Eur. J. Mech., A/Solids, **11**, n° 4, 447-466, 1992

A constitutive inequality for hyperelastic materials in finite strain

J. B. LEBLOND *

ABSTRACT. — The inequality $\check{\sigma}:\mathbf{D}>0$ ($\check{\sigma}$: Jaumann derivative of the Cauchy stress tensor; $\mathbf{D}$: Eulerian strain rate), which is a variant of Hill's "H_0 *inequality*", appears as an *a priori* viable answer to the "*Hauptproblem*", proposed by Truesdell in 1956, of formulating a constitutive inequality for finitely strained hyperelastic bodies; indeed it verifies the essential conditions of reducing to the usual restrictions $\mu>0$, $3\lambda+2\mu>0$ for an isotropic elastic solid in the natural (stress-free) state, and $dp/d\rho>0$ for a perfect (non-viscous) fluid. The aim of this paper is to confirm that this inequality is a possible solution, by considering various examples involving materials with or without internal constraints and checking that it yields reasonable restrictions. Connections with other inequalities are also investigated with special emphasis on Hill's H_0 inequality, and also on Ball's "*polyconvexity*" property because of its mathematical importance.

1. Introduction

The problem of finding a "reasonable" constitutive inequality for hyperelastic bodies in finite strain was first considered by [Truesdell, 1956]. (In fact Truesdell formulated the problem more generally for *elastic* materials, which differ from hyperelastic materials in that there may not exist a stored-energy function; only hyperelastic bodies will be considered here.) This question arises in two contexts. The first one is the search for phenomenological models of hyperelastic media; the role of the constitutive inequality is then to place restrictions upon the envisageable stored-energy functions (*see* e. g. [Ogden, 1972 *a*]). The second one is the investigation of such mathematical questions as the existence of solutions; the inequality serves then as a hypothesis needed for the derivation of certain theorems (*see* e. g. [Ball, 1977]).

Two essential criteria for appreciating the interest of a proposed inequality, used notably by [Wang & Truesdell, 1973], are as follows:

- It should reduce to the usual restrictions $\mu>0$, $3\lambda+2\mu>0$ (λ and μ being the Lamé coefficients) in the case of an isotropic elastic solid in the natural (stress-free) state.
- It should also reduce to the equally classical condition $dp/d\rho>0$ (where p and ρ are the pressure and mass per unit volume) in the case of a perfect (non-viscous) fluid.

Another, vaguer but equally important requirement is that it should yield "reasonable" restrictions in some simple cases, like that of the Neo-Hookean material for instance.

---

* Laboratoire de Modélisation en Mécanique, Université Pierre et Marie Curie, Tour n° 66, 4, place Jussieu, 75005 Paris, France.

EUROPEAN JOURNAL OF MECHANICS, A/SOLIDS, VOL. 11, N° 4, 1992
0997-7538/92/04 447 20/$ 4.00/

Among all inequalities proposed so far, the most important ones are probably the Coleman-Noll inequality, the Legendre-Hadamard strong ellipticity condition, Hill's inequalities and Ball's *"polyconvexity"* property. The first one [Coleman & Noll, 1959] states that

$$[\mathscr{B}(\mathbf{F}^*)-\mathscr{B}(\mathbf{F})]:[\mathbf{F}^{*\,\mathrm{T}}-\mathbf{F}^{\mathrm{T}}]>0 \tag{1}$$

In this inequality $\mathbf{F}$ is the gradient of the transformation, $\mathscr{B}=\mathrm{J}\,\boldsymbol{\sigma}\,.\,(\mathbf{F}^{\mathrm{T}})^{-1}$ (J: determinant of $\mathbf{F}$; $\boldsymbol{\sigma}$: Cauchy stress tensor) the Boussinesq, or Piola-Lagrange, or first Piola-Kirchhoff stress tensor, and $\mathbf{F}^*$ a tensor differing from $\mathbf{F}$ by a pure deformation: $\mathbf{F}^*=\mathbf{S}\,.\,\mathbf{F}$, where $\mathbf{S}$ is symmetric, positive-definite and not equal to $\mathbf{1}$. The corresponding (slightly stronger) differential inequality is

$$(\mathbf{F}^{\mathrm{T}}\,.\,\mathbf{D}):\frac{\partial^2\psi}{\partial\mathbf{F}\,\partial\mathbf{F}}:(\mathbf{F}^{\mathrm{T}}\,.\,\mathbf{D})>0 \tag{2}$$

for all non-zero Eulerian strain rates $\mathbf{D}=(1/2)\,[\dot{\mathbf{F}}\,.\,\mathbf{F}^{-1}+(\mathbf{F}^{-1})^{\mathrm{T}}\,.\,\dot{\mathbf{F}}^{\mathrm{T}}]$, where ψ is the stored-energy per unit volume of the reference configuration. This inequality does reduce to the conditions $\mu>0$, $3\lambda+2\mu>0$ for an isotropic solid in the natural state; however it yields $dp/d\rho>2p/3\rho$ in the case of a perfect fluid (*see* [W & T, 1973]), which is not true near the critical point of a fluid capable of phase change. [Ogden, 1970] and [Sidoroff, 1974] also showed that inequality (2) yields unsatisfactory consequences for some compressible solid materials. (A similar argument involving incompressible materials was presented by [Hill, 1968] but, as remarked by [W & T, 1973], the reasoning was based on a debatable interpretation of inequality (2) in the case of materials subject to internal constraints.)

The Legendre-Hadamard strong ellipticity condition states that

$$(\mathbf{U}\otimes\mathbf{v}):\frac{\partial^2\psi}{\partial\mathbf{F}\,\partial\mathbf{F}}:(\mathbf{U}\otimes\mathbf{v})>0 \tag{3}$$

for all non-zero vectors $\mathbf{U}$, $\mathbf{v}$. It has often been considered as a constitutive restriction (*see* e.g. [Antman, 1983] and [B, 1977]) but is too weak to be acceptable as an answer to the *"Hauptproblem"*; indeed it reduces to $\mu>0$, $\lambda+2\mu>0$ (instead of $3\lambda+2\mu>0$) for an isotropic elastic solid in the natural state [W & T, 1973].

The Hill inequalities [Hill, 1968, 1970] are defined as follows. Let $\boldsymbol{\tau}=\mathrm{J}\,\boldsymbol{\sigma}$ denote the Kirchhoff stress tensor, ˘ the Jaumann derivative, m a real parameter and $\mathscr{D}_m\boldsymbol{\tau}/\mathscr{D}_m t$ the objective stress-rate given by

$$\frac{\mathscr{D}_m\boldsymbol{\tau}}{\mathscr{D}_m t}=\breve{\boldsymbol{\tau}}-m\,(\boldsymbol{\tau}\,.\,\mathbf{D}+\mathbf{D}\,.\,\boldsymbol{\tau}). \tag{4}$$

Inequality H_m then stipulates that

$$\frac{\mathscr{D}_m\boldsymbol{\tau}}{\mathscr{D}_m t}:\mathbf{D}>0 \tag{5}$$

for all non-zero strain rates. Hill also introduced a one-parameter family of strain measures $\mathbf{E}_m$ in the following way: v_1, v_2, v_3 being the square roots of the eigenvalues of the tensor $\mathbf{C}=\mathbf{F}^T . \mathbf{F}$ (sometimes called the *"singular values"* of $\mathbf{F}$), $\mathbf{E}_m$ has the same eigenvectors as $\mathbf{C}$ and eigenvalues given by $f_m(v_1), f_m(v_2), f_m(v_3)$ where

$$f_m(v) \equiv \frac{v^{2m}-1}{2m} \quad \text{if } m \neq 0, \qquad \ln v \quad \text{if } m=0. \tag{6}$$

He then showed that H_m is equivalent to demanding that the Hessian form of ψ with respect to $\mathbf{E}_m$ be positive-definite at the reference configuration, whatever the choice of the latter.

For $m=1/2$, H_m is identical to the strengthened version (2) of the Coleman-Noll inequality [H, 1970]. Hill's recommendation, however, was to adopt the value $m=0$. His reasoning was based on the consideration of incompressible bodies. Since for such constrained materials, an indeterminate Lagrange multiplier is involved in the expression(s) of the stress tensor(s), two interpretations of the constitutive inequalities are possible according to whether one considers that the latter apply to the whole stress tensor(s) or only to that part which is independent of the Lagrange multiplier. Hill adopted the first interpretation and showed then that for an incompressible material, inequality H_m places a restriction upon the Lagrange multiplier, except for the value $m=0$. Since this multiplier then represents a hydrostatic pressure or tension for the Cauchy stress tensor and typical incompressible materials like rubber are observed to exist in nature without any apparent limitation on the trace of this tensor, such a restriction is undesirable. This favours the choice $m=0$.

