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The effect of particle size, shape, distribution and their evolution on the constitutive response of nonlinearly viscous composites. I. Theory

By M. Kailasam[1], P. Ponte Castañeda[1] and J. R. Willis[2]

[1] *Department of Mechanical Engineering and Applied Mechanics, University of Pennsylvania, Philadelphia, PA 19104, USA*

[2] *Department of Applied Mathematics and Theoretical Physics, University of Cambridge, Cambridge CB3 9EW, UK*

This work deals with the development of constitutive models for two-phase nonlinearly viscous and perfectly plastic composites with evolving microstructures. The work builds on the earlier models of Ponte Castañeda & Zaidman (1994) for composites with particulate microstructures subjected to finite deformation, where the influence of the evolution of the average shape and size of the inclusions (or voids) on the overall anisotropic response of the composites was considered. The present model additionally takes into account the effect of independent changes in the random distribution of the inclusions as the deformation progresses. Thus, appropriate 'internal variables' characterizing the state of the microstructure are incorporated into the 'instantaneous' constitutive equations for the composite and 'evolution laws' for these variables are proposed. The first part of this work deals with the development of the instantaneous constitutive relations for a sufficiently broad class of microstructures to be able to consider the evolution problem under general triaxial loading conditions (with fixed loading axes). The 'aspect ratios' of the two-point distribution function are introduced as new microstructural variables, along with the aspect ratios and the volume fraction of the inclusions as proposed in the earlier models. Evolution laws are then developed for all these variables, which—when integrated together with the instantaneous constitutive relations—serve to determine the effective anisotropic response of the composite under the prescribed loading conditions. Part II of this work is concerned with the application of the model to some specific classes of two-phase composite materials subjected to axisymmetric loading conditions.

1. Introduction

Over the past few years, various authors have proposed homogenization models to estimate the effective behaviour of linear composite materials. Among them, Hashin & Shtrikman (1963) made use of new variational principles to obtain rigorous bounds for the effective behaviour of composites with statistically homogeneous and isotropic distributions of linear elastic phases. Willis (1977) generalized these results by introducing the statistical notion of 'ellipsoidal symmetry' and later showed (Willis 1978) that his earlier results could be identified with specific types of particulate microstructures. Thus, Willis obtained estimates for composites whose microstructure consists of identically shaped ellipsoidal inclusions distributed in such a way that the two-point correlation functions characterizing the relative positions of the centres of the inclusions are also ellipsoidal with the same shape as the inclusions. Recently, Ponte Castañeda & Willis (1995) made use of this early work to give explicit estimates for the overall behaviour of linear composites consisting of ellipsoidal inclusions with pair distribution functions that are also ellipsoidal in shape, but with aspect ratios that are generally different from those of the inclusions.

Among several models proposed to estimate the effective behaviour of nonlinear composite materials, Ponte Castañeda (1991) (see also Talbot & Willis 1992; Suquet 1993) introduced a variational representation by means of which estimates for the effective behaviour of nonlinear composites can be generated from corresponding estimates for the effective behaviour of 'linear comparison composites' with the same microstructures as the nonlinear composites. In particular, this suggests that the new linear estimates of Ponte Castañeda & Willis (1995) may be used in conjunction with the aforementioned variational representation to obtain estimates for the effective behaviour of nonlinear composites with general ellipsoidal particulate microstructures. It should be mentioned that an alternative approach would be to make use of the nonlinear Hashin–Shtrikman variational principles of Talbot & Willis (1985) (see also Ponte Castañeda & Willis 1988; Willis 1991). However, for the class of nonlinear material models of interest here, it can be shown that the same estimates would result from both approaches. Therefore, given that the linear estimates are already available from the work of Ponte Castañeda & Willis (1995), it is perhaps simplest to make use of the variational procedure of Ponte Castañeda (1991) to obtain the corresponding estimates for the nonlinear composites; this is the approach that will be taken in this work.

