9) on Σ while the **x*** in (65) is not so restricted. Correspondingly, the s* in (61) is not subject to any boundary constraint, whereas the s* in (63) satisfies (10) on Σ.

For the first and second boundary-value problems respectively we may choose $\boldsymbol{\alpha}_0=\boldsymbol{\alpha}^*$ in (62) and $\mathrm{s}_0=\mathrm{s}^*$ in (66). Each of (62) and (66) then becomes

$$V^{-1}\int_V \{W(\boldsymbol{\alpha}^*)+W_c(\mathrm{s}^*)-\mathrm{s}^*\boldsymbol{\alpha}^*\}\,\mathrm{d}V \tag{67}$$

although the admissible $\boldsymbol{\alpha}^*$ and s* are different in the two problems.

More generally, for the mixed boundary-value problem it can be shown immediately from (31), using the fact that **x*** satisfies (9) and s* satisfies (10), that the difference between the upper and lower bounds on $\hat{W}(\bar{\boldsymbol{\alpha}})$ or $\hat{W}_c(\bar{\mathrm{s}})$ can also be written as (67). *The problem of bounding* $\hat{W}(\bar{\boldsymbol{\alpha}})$ *and* $\hat{W}_c(\bar{\mathrm{s}})$ *as closely as desired is therefore reduced to that of minimising the expression* (67) *within the class of admissible* **x*** *and* s*.

Incidentally, even if the inequalities in (31) are reversed the difference between the bounds is still (67), which is then negative.

The inequalities (59), (61), (65) and (63) respectively are generalizations to finite elasticity of those described in the infinitesimal theory by Hill ([34], eqns 1.1–1.4).

4.3 *Generalized Voigt–Reuss bounds*

When the boundary condition is more specific, of the form

$$x_i=\bar{\alpha}_{i\mu}X_\mu \qquad \text{on } \Sigma,$$

we may take $\boldsymbol{\alpha}_0=\bar{\boldsymbol{\alpha}}$ in (61). As our comparison field we may choose $\boldsymbol{\alpha}^*=\bar{\boldsymbol{\alpha}}$ in V since this is compatible with the boundary condition. For an inhomogeneous material such a deformation field cannot of course correspond to an actual equilibrium configuration. We recall that we are restricting attention to admissible $\boldsymbol{\alpha}^*$ and therefore it must be assumed that $\boldsymbol{\alpha}^*=\bar{\boldsymbol{\alpha}}$ is admissible.

Inequalities (59) and (61) now become

$$V^{-1}\int_V \{\mathrm{s}^*\bar{\boldsymbol{\alpha}}-W_c(\mathrm{s}^*)\}\,\mathrm{d}V\le \hat{W}(\bar{\boldsymbol{\alpha}})\le V^{-1}\int_V W(\bar{\boldsymbol{\alpha}})\,\mathrm{d}V, \tag{68}$$

where s* is any admissible self-equilibrated stress field. In particular, we may choose s* to be any uniform field in V, which may or may not be equal to the actual mean $\bar{\mathrm{s}}$ associated with $\bar{\boldsymbol{\alpha}}$.

Dually, for the boundary condition

$$t_i=n_\mu s_{\mu i} \qquad \text{on } \Sigma$$

we may put $\mathrm{s}_0=\bar{\mathrm{s}}$ in (65), and choose $\mathrm{s}^*=\bar{\mathrm{s}}$ in V (assumed admissible). Inequalities (63) and (65) then become

$$V^{-1}\int_V \{\boldsymbol{\alpha}^*\bar{\mathrm{s}}-W(\boldsymbol{\alpha}^*)\}\,\mathrm{d}V\le \hat{W}_c(\bar{\mathrm{s}})\le V^{-1}\int_V W_c(\bar{\mathrm{s}})\,\mathrm{d}V \tag{69}$$

for any admissible **x*** whether or not $\boldsymbol{\alpha}^*=\bar{\boldsymbol{\alpha}}$, the actual mean of $\boldsymbol{\alpha}$ associated with the mean $\bar{\mathrm{s}}$.