Hill's conclusion was criticized by [W and T, 1973] on the following grounds. First, his reasoning does not apply if one adopts the second interpretation mentioned above, no restriction being then placed, by definition, on the Lagrange multiplier. Second, and more importantly, whereas inequality H_m reduces to $\mu>0$, $3\lambda+2\mu>0$ for an isotropic solid in the natural state, as desired, it yields

$$mp>0; \qquad \frac{dp}{d\rho} > \left(1-\frac{2m}{3}\right)\frac{p}{\rho} \tag{7}$$

for a perfect fluid [H, 1968]. Hence H_0 implies that $dp/d\rho > p/\rho$, which is not satisfactory.

It should be remarked that according to Eq. (7), $m=3/2$ is the only value for which H_m yields $dp/d\rho>0$ for a perfect fluid (plus the restriction $p>0$, which is reasonable). In view of this, the author's opinion is that inequality $\mathrm{H}_{3/2}$ has received insufficient attention (in fact, it seems to have attracted *no* attention at all). It will be studied incidentally in this paper. Unfortunately, it will be seen to yield unsatisfactory consequences in some simple cases (other than an isotropic solid in the natural state and a perfect fluid).

Ball's *"polyconvexity"* condition [B, 1977] states that there exists a *convex* function $\Phi(\mathbf{A}, \mathbf{B}, \delta)$ such that

$$\psi(\mathbf{F}) = \Phi(\mathbf{F}, \operatorname{adj}\mathbf{F}, \det\mathbf{F}) \tag{8}$$

where adj $\mathbf{F}$ denotes the adjoint of $\mathbf{F}\,[=(\det \mathbf{F})\,\mathbf{F}^{-1}$ since $\mathbf{F}$ is invertible]. (The function ψ itself cannot be convex with respect to $\mathbf{F}$, because this would contradict the objectivity principle: *see* e.g. [C and N, 1959].) This property is of considerable mathematical interest: indeed [B, 1977] showed that together with some additional assumptions, it implies the existence of a displacement field which minimizes the energy integral. It is not known, unfortunately, whether the corresponding stresses do satisfy the equilibrium equations, even in the weak sense. Still, this seems to be the first existence theorem in non-linear elasticity established under *realistic* hypotheses.

With respect to the two criteria mentioned above, the situation is the following. As shown in Appendix A, the polyconvexity property "almost" reduces to the condition $dp/d\rho>0$ for a perfect fluid in the sense that

$$(9)\qquad \frac{dp}{d\rho}>0 \;\Rightarrow\; \text{polyconvexity} \;\Rightarrow\; \frac{dp}{d\rho}\geqq 0.$$

On the other hand, it is shown in Appendix B that for an isotropic solid in the natural state,

$$(10)\qquad \mu>0,\quad \lambda+2\mu>0 \;\Rightarrow\; \text{polyconvexity} \;\Rightarrow\; \mu\geqq 0,\quad \lambda+2\mu\geqq 0.$$

The polyconvexity condition is therefore too weak in this case to be acceptable as a solution to the *"Hauptproblem"*. This conclusion is also supported by the consideration of some other examples, as will be seen below.

The aim of this paper is to study the inequality

$$(11)\qquad \check{\boldsymbol{\sigma}}:\mathbf{D}>0$$

(for all non-zero $\mathbf{D}$'s) where $\check{}$ denotes the Jaumann derivative as above. It is a variant of Hill's inequality H_0 [*see* Eqs. (4), (5)]; in particular the two are equivalent in the incompressible case ($\mathrm{J}=\mathrm{Const.}$). Inequality (11) has been considered elsewhere in the context of plasticity theory, but does not seem to have been proposed as a constitutive restriction for hyperelastic materials. It satisfies the two criteria mentioned above and thus appears as an *a priori* viable proposition. Indeed, for a perfect fluid, $\boldsymbol{\sigma}=-p\,\mathbf{1}$, $\check{\boldsymbol{\sigma}}=-\dot{p}\,\mathbf{1}$ and $\check{\boldsymbol{\sigma}}:\mathbf{D}=-\dot{p}\,\dot{\mathrm{J}}/\mathrm{J}=\dot{p}\,\dot{\rho}/\rho$; hence inequality (11) is equivalent to $dp/d\rho>0$ (provided it is exceptionally required to be strict *only for those* $\mathbf{D}$*'s which have a non-zero trace*; this restriction is natural since a perfect fluid offers a resistance only to solicitations involving a volume change). For an isotropic solid in the natural state, one gets, taking the latter as the reference configuration, $\check{\boldsymbol{\sigma}}:\mathbf{D}\equiv\dot{\boldsymbol{\sigma}}:\dot{\boldsymbol{\varepsilon}}\equiv\dot{\boldsymbol{\varepsilon}}:(\partial^2\psi/\partial\boldsymbol{\varepsilon}\,\partial\boldsymbol{\varepsilon}):\dot{\boldsymbol{\varepsilon}}$ where $\boldsymbol{\varepsilon}$ denotes the linearized strain; hence (11) is equivalent to the requirement that the Hessian form of ψ with respect to $\boldsymbol{\varepsilon}$ be positive-definite, which is identical to the conditions $\mu>0$, $3\lambda+2\mu>0$.

The paper is organized as follows. First, the Lagrangian expression of inequality (11) is derived for both unconstrained and constrained materials. The important particular case of isotropic materials is envisaged next. We then study connections with some other constitutive restrictions, namely the H_0 inequality [because it was that recommended by Hill, and also because of its strong resemblance with (11)] and the polyconvexity condition (because of its mathematical importance). Illustrative examples are finally provided, paying again attention to the comparison with respect to the H_0 inequality and the polyconvexity condition (and also incidentally the $H_{3/2}$ inequality, for the reason explained above).

2. Lagrangian expression of the inequality $\breve{\boldsymbol{\sigma}}:\mathbf{D}>0$

2 *a*. Case of unconstrained materials

First, $\breve{\boldsymbol{\sigma}}$ can be expressed in terms of objective quantities by using the relations $\breve{\boldsymbol{\sigma}}=\dot{\boldsymbol{\sigma}}+\boldsymbol{\sigma}\,.\,\boldsymbol{\Omega}-\boldsymbol{\Omega}\,.\,\boldsymbol{\sigma}$ and $\boldsymbol{\sigma}=(1/J)\,\mathbf{F}\,.\,\boldsymbol{\pi}\,.\,\mathbf{F}^T$, where $\boldsymbol{\Omega}=(1/2)\,[\dot{\mathbf{F}}\,.\,\mathbf{F}^{-1}-(\mathbf{F}^{-1})^T\,.\,\dot{\mathbf{F}}^T]$ denotes the rotation rate of the matter's comoving frame and $\boldsymbol{\pi}$ the second Piola-Kirchhoff stress tensor:

$$\breve{\boldsymbol{\sigma}}=\frac{1}{J}\left[\mathbf{F}\,.\,\dot{\boldsymbol{\pi}}\,.\,\mathbf{F}^T+\mathbf{D}\,.\,\mathbf{F}\,.\,\boldsymbol{\pi}\,.\,\mathbf{F}^T+\mathbf{F}\,.\,\boldsymbol{\pi}\,.\,\mathbf{F}^T\,.\,\mathbf{D}-\frac{\dot{J}}{J}\mathbf{F}\,.\,\boldsymbol{\pi}\,.\,\mathbf{F}^T\right].$$

Second, the inequality $\breve{\boldsymbol{\sigma}}:\mathbf{D}=\operatorname{tr}(\breve{\boldsymbol{\sigma}}\,.\,\mathbf{D})>0$ can be formulated in terms of $\boldsymbol{\pi}$ and the conjugate Green-Lagrange, or Green-Saint Venant strain tensor $\mathbf{e}=(1/2)(\mathbf{C}-\mathbf{1})$ with the aid of the equations $\dot{J}/J=\operatorname{tr}\mathbf{D}$ and $\mathbf{D}=(\mathbf{F}^{-1})^T\,.\,\dot{\mathbf{e}}\,.\,\mathbf{F}^{-1}$:

$$J\breve{\boldsymbol{\sigma}}:\mathbf{D}=\dot{\boldsymbol{\pi}}:\dot{\mathbf{e}}+2\operatorname{tr}(\mathbf{C}^{-1}\,.\,\dot{\mathbf{e}}\,.\,\boldsymbol{\pi}\,.\,\dot{\mathbf{e}})-(\mathbf{C}^{-1}:\dot{\mathbf{e}})(\boldsymbol{\pi}:\dot{\mathbf{e}})>0 \tag{12}$$

for all non-zero $\dot{\mathbf{e}}$'s. Since $\boldsymbol{\pi}=\partial\psi/\partial\mathbf{e}$, this is equivalent to requiring that the quadratic form

$$Q(\dot{\mathbf{e}})=\dot{\mathbf{e}}:\frac{\partial^2\psi}{\partial\mathbf{e}\,\partial\mathbf{e}}:\dot{\mathbf{e}}+2\operatorname{tr}\left(\mathbf{C}^{-1}\,.\,\dot{\mathbf{e}}\,.\,\frac{\partial\psi}{\partial\mathbf{e}}\,.\,\dot{\mathbf{e}}\right)-(\mathbf{C}^{-1}:\dot{\mathbf{e}})\left(\frac{\partial\psi}{\partial\mathbf{e}}:\dot{\mathbf{e}}\right) \tag{13}$$

be positive-definite over the space of second rank symmetric tensors.