When composites are subjected to finite deformation, their microstructure changes continuously and consequently their effective response is affected. In particular, an initially isotropic composite may develop strongly anisotropic behaviour under appropriately chosen loading conditions. For example, a steel reinforced by an isotropic distribution of hard spherical particles would develop significant anisotropies if it was subjected to a hot-rolling operation in order to generate a large extension in the rolling direction coupled with a large reduction in thickness. The usual homogenization models, including the ones described above, do not normally take into account this important feature, implicitly assuming that the microstructure remains frozen during the deformation process. Recently, however, Ponte Castañeda & Zaidman (1994) (see also Zaidman & Ponte Castañeda 1996) proposed constitutive models which are capable of accounting for the evolution of the microstructure in porous and two-phase composites with particulate microstructures and nonlinearly viscous constitutive behaviour. These models make use of the variational representation of Ponte Castañeda to obtain estimates for the 'instantaneous' effective constitutive response, as well as for the average strain-rate in the phases of the nonlinear composites. In addition, appropriate internal variables were identified to characterize the state of the microstructure and evolution laws for these variables were proposed. Thus, the volume fraction (characterizing the average size) and the aspect ratios (characterizing the average shape) of the inclusions were introduced as the relevant microstructural variables and kinematically based evolution laws were prescribed for these variables. This was carried out under general triaxial loading conditions, with the assumption that the loading axes remain aligned with the material symmetry axes of the composite, in order to preclude the (average) rotation of the inclusions. Furthermore, the model involved the assumption that the spatial distribution of the inclusions exhibit the same 'ellipsoidal symmetry' as the shape of the inclusions at every instant in the deformation process, thus permitting the use of the results of Willis (1978) to estimate the effective response of the linear comparison composites.

In this work, we shall extend the model of Ponte Castañeda & Zaidman by treating the spatial distribution of the inclusions as an independent microstructural variable. Thus, the relevant microstructural variables, in the context of the improved model, are taken to be the aspect ratios of the (ellipsoidal) pair-distribution function for the inclusions, as well as the volume fraction and aspect ratios of the (also ellipsoidal) inclusions. The new estimates of Ponte Castañeda & Willis will be used in conjunction with the variational procedure of Ponte Castañeda to obtain the instantaneous constitutive relations for the nonlinear composite. Evolution laws will then be developed for the relevant microstructural variables, which will be integrated simultaneously with the instantaneous constitutive relations for the composite in order to describe the effective anisotropic response of the nonlinear composites under triaxial loading conditions (with fixed axes). In part II of this work (Kailasam *et al.* 1997), the behaviour of specific composites subjected to axisymmetric loading conditions will be considered in some detail. In particular, the case of composites with initially isotropic distributions of rigid inclusions of spherical shape will be studied in an attempt to isolate the effects of changes in the pair-distribution function of the inclusions from the effects of corresponding changes in the size and shape of the inclusions, which was the focus of the earlier work of Ponte Castañeda & Zaidman (1994).

The ultimate goal of this work is to develop simple constitutive models capable of describing the anisotropic and non-uniform development of microstructure in composite materials subjected to standard forming techniques such as forging, rolling and extrusion. Experimental evidence that the evolution of the microstructure affects the overall response of the materials can be found, for example, in the work of Spitzig *et al.* (1988) on iron compacts.

2. Effective properties of composite materials

In this work, a composite is defined as a heterogeneous material with two distinct length scales; a macroscopic one characterizing the overall dimensions of the specimen and the scale of variation of the applied loading conditions and a microscopic one characterizing the size of the typical heterogeneity (e.g. inclusions). By effective properties, we mean the relation between the averages of the local stress and strain-rate fields within the composite.

The local behaviour of the composite is assumed to be nonlinearly viscous and is governed by a stress potential U, through the relation

$$\boldsymbol{D} = \frac{\partial U}{\partial \boldsymbol{\sigma}}(\boldsymbol{x}, \boldsymbol{\sigma}), \tag{2.1}$$

where $\boldsymbol{\sigma}$ and $\boldsymbol{D}$ represent the Cauchy stress and the Eulerian strain rate, respectively. For a composite with two phases, this potential can be expressed in the form

$$U(\boldsymbol{x}, \boldsymbol{\sigma}) = \sum_{r=1}^{2} \chi^{(r)}(\boldsymbol{x}) U^{(r)}(\boldsymbol{\sigma}), \tag{2.2}$$

where $U^{(r)}$ and $\chi^{(r)}$ denote the stress potential and the characteristic function (equal to 1 if $\boldsymbol{x}$ is in phase r, and 0 otherwise) of phase r ($r = 1, 2$), respectively. The effective behaviour of the composite is then defined by the relation (Hill 1963)

$$\bar{\boldsymbol{D}} = \frac{\partial \tilde{U}}{\partial \bar{\boldsymbol{\sigma}}}(\bar{\boldsymbol{\sigma}}), \tag{2.3}$$

where $\bar{\boldsymbol{D}}$ is the average strain rate in the composite, $\bar{\boldsymbol{\sigma}}$ is the average stress and $\tilde{U}$ denotes the effective potential of the composite. The effective potential may be obtained from the variational representation