The right hand inequalities in (68) and (69) respectively generalize to finite elasticity the Voigt and Reuss bounds on the strain energy of an elastic solid in the infinitesimal theory (in particular, bounds on the bulk and shear moduli of an isotropic elastic solid), as discussed by Hill[5] for two-phase composites. We recall that $W_c = W$ in linear elasticity provided $\boldsymbol{\alpha}$ is interpreted as the *displacement* gradient.

The left-hand inequalities in (68) and (69) reduce to the right-hand ones in (69) and (68) respectively if $\mathbf{s}^* = \bar{\mathbf{s}}$ and $\boldsymbol{\alpha}^* = \bar{\boldsymbol{\alpha}}$, as can be shown with the help of (51). For a two-phase composite with volume concentrations c_1 and c_2 the right-hand sides of (68) and (69) respectively give

$$\hat{W}(\bar{\boldsymbol{\alpha}}) \leq c_1 W_1(\bar{\boldsymbol{\alpha}}) + c_2 W_2(\bar{\boldsymbol{\alpha}}) \tag{70}$$

and

$$\hat{W}_c(\bar{\mathbf{s}}) \leq c_1 W_{1c}(\bar{\mathbf{s}}) + c_2 W_{2c}(\bar{\mathbf{s}}), \tag{71}$$

where W_1 and W_2 are the strain-energy functions of the (homogeneous) constituent phases.

In the context of second-order elasticity and for particular material symmetries Barsch[35] has obtained estimates for the overall moduli corresponding to those of Voigt, Reuss and Hill in the linear theory. To my knowledge there is no other work in the literature concerned with problems of this kind in the non-linear context, other than for porous materials (see [21] and the refs. therein).

5. STRENGTHENING AND WEAKENING THEOREMS

5.1 *Strengthening*

Hill's strengthening theorem ([5], p. 370) has an immediate generalization to finite elasticity on the basis of the extremum principles of Section 3. This is now discussed.

Firstly we consider an elastic solid (homogeneous or inhomogeneous) with strain-energy function W. It occupies volume V in its undeformed configuration and is subject to the boundary conditions (9) and (10). A change in the constitution of the material is then made so that its strain-energy becomes W^+, the original boundary conditions being maintained on Σ (which is unchanged).

Let $\boldsymbol{\alpha}$ and $\boldsymbol{\alpha}^+$ respectively be the deformation gradients in the W and W^+ materials. Following Hill[5] the material is said to be strengthened in the transition if

$$W(\boldsymbol{\alpha}^+) \leqslant W^+(\boldsymbol{\alpha}^+) \tag{72}$$

with strict inequality holding somewhere in V. This just states that the energy of the stronger material is larger than that of the weaker one *at the same deformation.*

When $(\mathbf{x}, \mathbf{s})$ and $(\mathbf{x}^+, \mathbf{s}^+)$ correspond to the actual solutions of the general mixed boundary-value problem (for the W and W^+ materials respectively) the theorem of minimum energy (28) gives

$$E(\mathbf{x}) \leqslant E(\mathbf{x}^+), \qquad E^+(\mathbf{x}^+) \leq E^+(\mathbf{x})$$

when applied to each of the equilibrium configurations. (It is assumed that $\mathbf{x}^+$ and $\mathbf{x}$ are such that $\boldsymbol{\alpha}^+$ and $\boldsymbol{\alpha}$ are admissible for the validity of (28) in respect of W and W^+ respectively.)