Just like Hill's inequalities H_m, this condition is equivalent to demanding that the Hessian form of ψ with respect to a certain strain measure be positive-definite in the reference configuration, whatever the choice of the latter; but this strain measure is *not* of the type considered by Hill [Eq. (6)]. Indeed let the current state be taken as the reference configuration (such a choice is possible since the condition $Q(\dot{\mathbf{e}})>0$, which derives from inequality (11), is obviously independent of the reference configuration). Furthermore, let $\mathbf{E}$ be any strain tensor such that

$$\mathbf{E}=\mathbf{e}-\mathbf{e}^2+\frac{1}{2}(\operatorname{tr}\mathbf{e})\,\mathbf{e}+O(\|\mathbf{e}\|^3). \tag{14}$$

(The first two terms of the right-hand side are the same as for the logarithmic strain involved in the H_0 inequality, but the last one is new.) Using the obvious relations

$$\mathbf{e}=\mathbf{E}+\mathbf{E}^2-\frac{1}{2}(\operatorname{tr}\mathbf{E})\,\mathbf{E}+O(\|\mathbf{E}\|^3)$$

$$\Rightarrow \frac{\partial e_{ij}}{\partial E_{kl}}=\frac{1}{2}\left[\left(1-\frac{\operatorname{tr}\mathbf{E}}{2}\right)(\delta_{ik}\,\delta_{jl}+\delta_{il}\,\delta_{jk})\right.$$

$$\left.+\delta_{ik}\,E_{jl}+\delta_{il}\,E_{jk}+\delta_{jk}\,E_{il}+\delta_{jl}\,E_{ik}-\delta_{kl}\,E_{ij}\right]+O(\|\mathbf{E}\|^2),$$

one readily calculates the second derivatives of ψ with respect to the E_{ij}'s in the reference configuration ($\mathbf{E}=\mathbf{O}$):

$$\frac{\partial^2\psi}{\partial E_{ij}\,\partial E_{kl}}=\frac{\partial^2\psi}{\partial e_{ij}\,\partial e_{kl}}+\frac{1}{2}\left(\delta_{ik}\frac{\partial\psi}{\partial e_{jl}}+\delta_{il}\frac{\partial\psi}{\partial e_{jk}}+\delta_{jk}\frac{\partial\psi}{\partial e_{il}}+\delta_{jl}\frac{\partial\psi}{\partial e_{ik}}-\delta_{ij}\frac{\partial\psi}{\partial e_{kl}}-\delta_{kl}\frac{\partial\psi}{\partial e_{ij}}\right).$$

It follows that in this configuration,

$$\dot{\mathbf{E}}:\frac{\partial^2\psi}{\partial\mathbf{E}\,\partial\mathbf{E}}:\dot{\mathbf{E}}=\dot{\mathbf{e}}:\frac{\partial^2\psi}{\partial\mathbf{e}\,\partial\mathbf{e}}:\dot{\mathbf{e}}+2\operatorname{tr}\left(\dot{\mathbf{e}}\,.\,\frac{\partial\psi}{\partial\mathbf{e}}\,.\,\dot{\mathbf{e}}\right)-(\operatorname{tr}\dot{\mathbf{e}})\left(\frac{\partial\psi}{\partial\mathbf{e}}:\dot{\mathbf{e}}\right),$$

which is identical to the right-hand side of Eq. (13) (with $\mathbf{C}=\mathbf{1}$). Hence it is equivalent to require the positive-definiteness of $Q(\dot{\mathbf{e}})$ or that of the Hessian form of ψ with respect to $\mathbf{E}$, provided that the current state is taken as the reference configuration.

2*b*. Case of constrained materials

We now consider materials subject to internal constraints of the form

$$\varphi_p(\mathbf{e})=0 \qquad (p=1,\ \ldots,\ n) \tag{15}$$

where the φ_p's are given functions. Then all envisageable strain rates obey the restrictions

$$\frac{\partial\varphi_p}{\partial\mathbf{e}}:\dot{\mathbf{e}}=0 \qquad (p=1,\ \ldots,\ n) \tag{16}$$

and the constitutive law reads

$$\pi=\frac{\partial\psi}{\partial\mathbf{e}}+\sum_{p=1}^{n}\eta_p\frac{\partial\varphi_p}{\partial\mathbf{e}} \tag{17}$$

where the η_p's are Lagrange multipliers.

As mentioned in the Introduction, there are two possibilities for such materials: the constitutive inequality (12) can be considered to be applicable either directly to π, or only to its first term $\partial\psi/\partial\mathbf{e}$. Examples provided below show that restrictions on the

Lagrange multipliers are desired in general (the case of incompressible materials representing an exception). This favours the first possibility, which will therefore be adopted here, in conformity with Hill's choice [H, 1968, 1970] and in conflict with that suggested by Wang and Truesdell [W and T, 1973]. This leads to demanding that the quadratic form

$$(18)\quad Q(\dot{\mathbf{e}})=\dot{\mathbf{e}}:\frac{\partial^2\psi}{\partial\mathbf{e}\,\partial\mathbf{e}}:\dot{\mathbf{e}}+2\,\mathrm{tr}\left(\mathbf{C}^{-1}\,.\,\dot{\mathbf{e}}\,.\,\frac{\partial\psi}{\partial\mathbf{e}}\,.\,\dot{\mathbf{e}}\right)-(\mathbf{C}^{-1}:\dot{\mathbf{e}})\left(\frac{\partial\psi}{\partial\mathbf{e}}:\dot{\mathbf{e}}\right)$$
$$+\sum_{p=1}^{n}\eta_p\left[\dot{\mathbf{e}}:\frac{\partial^2\varphi_p}{\partial\mathbf{e}\,\partial\mathbf{e}}:\dot{\mathbf{e}}+2\,\mathrm{tr}\left(\mathbf{C}^{-1}\,.\,\dot{\mathbf{e}}\,.\,\frac{\partial\varphi_p}{\partial\mathbf{e}}\,.\,\dot{\mathbf{e}}\right)\right]$$

be positive-definite over the subspace of strain rates verifying conditions (16).

In the case of the incompressibility constraint, the above condition is, exceptionally, independent of the Lagrange multiplier. This can be deduced either from the fact that it is then equivalent to inequality H_0 which does satisfy this property (this was even, as explained in the Introduction, the reason that motivated Hill's choice of the value $m=0$), or from a direct calculation using Eq. (18) and the expression of the incompressibility constraint, *i.e.* $\varphi(\mathbf{e})=\det(\mathbf{1}+2\,\mathbf{e})-1=0$.

3. Isotropic materials

We shall now investigate the important special case of (unconstrained) isotropic materials. The results derived below offer strong similarities with those obtained by [H, 1970] for the H_0 inequality, but the treatment is somewhat different.

Provided that isotropy is preserved in the reference configuration chosen (this means that the corresponding stress state must be hydrostatic), the stored-energy is an isotropic function of $\mathbf{e}$ and $\boldsymbol{\pi}$ is given by

$$(19)\qquad \boldsymbol{\pi}=\sum_i\frac{\partial\psi}{\partial e_i}\mathbf{U}_i\otimes\mathbf{U}_i$$

where the e_i's and $\mathbf{U}_i$'s denote the eigenvalues and (unit) eigenvectors of $\mathbf{e}$. To obtain the expression of $\dot{\boldsymbol{\pi}}$, that of the $\dot{\mathbf{U}}_i$'s is needed. The latter can be derived by differentiating the expression of $\mathbf{e}$:

$$\mathbf{e}=\sum_i e_i\,\mathbf{U}_i\otimes\mathbf{U}_i\quad\Rightarrow\quad\dot{\mathbf{e}}=\sum_i(\dot{e}_i\,\mathbf{U}_i\otimes\mathbf{U}_i+e_i\,\dot{\mathbf{U}}_i\otimes\mathbf{U}_i+e_i\,\mathbf{U}_i\otimes\dot{\mathbf{U}}_i),$$

and contracting both sides between $\mathbf{U}_j$ and $\mathbf{U}_k$; since $\dot{\mathbf{U}}_j\,.\,\mathbf{U}_k+\mathbf{U}_j\,.\,\dot{\mathbf{U}}_k=0$, the result is

$$\dot{e}_{jk}=\dot{e}_j\,\delta_{jk}+(e_j-e_k)\,\dot{\mathbf{U}}_j\,.\,\mathbf{U}_k$$

where the indices are not summed and the $\dot{e}_{jk}$'s denote the components of $\dot{\mathbf{e}}$ in a *fixed* basis coincident with the basis $(\mathbf{U}_1, \mathbf{U}_2, \mathbf{U}_3)$ at the instant considered. This implies that

$$\dot{e}_j=\dot{e}_{jj};\qquad \dot{\mathbf{U}}_j\,.\,\mathbf{U}_k=\frac{\dot{e}_{jk}}{e_j-e_k}\quad\text{for } j\neq k$$