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \min_{\boldsymbol{\sigma}\in S(\bar{\boldsymbol{\sigma}})} \int_{\Omega} U(\boldsymbol{x}, \boldsymbol{\sigma})\, \mathrm{d}v, \tag{2.4}$$

where

$$S(\bar{\boldsymbol{\sigma}}) = \{\boldsymbol{\sigma} | \nabla \cdot \boldsymbol{\sigma} = 0 \text{ in } \Omega \text{ and } \boldsymbol{\sigma n} = \bar{\boldsymbol{\sigma}}\boldsymbol{n} \text{ on } \partial\Omega\}$$

is the set of statically admissible stress fields corresponding to the applied uniform stress $\bar{\boldsymbol{\sigma}}$ on the boundary.

In this work, we consider the nonlinear phases to be isotropic with potentials

$$U^{(r)}(\boldsymbol{\sigma}) = \phi^{(r)}(\sigma_{\mathrm{e}}) + \frac{1}{2k^{(r)}}\sigma_{\mathrm{m}}^2, \tag{2.5}$$

where $\sigma_{\mathrm{e}} = \frac{3}{2}\boldsymbol{\sigma}' \cdot \boldsymbol{\sigma}'$ is the equivalent stress ($\boldsymbol{\sigma}'$ is the deviatoric stress), $\sigma_{\mathrm{m}} = \frac{1}{3}(\mathrm{tr}\,\boldsymbol{\sigma})$ is the hydrostatic stress and $k^{(r)}$ is the bulk viscosity of phase r. The functions $\phi^{(r)}$, which are assumed to be convex in σ_{e}, may be taken to have the power-law form

$$\phi^{(r)}(\sigma_{\mathrm{e}}) = \frac{\sigma_y^{(r)}}{n^{(r)}+1}\left[\frac{\sigma_{\mathrm{e}}}{\sigma_y^{(r)}}\right]^{n^{(r)}+1} = \frac{1}{3(n^{(r)}+1)\mu^{(r)}}\sigma_{\mathrm{e}}^{n^{(r)}+1}, \tag{2.6}$$

which is commonly used in high-temperature creep. In the above relation, $n^{(r)}$ and $\sigma_y^{(r)} = (3\mu^{(r)})^{1/n^{(r)}}$ are the creep exponent and the reference stress of phase r, respectively.

In the expression for $\phi^{(r)}$, $n^{(r)} = 1$ corresponds to linearly viscous behaviour which is governed by the viscosity coefficient $\mu^{(r)}$. We then write the constitutive relation of phase r in the form

$$\boldsymbol{\sigma} = \boldsymbol{L}^{(r)}\boldsymbol{D}, \tag{2.7}$$

where $\boldsymbol{L}^{(r)} = (3k^{(r)}, 2\mu^{(r)})$ is the viscosity tensor for phase r, in the sense of Hill (1965). We note that the bulk viscosity is introduced in order to be able to consider the limit of vacuous phases, in which case $\boldsymbol{L}^{(r)} = (0,0)$; however, non-vacuous phases will be taken to be incompressible so that $\boldsymbol{L}^{(r)} = (\infty, 2\mu^{(r)})$. The effective potential of the composite may then be expressed as

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \tfrac{1}{2}\bar{\boldsymbol{\sigma}} \cdot (\tilde{\boldsymbol{M}}\bar{\boldsymbol{\sigma}}), \tag{2.8}$$

where $\tilde{\boldsymbol{M}}$ is the effective viscous compliance tensor. Making use of relation (2.3), we obtain the following relation between the average stress and the average strain rate in the composite:

$$\bar{\boldsymbol{\sigma}} = \tilde{\boldsymbol{L}}\bar{\boldsymbol{D}}, \tag{2.9}$$

where $\tilde{\boldsymbol{L}}$ is the effective viscosity tensor of the composite and is related to the effective viscous compliance tensor through the relation $\tilde{\boldsymbol{M}} = \tilde{\boldsymbol{L}}^{-1}$.