Use of (72) then enables a further inequality to be inserted, so that

$$E(\mathbf{x}) \leq E(\mathbf{x}^+) \leq E^+(\mathbf{x}^+) \leq E^+(\mathbf{x}) \tag{73}$$

and, in particular,

$$E(\mathbf{x}) \leq E^+(\mathbf{x}^+). \tag{74}$$

Thus, the total-energy functional is increased when the material is strengthened (see Walpole[22] for a discussion of the corresponding inequality in the infinitesimal theory). Note that in order to establish (74) the final inequality in (73) is not necessary.

By means of (14) we may rewrite (74) as

$$E_c(\mathbf{s}) \le E_c^+(\mathbf{s}^+). \tag{75}$$

This prompts consideration of a possible definition of strengthening in terms of the complementary energy. One candidate for this (by analogy with (72)) is $W_c(\mathbf{s}^+) \ge W_c^+(\mathbf{s}^+)$, but this leads to $E_c(\mathbf{s}) \ge E_c(\mathbf{s}^+) \le E_c^+(\mathbf{s}^+) \ge E_c^+(\mathbf{s})$ and, in particular, does not establish (75). A variant of this, not equivalent to (72) in general, is

$$W_c(\mathbf{s}) \ge W_c^+(\mathbf{s}). \tag{76}$$

This leads to the chain of inequalities

$$E_c(\mathbf{s}^+) \le E_c(\mathbf{s}) \le E_c^+(\mathbf{s}) \le E_c^+(\mathbf{s}^+) \tag{77}$$

on use of the theorem of maximum complementary energy (30) with $\mathbf{s}^+$ and $\mathbf{s}$ as admissible fields for (30) to be valid in respect of W_c and W_c^+ respectively. In particular, (77) includes (75) and therefore (74) as required. The first inequality in (77) is not necessary in order to establish this result.

Although the two definitions of strengthening (72) and (76) are not in general equivalent they both imply that the total energy is increased when the material is strengthened without relaxing the boundary conditions.

The inequality (76) merely states that the complementary energy of the original material is greater than that of the strengthened material *at the same stress*.

Using the definition of the overall constitutive law introduced in Section 4.1 we may rewrite (74) and (75) as

$$\hat{W}(\bar{\boldsymbol{\alpha}}) \le \hat{W}^+(\bar{\boldsymbol{\alpha}}^+) + V^{-1}\int_\Sigma (\mathbf{x}-\mathbf{x}^+)\mathbf{t}\,\mathrm{d}\Sigma \tag{78}$$

and

$$\hat{W}_c(\bar{\mathbf{s}}) \ge \hat{W}_c^+(\bar{\mathbf{s}}^+) + V^{-1}\int_\Sigma (\mathbf{t}-\mathbf{t}^+)\mathbf{x}\,\mathrm{d}\Sigma \tag{79}$$

respectively. In (78) $\mathbf{t}$ may be replaced by $\mathbf{t}^+$, and in (79) $\mathbf{x}$ by $\mathbf{x}^+$.

For the first boundary-value problem (78) and (79) each give

$$\hat{W}(\bar{\boldsymbol{\alpha}}) \le \hat{W}^+(\bar{\boldsymbol{\alpha}}^+), \tag{80}$$

where $\bar{\boldsymbol{\alpha}}^+ = \bar{\boldsymbol{\alpha}}$ by (46) and use has been made of (51). For the second boundary-value problem they each yield

$$\hat{W}_c(\bar{\mathbf{s}}) \ge \hat{W}_c^+(\bar{\mathbf{s}}^+), \tag{81}$$

where $\bar{\mathbf{s}}^+ = \bar{\mathbf{s}}$.

For strengthening, therefore, the overall energy is increased when the boundary is fixed, and the overall complementary energy is decreased under fixed boundary tractions. In the infinitesimal theory, since $W_c = W$, (81) states that the total energy is decreased under fixed traction[5, 22].