(where the e_i's are assumed to be distinct). It follows that

$$\dot{\mathrm{U}}_i = \sum_{j \neq i} (\dot{\mathrm{U}}_i \,.\, \mathrm{U}_j)\, \mathrm{U}_j = \sum_{j \neq i} \frac{\dot{e}_{ij}}{e_i - e_j} \mathrm{U}_j$$

and hence that

$$\dot{\pi} = \sum_{i,\,j} \frac{\partial^2 \psi}{\partial e_i \partial e_j} \dot{e}_{jj} \mathrm{U}_i \otimes \mathrm{U}_i + \sum_{i \neq j} \frac{\partial \psi / \partial e_i - \partial \psi / \partial e_j}{e_i - e_j} \dot{e}_{ij} \mathrm{U}_i \otimes \mathrm{U}_j.$$

Contracting both sides with $\dot{\mathbf{e}} = \sum_{k,\,l} \dot{e}_{kl} \mathrm{U}_k \otimes \mathrm{U}_l$, one gets

$$\dot{\pi} : \dot{\mathbf{e}} = \sum_{i,\,j} \frac{\partial^2 \psi}{\partial e_i \partial e_j} \dot{e}_{ii} \dot{e}_{jj} + \sum_{i \neq j} \frac{\partial \psi / \partial e_i - \partial \psi / \partial e_j}{e_i - e_j} \dot{e}_{ij}^2.$$

Furthermore,

$$\operatorname{tr}(\mathrm{C}^{-1} \,.\, \dot{\mathbf{e}} \,.\, \pi \,.\, \dot{\mathbf{e}}) = \sum_{i,\,j} \frac{1}{v_i^2} \dot{e}_{ij} \frac{\partial \psi}{\partial e_j} \dot{e}_{ji} = \sum_i \frac{1}{v_i^2} \frac{\partial \psi}{\partial e_i} \dot{e}_{ii}^2 + \frac{1}{2} \sum_{i \neq j} \left(\frac{1}{v_i^2} \frac{\partial \psi}{\partial e_j} + \frac{1}{v_j^2} \frac{\partial \psi}{\partial e_i} \right) \dot{e}_{ij}^2$$

(where $v_i = \sqrt{1 + 2 e_i}$) and

$$(\mathrm{C}^{-1} : \dot{\mathbf{e}})(\pi : \dot{\mathbf{e}}) = \left(\sum_i \frac{\dot{e}_{ii}}{v_i^2} \right) \left(\sum_j \frac{\partial \psi}{\partial e_j} \dot{e}_{jj} \right) = \sum_i \frac{1}{v_i^2} \frac{\partial \psi}{\partial e_i} \dot{e}_{ii}^2 + \frac{1}{2} \sum_{i \neq j} \left(\frac{1}{v_i^2} \frac{\partial \psi}{\partial e_j} + \frac{1}{v_j^2} \frac{\partial \psi}{\partial e_i} \right) \dot{e}_{ii} \dot{e}_{jj};$$

thus the quadratic form defined by Eq. (13) is the sum of two *independent* quadratic forms, each of which must therefore be positive-definite:

(20) $$\mathrm{Q}(\dot{\mathbf{e}}) = \mathrm{Q}_1(\dot{e}_{11}, \dot{e}_{22}, \dot{e}_{33}) + \mathrm{Q}_2(\dot{e}_{12}, \dot{e}_{23}, \dot{e}_{31})$$

where

$$\mathrm{Q}_1(\dot{e}_{11}, \dot{e}_{22}, \dot{e}_{33}) = \sum_i \left(\frac{\partial^2 \psi}{\partial e_i^2} + \frac{1}{v_i^2} \frac{\partial \psi}{\partial e_i} \right) \dot{e}_{ii}^2 + \sum_{i \neq j} \left[\frac{\partial^2 \psi}{\partial e_i \partial e_j} - \frac{1}{2} \left(\frac{1}{v_i^2} \frac{\partial \psi}{\partial e_j} + \frac{1}{v_j^2} \frac{\partial \psi}{\partial e_i} \right) \right] \dot{e}_{ii} \dot{e}_{jj};$$

$$\mathrm{Q}_2(\dot{e}_{12}, \dot{e}_{23}, \dot{e}_{31}) = \sum_{i \neq j} \left(\frac{\partial \psi / \partial e_i - \partial \psi / \partial e_j}{e_i - e_j} + \frac{1}{v_i^2} \frac{\partial \psi}{\partial e_j} + \frac{1}{v_j^2} \frac{\partial \psi}{\partial e_i} \right) \dot{e}_{ij}^2.$$

The expressions of Q_1 and Q_2 can be simplified by noting that by Eq. (19),

$$\boldsymbol{\sigma} = \frac{1}{\mathrm{J}} \mathbf{F} \,.\, \left(\sum_i \frac{\partial \psi}{\partial e_i} \mathrm{U}_i \otimes \mathrm{U}_i \right) . \, \mathbf{F}^{\mathrm{T}} = \frac{1}{\mathrm{J}} \sum_i \frac{\partial \psi}{\partial e_i} \mathbf{u}_i \otimes \mathbf{u}_i$$

where $\mathbf{u}_i = \mathbf{F} \,.\, \mathrm{U}_i$; since $\| \mathbf{u}_i \| = v_i$, this implies that the eigenvalues σ_i of $\boldsymbol{\sigma}$ are given by

(21) $$\sigma_i = \frac{v_i^2}{\mathrm{J}} \frac{\partial \psi}{\partial e_i} = \frac{1}{\mathrm{J}} \frac{\partial \psi}{\partial (\ln v_i)}.$$

It is then easy to show that

$$\frac{\partial^2 \psi}{\partial e_i^2} + \frac{1}{v_i^2} \frac{\partial \psi}{\partial e_i} = \frac{J}{v_i^4} \frac{\partial \sigma_i}{\partial (\ln v_i)}$$

and

$$\frac{\partial^2 \psi}{\partial e_i \partial e_j} - \frac{1}{2}\left(\frac{1}{v_i^2} \frac{\partial \psi}{\partial e_j} + \frac{1}{v_j^2} \frac{\partial \psi}{\partial e_i}\right) = \frac{J}{2 v_i^2 v_j^2}\left(\frac{\partial \sigma_i}{\partial (\ln v_j)} + \frac{\partial \sigma_j}{\partial (\ln v_i)}\right) \quad \text{for} \quad i \neq j;$$

it follows that

$$\text{(22)} \qquad Q_1(\dot{e}_{11}, \dot{e}_{22}, \dot{e}_{33}) = \frac{J}{2} \sum_{i,j} \left(\frac{\partial \sigma_i}{\partial (\ln v_j)} + \frac{\partial \sigma_j}{\partial (\ln v_i)}\right) \frac{\dot{e}_{ii}}{v_i^2} \frac{\dot{e}_{jj}}{v_j^2}$$

and hence that Q_1 is positive-definite if, and only if, the same is true of the tensor **A** of components

$$\text{(23)} \qquad A_{ij} = \frac{1}{2}\left(\frac{\partial \sigma_i}{\partial (\ln v_j)} + \frac{\partial \sigma_j}{\partial (\ln v_i)}\right).$$

Furthermore Q_2 is reduced, after a few manipulations, to the form

$$\text{(24)} \qquad Q_2(\dot{e}_{12}, \dot{e}_{23}, \dot{e}_{31}) = J \sum_{i \neq j} \frac{\sigma_i - \sigma_j}{v_i^2 - v_j^2} \left(\frac{1}{v_i^2} + \frac{1}{v_j^2}\right) \dot{e}_{ij}^2,$$

and is thus seen to be positive-definite if, and only if, the ordering of the principal Cauchy stresses is the same as that of the principal strains. Now [H, 1970] has shown in a very elegant way that this second condition is automatically satisfied provided that the first one is (it suffices to replace the e_i's and t_i's in Hill's reasoning by the $\ln v_i$'s and σ_i's). Hence, for an isotropic material, inequality (11) is equivalent to the requirement that *the tensor* **A** *be positive-definite*. It is recalled that the σ_i's in the expression (23) of this tensor are given by Eq. (21).

In contrast, Hill's H_0 inequality $\overset{\circ}{\boldsymbol{\tau}} : \mathbf{D} > 0$ (τ: Kirchhoff stress tensor) is satisfied if, and only if, the tensor of components $\partial \tau_i / \partial (\ln v_j)$ [which is symmetric since $\tau_i = J \sigma_i = \partial \psi / \partial (\ln v_i)$] is positive-definite [H, 1970]. Hill also established the equivalence of this condition and the positive-definiteness of the Hessian form of ψ with respect to the logarithmic strain $(1/2) \ln (\mathbf{1} + 2\mathbf{e})$. (The latter condition differs from that mentioned in the Introduction for arbitrary, non-isotropic materials in that here the reference configuration is not the current state but any configuration corresponding to a hydrostatic stress). It does not seem possible, however, to put inequality (11) under a similar form (the difficulty being that the A_{ij}'s cannot be expressed as second derivatives of ψ, because of the factor $1/J$ in the expression (21) of σ_i).