When phase r is taken to be incompressible and $n^{(r)} \to \infty$, the expression for $\phi^{(r)}$ corresponds to rigid–perfectly plastic behaviour of the von Mises type, with tensile

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yield stress $\sigma_y^{(r)}$. The function $\phi^{(r)}$ then takes the form

$$\phi^{(r)}(\sigma_\mathrm{e}) = \begin{cases} 0, & \text{if } \sigma_\mathrm{e} \leqslant \sigma_y^{(r)}, \\ \infty, & \text{otherwise,} \end{cases} \tag{2.10}$$

and relation (2.1) for the local behaviour of the composite is replaced by the normality condition

$$\boldsymbol{D} = \dot{\lambda}\boldsymbol{\sigma}', \tag{2.11}$$

where $\dot{\lambda}$ is the (non-negative) plastic loading parameter. In order to describe the effective behaviour of composites made of such phases, it is convenient to introduce an effective yield domain $\tilde{P}$ (Suquet 1983), such that

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \begin{cases} 0, & \text{if } \bar{\boldsymbol{\sigma}} \in \tilde{P}, \\ \infty, & \text{otherwise.} \end{cases} \tag{2.12}$$

The boundary of $\tilde{P}$ then defines an effective yield function

$$\tilde{\Phi}(\bar{\boldsymbol{\sigma}}) = 0, \tag{2.13}$$

in terms of which we have (Hill 1967)

$$\bar{\boldsymbol{D}} = \dot{\Lambda}\frac{\partial \tilde{\Phi}}{\partial \bar{\boldsymbol{\sigma}}}(\bar{\boldsymbol{\sigma}}), \tag{2.14}$$

where $\dot{\Lambda}$ is a non-negative parameter to be determined from the consistency condition.

3. Linearly viscous composites

(*a*) *Effective constitutive relations*

In this work, we make use of the Hashin–Shtrikman estimates of Ponte Castañeda & Willis (1995) for the effective viscosity tensor $\tilde{\boldsymbol{L}}$ of linearly viscous (mathematically analogous to linearly elastic) composites with particulate microstructures consisting of random 'ellipsoidal' distributions of ellipsoidal inclusions of one material (denoted by the superscript 2) in a matrix of a different material (denoted 1). Referring to figure 1, the aspect ratios of the inclusions are defined by $w_1^\mathrm{i} = l_3/l_1$ and $w_2^\mathrm{i} = l_3/l_2$ (see figure 1*a*), whereas those of the distribution function are defined by $w_1^\mathrm{d} = L_3/L_1$ and $w_2^\mathrm{d} = L_3/L_2$ (see figure 1*b*). (Note that the microstructure in figure 1*b* has been depicted as being periodic, for ease of visualization only.)

These estimates for the effective viscosity of the composite can be written in terms of the strain-rate concentration tensors $\boldsymbol{A}^{(r)}$ $(r = 1, 2)$, which are such that the average strain rate in phase r is given by $\boldsymbol{D}^{(r)} = \boldsymbol{A}^{(r)}\bar{\boldsymbol{D}}$ (Hill 1965), via the expression

$$\tilde{\boldsymbol{L}} = \sum_{r=1}^{2} c^{(r)}\boldsymbol{L}^{(r)}\boldsymbol{A}^{(r)}, \tag{3.1}$$

where the expression for $\boldsymbol{A}^{(2)}$, obtained from the Hashin–Shtrikman procedure (see Ponte Castañeda & Willis 1995), can be written as

$$\boldsymbol{A}^{(2)} = [\boldsymbol{I} + (\boldsymbol{P}^\mathrm{i} - c^{(2)}\boldsymbol{P}^\mathrm{d})(\boldsymbol{L}^{(2)} - \boldsymbol{L}^{(1)})]^{-1}, \tag{3.2}$$