If the whole class of possible equilibrium fields is considered $\boldsymbol{\alpha}^+$ in (72) and $\mathbf{s}$ in (76) may be regarded as arbitrary, the material being strengthened for all modes of deformation; and then (72) and (76) can be shown to be equivalent on the basis of the convexity inequalities (15) and (22). Then, since $\bar{\boldsymbol{\alpha}}^+ = \bar{\boldsymbol{\alpha}}$ and $\bar{\mathbf{s}}^+ = \bar{\mathbf{s}}$, (80) and (81) are direct overall analogues of (72) and (76).

More generally, for the mixed boundary-value problem (all possible boundary data being considered) it is easily shown from (78) and (79), with the help of (57), (58), (51) and the divergence theorem, that

$$\hat{W}(\bar{\boldsymbol{\alpha}}) \le \hat{W}^+(\bar{\boldsymbol{\alpha}}) \tag{82}$$

and, equivalently,

$$\hat{W}_c(\bar{s}^+) \geq \hat{W}_c^+(\bar{s}^+) \tag{83}$$

for all admissible $\bar{\alpha}$ and $\bar{s}^+$. The overall characterization of strengthening (82) (the direct overall analogue of (72)) is equivalent to (74) and (78) on the basis of (57) applied to $\hat{W}^+$.

It is important to remark that (72) and (76) are frame-indifferent and can therefore be expressed as

$$W(\mathbf{u}^*) \leq W^+(\mathbf{u}^*), \qquad W_c(\boldsymbol{\tau}^*) \geq W_c^+(\boldsymbol{\tau}^*) \tag{84}$$

where $\mathbf{u}^*$ is now arbitrary, symmetric and positive definite and $\boldsymbol{\tau}^*$ is arbitrary and symmetric. On the assumption that (18) holds it is easily shown, by means of (18) and (21), that $(84)_1$ and $(84)_2$ are equivalent and hence *there is no need to distinguish* (76) *from* (72) *even when* (15) *and* (22) *fail*. At the overall level $\hat{W}$ and $\hat{W}_c$ are frame-indifferent (as remarked in Section 4.1) and hence the overall analogues of (84), namely (82) and (83), could be expressed in terms of the variables $\hat{\mathbf{u}}$ and $\hat{\boldsymbol{\tau}}^+$.

The definition of local strengthening used here is geometry independent and local strengthening is a notion *intrinsic* to the material. In view of the above remarks about frame indifference it might appear that the derived overall characterization of strengthening, as portrayed by (82), is also geometry independent. However, (82) is arrived at only after certain assumptions of admissibility are made for the validity of (74). Failure of (74) may in essence be interpreted as geometry dependent instability so the transition from (72) to (82) involves geometrical considerations which enter via (15) from the boundary data. It is worth remarking at this point that for the first boundary-value problem (when the material is homogeneous and homogeneously deformed) the question of stability is independent of the constitutive law and shape of the body ([14], Section 68b; [36], Section 3). On the other hand, for the traction boundary-value problem the shape is critical.

Thus, in general, (82) is geometry dependent and does not always follow if the material is strengthened in the local sense. Nevertheless, (82) serves as a characterization of overall strengthening without reference to the local definition, and certain instabilities are ruled out when the material is strengthened in this overall sense.

We have assumed that the overall constitutive law $\hat{W}(\bar{\alpha})$ is independent of the boundary data. This, however, may not be a valid assumption for all inhomogeneous solids, in which case the geometry dependence of (82) is enhanced.

In the linear theory, of course, questions of instability and inadmissibility do not arise and the transition from the local to the overall definition of strengthening is straightforward. It is worth remarking that although a material may be strengthened in the overall sense, it is not necessarily strengthened locally at every point.

An immediate application of these theorems is in the examination of the relation between the properties of a homogeneous material and those of a two-phase composite which consists of a matrix of the original material and inclusions of a stronger material.