In the case of a hydrostatic stress ($\boldsymbol{\sigma} = -p\mathbf{1}$), the diagonal terms of **A** are all equal to $\partial \sigma_1 / \partial (\ln v_1)$ and the off-diagonal ones to $\partial \sigma_1 / \partial (\ln v_2)$. **A** is therefore identical to the

Hessian matrix $\begin{bmatrix} \lambda+2\mu & \lambda & \lambda \\ \lambda & \lambda+2\mu & \lambda \\ \lambda & \lambda & \lambda+2\mu \end{bmatrix}$ of the stored-energy function of an isotropic material in the natural state with Lamé coefficients

$$\lambda = \frac{\partial \sigma_1}{\partial (\ln v_2)} \qquad \text{and} \qquad \mu = \frac{1}{2}\left(\frac{\partial \sigma_1}{\partial (\ln v_1)} - \frac{\partial \sigma_1}{\partial (\ln v_2)}\right).$$

Using the well-known conditions for such a matrix to be positive-definite, one concludes that inequality (11) reduces to the restrictions

$$\frac{\partial \sigma_1}{\partial (\ln v_1)} + 2\frac{\partial \sigma_1}{\partial (\ln v_2)} > 0; \qquad \frac{\partial \sigma_1}{\partial (\ln v_1)} - \frac{\partial \sigma_1}{\partial (\ln v_2)} > 0.$$

These inequalities are easily seen to be equivalent to (with obvious notations)

$$(25) \qquad \frac{\partial p}{\partial \rho} > 0; \qquad \frac{\partial \sigma_{12}}{\partial e_{12}} > 0;$$

this means that *the bulk and shear moduli must be positive*. For the H_0 inequality, the result is the same for the shear modulus but the bulk modulus must verify $\partial p/\partial \rho > p/\rho$ [H, 1970]. Thus this inequality tolerates negative bulk moduli in tension ($p<0$). This feature is plainly undesirable; it means for instance that the material can be unstable when subjected to a constant hydrostatic Cauchy stress, which is a pretty singular behaviour. Also, in rubbery materials for which the ratio of bulk to shear moduli is very large, the speed of longitudinal waves is almost equal to the square root of the former modulus; thus a negative value for this modulus means that no such waves exist.

4. Connections with other constitutive restrictions

4*a*. The H_0 inequality

Elements of comparison between inequalities (11) and H_0 have already been provided in the Introduction and Section 3, and can be summarized as follows:

Arbitrary materials (the current state is taken as the reference configuration):

H_0: $\breve{\boldsymbol{\tau}} : \mathbf{D} > 0$

$\Leftrightarrow$ The Hessian form of ψ with respect to $(1/2)\ln(\mathbf{1}+2\mathbf{e})$, or more generally any strain tensor of the form $\mathbf{e}-\mathbf{e}^2+O(\|\mathbf{e}^3\|)$, is positive-definite in the reference configuration

(11): $\breve{\boldsymbol{\sigma}} : \mathbf{D} > 0$

$\Leftrightarrow$ The Hessian form of ψ with respect to any strain tensor of the form $\mathbf{e}-\mathbf{e}^2+(1/2)(\operatorname{tr}\mathbf{e})\,\mathbf{e}^2+O(\|\mathbf{e}^3\|)$ is positive-definite in the reference configuration

Incompressible materials:

The two inequalities are equivalent

Isotropic materials (a hydrostatic stress state is taken as the reference configuration):

H_0:

$\Leftrightarrow$ The tensor of components $\partial\tau_i/\partial(\ln v_j)=\partial^2\psi/\partial(\ln v_i)\,\partial(\ln v_j)$ is positive-definite

$\Leftrightarrow$ The Hessian form of ψ with respect to $(1/2)\ln(\mathbf{1}+2\,\mathbf{e})$ is positive-definite

$$\Leftrightarrow \frac{\partial p}{\partial \rho}>\frac{p}{\rho}, \qquad \frac{\partial \sigma_{12}}{\partial e_{12}}>0 \quad \text{if } \boldsymbol{\sigma}=-p\,\mathbf{1}$$

(11):

$\Leftrightarrow$ The tensor of components $(1/2)\,(\partial\sigma_i/\partial(\ln v_j)+\partial\sigma_j/\partial(\ln v_i))$ is positive-definite

$$\Leftrightarrow \frac{\partial p}{\partial \rho}>0, \qquad \frac{\partial \sigma_{12}}{\partial e_{12}}>0 \quad \text{if } \boldsymbol{\sigma}=-p\,\mathbf{1}$$

4*b*. The polyconvexity property

We shall show here that neither inequality (11) nor polyconvexity entails the other property in general. To prove that polyconvexity does not imply inequality (11), it suffices to consider an isotropic solid in the natural state with Lamé coefficients such that $\mu>0$ and $\lambda+2\mu>0$ but $3\lambda+2\mu<0$: then polyconvexity holds but inequality (11) does not (*see* the Introduction).

Since polyconvexity implies the weak form (with "$\geqq$") of the Legendre-Hadamard condition (3) [B, 1977], proving that inequality (11) does not entail the latter is sufficient to establish that it does not imply polyconvexity. Proving this requires to know the Lagrangian form of the weak Legendre-Hadamard condition, which is easily found to be

$$(26) \qquad (\mathbf{U}\otimes\mathbf{V}):\frac{\partial^2\psi}{\partial\mathbf{e}\,\partial\mathbf{e}}:(\mathbf{U}\otimes\mathbf{V})+\left(\mathbf{U}\,.\,\frac{\partial\psi}{\partial\mathbf{e}}\,.\,\mathbf{U}\right)(\mathbf{V}\,.\,\mathbf{C}^{-1}\,.\,\mathbf{V})\geqq 0$$

(the vectors $\mathbf{v}$ in Eq. (3) and $\mathbf{V}$ here are connected by $\mathbf{V}=\mathbf{F}^{\mathrm{T}}\,.\,\mathbf{v}$).

Before proving that (11) does not imply (26), it is worth noting that it does, however, entail the following "symmetrized" variant of this inequality:

$$(\mathbf{U}\otimes\mathbf{V}):\frac{\partial^2\psi}{\partial\mathbf{e}\,\partial\mathbf{e}}:(\mathbf{U}\otimes\mathbf{V})+\frac{1}{2}\left[\left(\mathbf{U}\,.\,\frac{\partial\psi}{\partial\mathbf{e}}\,.\,\mathbf{U}\right)(\mathbf{V}\,.\,\mathbf{C}^{-1}\,.\,\mathbf{V})+\left(\mathbf{V}\,.\,\frac{\partial\psi}{\partial\mathbf{e}}\,.\,\mathbf{V}\right)(\mathbf{U}\,.\,\mathbf{C}^{-1}\,.\,\mathbf{U})\right]\geqq 0.$$

This follows at once from application of the condition $Q(\dot{\mathbf{e}})>0$ to the tensor $\dot{\mathbf{e}}=(1/2)\,(\mathbf{U}\otimes\mathbf{V}+\mathbf{V}\otimes\mathbf{U})$.

We shall consider some ψ's with first and second derivatives given by

$$\frac{\partial\psi}{\partial e_{ij}}=a_i \text{ if } i=j,\ 0 \text{ otherwise}; \qquad \frac{\partial^2\psi}{\partial e_{ij}\,\partial e_{kl}}=b \text{ if } i=j=k=l,\ 0 \text{ otherwise}$$

in the reference configuration, in some orthonomal basis. (Such a choice is possible since these formulae respect the symmetries under exchange of i and j, k and l, and (i, j) and (k, l)). Then, in this configuration,

$$\dot{\mathrm{e}} : \frac{\partial^2 \psi}{\partial \mathrm{e}\, \partial \mathrm{e}} : \dot{\mathrm{e}} = b \sum_i \dot{e}_{ii}^2;$$

$$\operatorname{tr}\left(\mathrm{C}^{-1} \cdot \dot{\mathrm{e}} \cdot \frac{\partial \psi}{\partial \mathrm{e}} \cdot \dot{\mathrm{e}}\right) = \sum_{i,j} \dot{e}_{ij}\, a_j\, \dot{e}_{ji} = \sum_i a_i\, \dot{e}_{ii}^2 + \frac{1}{2} \sum_{i \neq j} (a_i + a_j)\, \dot{e}_{ij}^2;$$

$$(\mathrm{C}^{-1} : \dot{\mathrm{e}}) \left(\frac{\partial \psi}{\partial \mathrm{e}} : \dot{\mathrm{e}}\right) = \sum_{i,j} \dot{e}_{ii}\, a_j\, \dot{e}_{jj} = \sum_i a_i\, \dot{e}_{ii}^2 + \frac{1}{2} \sum_{i \neq j} (a_i + a_j)\, \dot{e}_{ii}\, \dot{e}_{jj},$$

so that the quadratic form defined by Eq. (13) can be written as

$$\mathrm{Q}(\dot{e}) = \mathrm{Q}_1(\dot{e}_{11}, \dot{e}_{22}, \dot{e}_{33}) + \mathrm{Q}_2(\dot{e}_{12}, \dot{e}_{23}, \dot{e}_{31}),$$

$$\mathrm{Q}_1(\dot{e}_{11}, \dot{e}_{22}, \dot{e}_{33}) = \sum_i (a_i + b)\, \dot{e}_{ii}^2 - \frac{1}{2} \sum_{i \neq j} (a_i + a_j)\, \dot{e}_{ii}\, \dot{e}_{jj},$$

$$\mathrm{Q}_2(\dot{e}_{12}, \dot{e}_{23}, \dot{e}_{31}) = \sum_{i \neq j} (a_i + a_j)\, \dot{e}_{ij}^2.$$