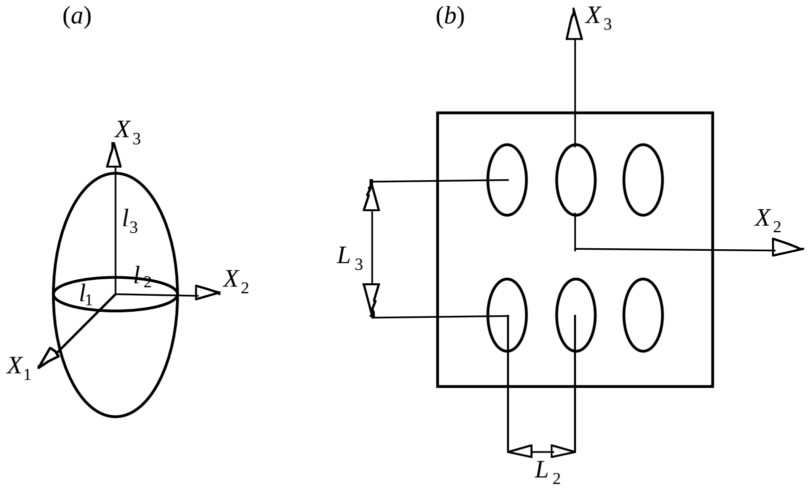

Figure 1. Inclusion (void) geometry and distribution. (*a*) Ellipsoidal inclusion (void) with aspect ratios, $w_1^{\rm i} = l_3/l_1$ and $w_2^{\rm i} = l_3/l_2$. (*b*) Ellipsoidal distribution of the inclusions with centres positioned such that the aspect ratio $w_2^{\rm d} = L_3/L_2$ and also $w_1^{\rm d} = L_3/L_1$ (not shown).

and $\boldsymbol{A}^{(1)}$ is such that

$$c^{(1)}\boldsymbol{A}^{(1)} + c^{(2)}\boldsymbol{A}^{(2)} = \boldsymbol{I}. \tag{3.3}$$

In the above expressions, $c^{(1)}$ and $c^{(2)}$ are the volume fractions of the matrix and the inclusions, respectively, $\boldsymbol{I}$ is the fourth-order identity tensor, $\boldsymbol{P}^{\rm i}$ and $\boldsymbol{P}^{\rm d}$ are geometric tensors associated with the inclusions (which are all identical in shape) and their spatial distributions, respectively. Both $\boldsymbol{P}^{\rm i}$ and $\boldsymbol{P}^{\rm d}$ depend on the properties of the matrix and, in addition, $\boldsymbol{P}^{\rm i}$ depends on the aspect ratios of the inclusions while $\boldsymbol{P}^{\rm d}$ depends on the aspect ratios of the spatial distribution of the inclusions. We note that the $\boldsymbol{P}$ tensors are related to the Eshelby (1957) tensors $\boldsymbol{S}$, corresponding to an isolated inclusion embedded in a matrix of phase 1, through the relation $\boldsymbol{P} = \boldsymbol{S}\boldsymbol{M}^{(1)}$. The corresponding expression for $\tilde{\boldsymbol{M}}$, obtained from (3.1) upon simplification, is given by

$$\tilde{\boldsymbol{M}} = \{\boldsymbol{I} + c^{(2)}[(\boldsymbol{M}^{(1)}\boldsymbol{L}^{(2)} - \boldsymbol{I})^{-1} + (\boldsymbol{S}^{\rm i} - c^{(2)}\boldsymbol{S}^{\rm d})]^{-1}\}^{-1}\boldsymbol{M}^{(1)}. \tag{3.4}$$

We observe that when $\boldsymbol{P}^{\rm i} = \boldsymbol{P}^{\rm d} = \boldsymbol{P}$ (or $\boldsymbol{S}^{\rm i} = \boldsymbol{S}^{\rm d} = \boldsymbol{S}$), the above expressions for $\tilde{\boldsymbol{M}}$ and $\boldsymbol{A}^{(2)}$ reduce to the expressions of Willis (1977, 1978). If, in addition, we consider the case where the inclusions are present in dilute concentrations ($c^{(2)} \rightarrow 0$), the expressions are in agreement with the well-known estimates of Eshelby (1957). Note that the distribution effects, as characterized by $\boldsymbol{P}^{\rm d}$ or $\boldsymbol{S}^{\rm d}$, become negligible in this limiting case. We also note that if $\boldsymbol{L}^{(1)} > \boldsymbol{L}^{(2)}$ ($\boldsymbol{L}^{(1)} < \boldsymbol{L}^{(2)}$), in the sense of quadratic forms, the expressions for $\tilde{\boldsymbol{L}}$ and $\tilde{\boldsymbol{M}}$ can be interpreted as upper (lower) and lower (upper) bounds, respectively, for the class of microstructures described here. Finally, the estimates (3.1) or (3.4) are believed to be adequate for general particulate composites in the low to intermediate range of volume fractions, ensuring that, on the average, the inclusions are not too close to each other and strong interaction effects can be safely neglected.