For any given boundary-value problem of the type we are considering let $(\boldsymbol{\alpha}, \mathbf{s})$ be a solution in respect of the homogeneous material and $(\boldsymbol{\alpha}^+, \mathbf{s}^+)$ for the strengthened material. We suppose that $(\boldsymbol{\alpha}, \mathbf{s})$ is known but $(\boldsymbol{\alpha}^+, \mathbf{s}^+)$ is not. Then, for the first boundary-value problem, or when $\mathbf{t}_0 = \mathbf{0}$ on Σ_s, the inequalities (73) yield

$$\int_V W(\boldsymbol{\alpha})\,\mathrm{d}V \leq \int_V W^+(\boldsymbol{\alpha}^+)\,\mathrm{d}V \leq \int_V W^+(\boldsymbol{\alpha})\,\mathrm{d}V. \tag{85}$$

If W_1 and W_2 are the strain-energy functions for the matrix and inclusions respectively (85) can be replaced by

$$\int_V W_1(\boldsymbol{\alpha})\,\mathrm{d}V \leq \int_V W^+(\boldsymbol{\alpha}^+)\,\mathrm{d}V \leq \int_{V_1} W_1(\boldsymbol{\alpha})\,\mathrm{d}V + \int_{V_2} W_2(\boldsymbol{\alpha})\,\mathrm{d}V, \tag{86}$$

where V_1 and V_2 are the volumes of the constituents. The right-hand inequality in (86) reduces to (70) for the first boundary-value problem only if the deformation $\boldsymbol{\alpha}$ is homogeneous.

The inequalities (86) provide bounds on the overall energy of the composite in terms of the known quantities W_1, W_2 and $\boldsymbol{\alpha}$, but, of course, the left-hand inequality is trivial, coming directly from the definition of strengthening. The right-hand inequality could also be obtained from (59) by putting $\boldsymbol{\alpha}^* = \boldsymbol{\alpha}$. Furthermore, from (60) with $\mathrm{s}^* = \mathrm{s}$, the left-hand bound in (86) can be replaced by the larger quantity

$$\int_{V_1} W_1(\boldsymbol{\alpha})\,\mathrm{d}V + \int_{V_2} \{\mathrm{s}\boldsymbol{\alpha} - W_{2c}(\mathrm{s})\}\,\mathrm{d}V. \tag{87}$$

For the second boundary-value problem, or when $\mathbf{x}_0 = \mathbf{0}$ on Σ_x, the corresponding inequalities for the overall complementary energy are obtained from (63) and (64) with $\mathrm{s}^* = \mathrm{s}$ and $\boldsymbol{\alpha}^* = \boldsymbol{\alpha}$ in the form

$$\int_{V_1} W_{1c}(\mathrm{s})\,\mathrm{d}V + \int_{V_2} \{\mathrm{s}\boldsymbol{\alpha} - W_2(\boldsymbol{\alpha})\}\,\mathrm{d}V \le \int_V W_c^+(\mathrm{s}^+)\,\mathrm{d}V \le \int_{V_1} W_{1c}(\mathrm{s})\,\mathrm{d}V + \int_{V_2} W_{2c}(\mathrm{s})\,\mathrm{d}V, \tag{88}$$

these bounds again being in terms of known quantities.

The bounds given in (86)–(88) can be improved by better choices of the test functions $\boldsymbol{\alpha}^*$ and s^* in (59), (60), (63) and (64). We note finally that the right hand inequality in (88) reduces to (71) when s is homogeneous.

5.2 *Weakening*

As remarked by Walpole [22] a corresponding weakening theorem can be given in the linear theory. It is obtained by reversing the inequality in (72). The same applies in the non-linear theory, and on this basis, with the help of the extremum principle (28), the chain of inequalities (73) is replaced by

$$E(\mathbf{x}^+) \ge E(\mathbf{x}) \ge E^+(\mathbf{x}) \ge E^+(\mathbf{x}^+).$$

In particular (74), and therefore (75), is reversed.

In consequence, weakening implies that (80) and (81) respectively are reversed for the first and second boundary-value problems.

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