Q_1 is positive-definite for sufficiently large values of b. On the other hand, Q_2 is positive-definite if, and only if, $a_i + a_j > 0$ for $i \neq j$. Hence there exists a function $b_0(a_1, a_2, a_3)$ such that the conditions

$$b > b_0(a_1, a_2, a_3); \quad a_1 + a_2 > 0; \quad a_2 + a_3 > 0; \quad a_3 + a_1 > 0 \tag{27}$$

ensure the positive-definiteness of $\mathrm{Q}(\dot{e})$.

On the other hand,

$$(\mathrm{U} \otimes \mathrm{V}) : \frac{\partial^2 \psi}{\partial \mathrm{e}\, \partial \mathrm{e}} : (\mathrm{U} \otimes \mathrm{V}) = b \sum_i \mathrm{U}_i^2 \mathrm{V}_i^2; \quad \left(\mathrm{U} \cdot \frac{\partial \psi}{\partial \mathrm{e}} \cdot \mathrm{U}\right)(\mathrm{V} \cdot \mathrm{C}^{-1} \cdot \mathrm{V}) = \sum_{i,j} a_i\, \mathrm{U}_i^2\, \mathrm{V}_j^2$$

so that inequality (26) reads

$$\sum_i \left(b\, \mathrm{V}_i^2 + a_i \sum_j \mathrm{V}_j^2\right) \mathrm{U}_i^2 \geqq 0;$$

this requires

$$b\, \mathrm{V}_i^2 + a_i \sum_j \mathrm{V}_j^2 = (a_i + b)\, \mathrm{V}_i^2 + a_i \sum_{j,\, j \neq i} \mathrm{V}_j^2 \geqq 0$$

or equivalently

$$\begin{cases} a_1 \geqq 0; & a_2 \geqq 0; & a_3 \geqq 0; \\ a_1 + b \geqq 0; & a_2 + b \geqq 0; & a_3 + b \geqq 0. \end{cases} \tag{28}$$

There exist some a_1, a_2, a_3, b satisfying (27) but not (28) (take a_1, a_2, a_3 such that $a_1+a_2>0$, $a_2+a_3>0$, $a_3+a_1>0$ but $a_1<0$, and b sufficiently great). For those parameters, inequality (11) holds but polyconvexity does not.

Incidentally, the same kind of proof can be used to show that Hill's inequality H_0 does not imply polyconvexity either. Indeed it is easy to see that this inequality is equivalent to requiring the positive-definiteness of a quadratic form identical to that defined by (13) except for the final term $-(C^{-1}:\dot{e})((\partial\psi/\partial e):\dot{e})$; this reduces to the conditions

$$\begin{aligned} &2a_1+b>0; \quad 2a_2+b>0; \quad 2a_3+b>0;\\ &a_1+a_2>0; \quad a_2+a_3>0; \quad a_3+a_1>0 \end{aligned}$$

for the ψ's considered above. It is easy to find some a_1, a_2, a_3, b satisfying these inequalities while violating (28).

5. Examples

We shall first give three simple examples involving materials with internal constraints, then two more complex examples of unconstrained media.

5*a*. The Neo-Hookean material

This is the simplest prototype of an incompressible isotropic material; the natural state being taken as the reference configuration, the stored-energy function is given by

$$\psi=\mu \operatorname{tr} e,$$

where μ is the shear modulus [Treloar, 1975].

For such an incompressible medium, inequality (11) is equivalent to H_0, and reduces to the requirement $\mu>0$ [H, 1968], which is reasonable enough. Writing $\operatorname{tr} e$ under the form $(1/2)(v_1^2+v_2^2+v_3^2-3)$ and using Ball's Theorem 5.2 [B, 1977], one sees that the (again reasonable) condition $\mu\geqq 0$ ensures polyconvexity. On the other hand, inequality $H_{3/2}$ yields unsatisfactory consequences. Indeed, if it is supposed to be applicable to π itself, an undesired restriction is put on the Lagrange multiplier [H, 1968]; if, conversely, it is applied only to that part of π which is independent of the Lagrange multiplier, it can be verified to reduce to $\mu<0$ in the natural state, which is absurd.

5*b*. A perfectly flexible, inextensible thread

We shall now consider a material made of parallel, perfectly flexible, unstretchable fibers (a thread or a rod for instance). The stored-energy function is zero and the internal constraint is described by

$$\varphi(e)=\mathbf{U}\,.\,e\,.\,\mathbf{U}=0,$$

where $\mathbf{U}$ is a unit vector parallel to the fibers in the reference configuration.

The quadratic form defined by Eq. (18) is given by

$$Q(\dot{e})=2\eta\,\mathrm{tr}[C^{-1}\,.\,\dot{e}\,.\,(U\otimes U)\,.\,\dot{e}]=2\eta\,(\dot{e}\,.\,U)\,.\,C^{-1}\,.\,(\dot{e}\,.\,U)=2\eta\,\|(F^{-1})^T\,.\,\dot{e}\,.\,U\|^2$$

where η is the Lagrange multiplier associated to the internal constraint. The condition $Q(\dot{e})>0$ (for all non-zero strain rates verifying (16), *i.e.* $U\,.\,\dot{e}\,.\,U=0$) implies $\eta>0$. This necessary condition is also sufficient provided that the inequality is required to be strict only for those $\dot{e}$'s verifying (16) *and such that* $\dot{e}\,.\,U\neq O$; this restriction is analogous to that made for perfect fluids (*see* the Introduction) and is natural since $\dot{e}\,.\,U=O$ means that the material is deformed only in the plane orthogonal to the fibers, which is a solicitation to which it offers no resistance. The meaning of the condition $\eta>0$ is easily elucidated by evaluating π and σ:

$$\pi=\eta\,U\otimes U\quad\Rightarrow\quad\sigma=\frac{\eta}{J}F\,.\,(U\otimes U)\,.\,F^T=\frac{\eta}{J}u\otimes u,\qquad u=F\,.\,U.$$

Since u is collinear to the fibers in the present configuration, $\eta>0$ means that the fibers must be under tension, which is a sensible result.

Inequality H_0 can be verified to be again equivalent to (11) in the case considered, provided that it is assumed to apply to π itself and not only to that part of this tensor which does not depend on the Lagrange multiplier. Concerning polyconvexity, it is not clear what precise property of ψ should be required for a material subject to the internal constraint considered here; however this property will surely be satisfied for the *zero* ψ which corresponds to a perfectly flexible rod. Hence "polyconvexity" of ψ (whatever it may be) is certainly too weak to forbid compressive states. Finally inequality $H_{3/2}$ again yields unsatisfactory consequences: if applied to π itself, it can be checked to reduce to $\eta<0$; if applied to that part of π which is independent of η, it does not prohibit compressive stresses.

In view of the fact that this example and the preceding one have clearly evidenced the inadequacy of inequality $H_{3/2}$, the discussion will be restricted from now on to inequalities (11) and H_0 and the polyconvexity property.

5 *c*. A perfectly flexible, inextensible membrane

As an interesting extension of the preceding example, we shall now study an initially plane membrane, perfectly flexible but undeformable in its plane (a very thin metallic foil for instance). This is described by

$$\psi=0;\qquad \varphi_1(e)=U_1\,.\,e\,.\,U_1=0;\qquad \varphi_2(e)=U_2\,.\,e\,.\,U_2=0;$$
$$\varphi_3(e)=U_1\,.\,e\,.\,U_2+U_2\,.\,e\,.\,U_1=0,$$

where (U_1, U_2) is an orthonormal basis of the membrane plane in the reference configuration.

The condition $Q(\dot{e})>0$ is readily put under the form

$$\eta_1\|v_1\|^2+\eta_2\|v_2\|^2+2\eta_3 v_1\,.\,v_2>0,$$
$$v_1=(F^{-1})^T\,.\,\dot{e}\,.\,U_1,\qquad v_2=(F^{-1})^T\,.\,\dot{e}\,.\,U_2.$$

To find necessary conditions for this to be true, let us consider an $\dot{e}$ with matrix $\begin{bmatrix} 0 & 0 & x \\ 0 & 0 & 1 \\ x & 1 & 0 \end{bmatrix}$ in the basis (U_1, U_2, U_3), where x is an arbitrary real number and $U_3 = U_1 \times U_2$. Then conditions (16) are fulfilled, *i.e.* $U_1 . \dot{e} . U_1 = U_2 . \dot{e} . U_2 = U_1 . \dot{e} . U_2 + U_2 . \dot{e} . U_1 = 0$, and the preceding inequality reads

$$\|(F^{-1})^T . U_3\|^2 (\eta_1 x^2 + \eta_2 + 2\eta_3 x) > 0 \quad \Leftrightarrow \quad \eta_1 > 0; \qquad \eta_1 \eta_2 - \eta_3^2 > 0.$$

To show that these necessary conditions are also sufficient, let us note that

$$\eta_1 \|v_1\|^2 + \eta_2 \|v_2\|^2 + 2\eta_3 v_1 . v_2 \geqq \eta_1 \|v_1\|^2 + \eta_2 \|v_2\|^2 - 2|\eta_3| . \|v_1\| . \|v_2\|.$$

The above conditions ensure the positive-definiteness of this quadratic form of the variables $\|v_1\|$, $\|v_2\|$; $Q(\dot{e})$ is then positive for all $\dot{e}$'s verifying (16) *and such that* $\dot{e} . U_1 \neq 0$ *or* $\dot{e} . U_2 \neq 0$. This restriction is again natural since $\dot{e} . U_1 = \dot{e} . U_2 = 0$ implies that $\dot{e}$ is proportional to $U_3 \otimes U_3$, and the material offers no resistance to such solicitations.

The seemingly strange conditions $\eta_1 > 0$; $\eta_1 \eta_2 - \eta_3^2 > 0$ possess an appealing interpretation. Indeed calculation of π and σ yields

$$\pi = \eta_1 U_1 \otimes U_1 + \eta_2 U_2 \otimes U_2 + \eta_3 (U_1 \otimes U_2 + U_2 \otimes U_1)$$
$$\Rightarrow \quad J\sigma = \eta_1 u_1 \otimes u_1 + \eta_2 u_2 \otimes u_2 + \eta_3 (u_1 \otimes u_2 + u_2 \otimes u_1),$$
$$u_1 = F . U_1, \qquad u_2 = F . U_2.$$

Hence these conditions mean that the quadratic form $\mathbf{n} . \sigma . \mathbf{n}$ must be positive-definite over the tangent plane to the membrane in the present configuration; in other words, all directions of the membrane must be under tension.

With regard to other constitutive restrictions, the conclusions are the same as for the preceding example: H_0 is equivalent to (11), and the polyconvexity property is automatically satisfied and thus tolerates compressive stress states.

5*d*. The compressible Neo-Hookean material

This is probably the simplest realistic model for compressible isotropic materials. The stored-energy function is

$$\psi = \mu \operatorname{tr} e + \lambda J - (\lambda + \mu) \ln J$$

where λ and μ are the Lamé coefficients [Blatz, 1971].

For simplicity, we consider only hydrostatic stress (and strain) states ($e = e\,\mathbf{1}$). Inequality (11) reduces to conditions (25), which are easily explicited as follows:

$$3(\lambda + \mu) - \mu(1 + 2e) > 0; \qquad \mu(1 + 2e) > 0.$$

Since $1+2e>0$, the second condition is equivalent to $\mu>0$; the first one reduces then to

$$e<1+\frac{3\lambda}{2\mu}.$$

In contrast, inequality H_0 is easily seen to be equivalent to the conditions $\mu>0$; $3\lambda\sqrt{1+2e}+2\mu>0$. If, for instance, λ is supposed to be positive, inequality H_0 is automatically satisfied (provided of course that $\mu>0$) whereas inequality (11) is not: this is because the former tolerates negative compressibilities (for $e>1+3\lambda/2\mu$), in contrast with the latter. Concerning polyconvexity, it is easy, using Ball's Theorem 5.2, to see that the conditions $\mu>0$, $\lambda+\mu>0$ are sufficient to warrant this property. These conditions are weaker than the usual restrictions in the natural state, $\mu>0$, $3\lambda+2\mu>0$; hence here again polyconvexity appears to be too weak to guarantee physically reasonable behaviour in all circumstances.

5 *e*. THE MODEL OF BLATZ AND KO

This model is interesting in that it was specifically designed to describe the behaviour of a highly compressible material, namely a foam rubber. The stored-energy function is [Blatz & Ko, 1962]

$$\psi=\frac{\mu}{2}\left(\frac{1}{v_1^2}+\frac{1}{v_2^2}+\frac{1}{v_3^2}\right)+\frac{\mu(1-2\nu)}{2\nu}J^{2\nu/(1-2\nu)}$$

where μ and ν are the shear modulus and Poisson ratio (in fact Blatz and Ko's original model contained an additional parameter f; the expression given here corresponds to the value of f which they finally adopted, namely 0); this form fits into a general class of models introduced by [Ogden, 1972 *b*].

For a hydrostatic stress, inequalities (25) yield the conditions $\mu>0$ and

$$5J^{-5/3}+3\frac{4\nu-1}{1-2\nu}J^{(4\nu-1)/(1-2\nu)}>0.$$

Since highly compressible materials are in question here, it is natural to demand that this inequality be satisfied for all values of J. If $1/4\leqq\nu<1/2$, this is true; indeed $(4\nu-1)/(1-2\nu)\geqq 0$ so that the second term is non-negative. On the other hand, if $\nu<1/4$ or $\nu>1/2$, it is not; indeed if $\nu\neq-1$, $(4\nu-1)/(1-2\nu)$ is negative and not equal to $-5/3$ so that the second term is negative and dominates over the first one for $J\to 0$ or $J\to+\infty$, and if $\nu=-1$, the whole left-hand side is zero. Hence the above inequality holds for all values of J if, and only if,

$$\frac{1}{4}\leqq\nu<\frac{1}{2}.$$

On the other hand, inequality H_0 is satisfied provided that $\mu>0$ and $0\leqq\nu<1/2$ [O, 1970]; here again, this inequality tolerates negative bulk moduli (for $0\leqq\nu<1/4$). In contrast, the sufficient conditions for polyconvexity provided by Ball's Theorem 5.2 are violated since the function of v_1, v_2, v_3 involved in the expression of ψ is decreasing with respect to each variable (but this does not, of course, prove that polyconvexity is violated).

The value of ν which provided the best fit with Blatz and Ko's experimental results for a foam rubber was 1/4; it is compatible with both inequalities (11) and H_0.

6. Conclusion

In no case has inequality (11) been observed to yield unreasonable restrictions. Hill's inequality H_0 also appears to be reasonable in a number of cases; however it presents the major drawback of not prohibiting negative bulk moduli, which lead to a number of unwanted consequences (some of which were mentioned in Section 3). In no case has Ball's polyconvexity property been remarked to be over-restrictive; but it is often weaker than desired (e. g., for the compressible Neo-Hookean material).

On the other hand, the polyconvexity hypothesis has mathematical power, as was shown by [B, 1977], whereas nothing is known about the mathematical consequences of inequality (11) (nor about those of H_0). It is precisely the aim of this paper to suggest that in view of the apparent physical relevance of this inequality. a thorough investigation of the said consequences would be worthwhile.

Acknowledgement

The author expresses his warm thanks to Pr. Michel Amestoy for several stimulating discussions, and to the referees for their most pertinent comments.

REFERENCES

Antman S. S., 1983, Regular and singular problems for large elastic deformations of tubes, wedges, and cylinders, *Arch. Rat. Mech. Anal.*, **83**, 1-52.

Ball J., 1977, Convexity conditions and existence theorems in non-linear elasticity, *Arch. Rat. Mech. Anal.*, **63**, 337-403.

Blatz P. J., 1971, On the thermostatic behaviour of elastomers, *Polymer Networks Structure and Mechanical Properties*, Plenum Press, London.

Blatz P. J., Ko W. L., 1962, Application of finite elasticity theory to the deformation of rubbery materials, *Trans. Soc. Rheol.*, **6**, 223-251.

Ciarlet P. G., Geymonat G., 1982, Sur les lois de comportement en élasticité non linéaire compressible, *C. R. Acad. Sci. Paris*, **295**, Série II, 423-426.

Coleman B. D., Noll W., 1959, On the thermostatic of continuous media, *Arch. Rat. Mech. Anal.*, **4**, 97-128; reprinted in *Continuum Mechanics III, Foundations of Elasticity Theory*, Int. Sci. Rev. Ser., **VIII**, C. Truesdell Ed., Gordon and Breach, New York, 1965.

Hill R., 1968, On constitutive inequalities for simple materials-I, *J. Mech. Phys. Solids*, **16**, 229-242.

Hill R., 1970, Constitutive inequalities for isotropic elastic solids under finite strain, *Proc. R. Soc. London* A, **314**, 457-472.

Ogden R. W., 1970, Compressible isotropic elastic solids under finite strain, constitutive inequalities, *Quart. J. Mech. Appl. Math.*, **23**, 457-468.

OGDEN R. W., 1972*a*, Large deformation isotropic elasticity, on the correlation of theory and experiment for incompressible rubberlike solids, *Proc. R. Soc. London* A, **326**, 565-584.

OGDEN R. W., 1972*b*, Large deformation isotropic elasticity, on the correlation of theory and experiment for compressible rubberlike solids, *Proc. R. Soc. London* A, **328**, 567-583.

SIDOROFF F., 1974, Sur les restrictions à imposer à l'énergie de déformation d'un matériau hyperélastique, *C. R. Acad. Sci. Paris*, **279**, Série A, 379-382.

TRELOAR L. R. G., 1975, *The Physics of Rubber Elasticity*, Oxford University Press, Oxford.

TRUESDELL C., 1956, Das ungelöste Hauptproblem der endlichen Elastizitäts-Theorie, *Z. angew. Math. Mech.*, **36**, 97-103; English translation in *Continuum Mechanics III, Foundations of Elasticity Theory*, Int. Sci. Rev. Ser., **VIII**, C. Truesdell Ed., Gordon and Breach, New York, 1965.

WANG C. C., TRUESDELL C., 1973, *Introduction to Rational Elasticity*, Noordhoff, Groningen.

APPENDIX A

The polyconvexity condition for a perfect fluid

A perfect fluid is a particular type of hyperelastic material for which the stored-energy function ψ depends on $\mathbf{F}$ only through its determinant J. The Cauchy stress tensor reduces to a hydrostatic pressure given by $p=-d\psi/dJ$. The inequality $dp/d\rho>0$ is equivalent to $dp/dJ<0$ or $d^2\psi/dJ^2>0$; hence it implies that ψ is convex with respect to J, and consequently polyconvex.

To show that conversely polyconvexity implies $dp/d\rho \geqq 0$, let us consider, for any positive real number J, the tensors $\mathbf{A}(J)$ and $\mathbf{B}(J)$ with respective matrices $\begin{bmatrix} J & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}$ and $\begin{bmatrix} 1 & 0 & 0 \\ 0 & J & 0 \\ 0 & 0 & J \end{bmatrix}$ in a given orthonormal basis. Then $\operatorname{adj}\mathbf{A}(J)=\mathbf{B}(J)$ and $\det\mathbf{A}(J)=J$ so that

$$\psi[\mathbf{A}(J)]\equiv\psi(J)=\Phi[\mathbf{A}(J),\ \mathbf{B}(J),\ J]$$

where Φ is the function associated to ψ by Eq. (8). Using the convexity of Φ and the fact that $\mathbf{A}(J)$ and $\mathbf{B}(J)$ are *affine* functions of J, we then get, for any J, J′ and $\theta\in[0, 1]$:

$$\begin{aligned}\psi[\theta J+(1-\theta)J']&=\Phi\{\mathbf{A}[\theta J+(1-\theta)J'],\ \mathbf{B}[\theta J+(1-\theta)J'],\ \theta J+(1-\theta)J'\}\\&=\Phi[\theta\mathbf{A}(J)+(1-\theta)\mathbf{A}(J'),\ \theta\mathbf{B}(J)+(1-\theta)\mathbf{B}(J'),\ \theta J+(1-\theta)J']\\&\leqq\theta\Phi[\mathbf{A}(J),\ \mathbf{B}(J),\ J]+(1-\theta)\Phi[\mathbf{A}(J'),\ \mathbf{B}(J'),\ J']=\theta\psi(J)+(1-\theta)\psi(J').\end{aligned}$$

Thus ψ is convex with respect to J. This implies that $d^2\psi/dJ^2\geqq 0$, or equivalently $dp/d\rho\geqq 0$.

APPENDIX B

The polyconvexity condition for an isotropic solid in the natural state

It was shown in [B, 1977] that polyconvexity implies a weaker property termed *"quasi-convexity"*, which in turn implies the weak form (*i.e.*, with "$\geqq$" instead of "$>$") of the

Legendre-Hadamard ellipticity condition [Eq. (3)]. This condition is known to be equivalent to the inequalities $\mu \geqq 0$, $\lambda + 2\mu \geqq 0$ for an isotropic solid in the natural state. Thus polyconvexity implies these inequalities for such a solid.

Let us now show that, conversely, the conditions $\mu > 0$, $\lambda + 2\mu > 0$ imply polyconvexity in (the vicinity of) the natural state. This state being taken as the reference configuration and the square roots of the eigenvalues of $\mathbf{C} = \mathbf{F}^T . \mathbf{F}$ being denoted v_1, v_2, v_3, let the stored-energy function be written under the form

$$\psi(\mathbf{F}) = \Phi_1(v_1, v_2, v_3) + \Phi_2(v_1 v_2 v_3)$$

where the functions Φ_1 and Φ_2 are defined by (a being a parameter)

$$\Phi_1(v_1, v_2, v_3) \equiv \psi(v_1, v_2, v_3) + a v_1 v_2 v_3; \qquad \Phi_2(\delta) \equiv -a\delta.$$

By Ball's Theorem 5.2 [B, 1977], showing that Φ_1 and Φ_2 are convex and Φ_1 non-decreasing with respect to each variable is sufficient to establish polyconvexity. The convexity of Φ_2 is obvious. Furthermore the expression of ψ near the natural state $[(v_1, v_2, v_3) = (1, 1, 1)]$ is

$$\psi = \frac{\lambda}{2}(e_1 + e_2 + e_3)^2 + \mu(e_1^2 + e_2^2 + e_3^2) + 0(\|\mathbf{e}\|^3)$$

where $e_1 = (v_1^2 - 1)/2$, $e_2 = (v_2^2 - 1)/2$, $e_3 = (v_3^2 - 1)/2$ are the eigenvalues of the Green-Lagrange, of Green-Saint Venant strain tensor $\mathbf{e}$; it follows that the first and second derivatives of Φ_1 in this state are given by

$$\frac{\partial \Phi_1}{\partial v_i}(1, 1, 1) = a; \qquad \frac{\partial^2 \Phi_1}{\partial v_i \partial v_j}(1, 1, 1) = \lambda + 2\mu \quad \text{if} \quad i = j, \qquad \lambda + a \quad \text{if} \quad i \neq j.$$

Thus the condition

$$a > 0$$

ensures that Φ_1 is non-decreasing with respect to each variable. To find conditions guaranteing the positive-definiteness of its Hessian matrix, one just needs to write the latter under the form

$$\begin{bmatrix} \lambda' + 2\mu' & \lambda' & \lambda' \\ \lambda' & \lambda' + 2\mu' & \lambda' \\ \lambda' & \lambda' & \lambda' + 2\mu' \end{bmatrix}$$

where $\lambda' = \lambda + a$, $\lambda' + 2\mu' = \lambda + 2\mu \Leftrightarrow \mu' = \mu - a/2$; this is the Hessian matrix of a stored-energy function with Lamé coefficients λ' and μ', which is well known to be positive-definite under the conditions

$$\mu' > 0 \quad \Leftrightarrow \quad a < 2\mu; \qquad 3\lambda' + 2\mu' > 0 \quad \Leftrightarrow \quad a > -\frac{1}{2}(3\lambda + 2\mu).$$

There exists an a satisfying the three above inequalities if, and only if, the conditions

$$0<2\mu \Leftrightarrow \mu>0; \qquad -\frac{1}{2}(3\lambda+2\mu)<2\mu \Leftrightarrow \lambda+2\mu>0$$

are fulfilled. This concludes the proof.

Remarks

1. The use of Ball's Theorem 5.2 can be avoided by writing

$$\psi(\mathbf{F})=\Phi_1(\mathbf{F})+\Phi_2(\det \mathbf{F})$$

with

$$\Phi_1(\mathbf{F})=\psi(\mathbf{F})+a\det \mathbf{F}; \qquad \Phi_2(\delta)=-a\delta,$$

and directly checking that the Hessian form of Φ_1 with respect to $\mathbf{F}$ is positive-definite at the point $\mathbf{F}=\mathbf{1}$ under the same conditions as above.

2. It was shown by [Ciarlet & Geymonat, 1982] that for any *positive* constants λ, μ, there exists an Ogden-type stored-energy function which is polyconvex and admits λ and μ as Lamé coefficients in the natural state. This shows in particular that the inequalities $\lambda>0$, $\mu>0$ ensure polyconvexity in (the vicinity of) this state. These conditions are more restrictive than those above; it does not seem feasible, unfortunately, to adapt Ciarlet and Geymonat's proof so as to weaken them. (In this respect, the result established here is stronger than that of Ciarlet and Geymonat. However it is also weaker in that polyconvexity is established *only in* (*the vicinity of*) *the natural state*, whereas Ciarlet and Geymonat's stored-energy function is polyconvex *everywhere*.)

(Manuscript received December 26, 1990;
accepted October 7, 1991.)