(*b*) *Evolution of the microstructure under triaxial loading conditions*

If a composite is subjected to finite deformation, it is clear that its microstructure will not remain fixed but instead will change at every stage of the deformation process. However, to account for the evolution of the microstructure exactly is a very complicated problem and so certain simplifying assumptions will be made here to capture the essential features of the problem. The first such assumption is that the applied loading will be taken to be triaxial with axes coinciding with the material symmetry axes of the composite. This ensures that the inclusions do not change orientation during the deformation process (at least on the average). It should be emphasized that this assumption could be relaxed, which would involve keeping track of the (average) orientation of the inclusions throughout the deformation process (see Kailasam & Ponte Castañeda 1997).

For composites with dilute concentrations of inclusions subjected to uniform loading conditions, Eshelby (1957) has shown that the strain rate within the inclusions is uniform. This implies that an initially ellipsoidal inclusion will deform into ellipsoidal shapes with possibly different size, aspect ratios and orientations (the last of which, as we mentioned earlier, is not accounted for in this work). Ponte Castañeda & Zaidman (1994) (see also Zaidman & Ponte Castañeda 1996) have argued that even for composites with non-dilute concentrations of the inclusions, subjected to triaxial loading conditions, initially spherical inclusions may be assumed to deform, on the average, into ellipsoidal inclusions which are aligned with the loading axes. The difference in this case is that the average strain rate in the inclusions is determined by using the 'non-dilute' Hashin–Shtrikman expression (3.2) for the strain-rate concentration tensor instead of the corresponding dilute expression of Eshelby.

In the works of Ponte Castañeda & Zaidman (1994) and Zaidman & Ponte Castañeda (1996), the volume fraction and the aspect ratios of the inclusions were the only microstructural variables considered. This simplification was justified by the fact that the effect of the distribution of the inclusions is of second order, relative to the shape of the inclusions, which is of first order in the volume fraction of the inclusions. It should also be noted that the more general estimates of the form (3.4), which take into account the effect of the distribution, were not available until very recently. It was thus implicitly assumed that the inclusions and the distribution had the same aspect ratios through every stage of the deformation process, thereby allowing the use of the simplified estimates of Willis (1978). In the present work, we relax this assumption and treat the distribution of the inclusions as an independent variable, which will be allowed to evolve differently from the shape of the inclusions. Thus, in summary, the relevant internal variables which characterize the state of the microstructure will be taken to be the volume fraction of the inclusions $c^{(2)}$, the two aspect ratios of the ellipsoidal inclusions w_1^{i}, w_2^{i} and the two aspect ratios of the spatial distribution of the inclusions w_1^{d}, w_2^{d} (see figure 1).

Having identified the relevant microstructural variables, we next deduce appropriate evolution equations for these variables. An evolution law for the volume fraction of the inclusions is easily obtained from the kinematical relation

$$\dot{c}^{(2)} = c^{(1)}c^{(2)}(D_{kk}^{(2)} - D_{kk}^{(1)}). \tag{3.5}$$

When the matrix is incompressible, $D_{kk}^{(1)} = 0$ and therefore $D_{kk}^{(2)} = \bar{D}_{kk}/c^{(2)}$, so that the above evolution law takes the form

$$\dot{c}^{(2)} = c^{(1)}\bar{D}_{kk}. \tag{3.6}$$

The change in the aspect ratios of the inclusions is governed by the well-known kinematical relations

$$\dot{w}_1^{\mathrm{i}} = w_1^{\mathrm{i}}(D_{33}^{(2)} - D_{11}^{(2)}) \quad \text{and} \quad \dot{w}_2^{\mathrm{i}} = w_2^{\mathrm{i}}(D_{33}^{(2)} - D_{22}^{(2)}), \tag{3.7}$$

involving only the average strain rate in the inclusions $\boldsymbol{D}^{(2)} = \boldsymbol{A}^{(2)}\bar{\boldsymbol{D}}$, where the strain-rate concentration tensor $\boldsymbol{A}^{(2)}$ is given by equation (3.2). It is noted that $\boldsymbol{A}^{(2)}$ is, in general, a function of all the microstructural variables, as well as of the material properties of the inclusion and the matrix phases; that is, $\boldsymbol{A}^{(2)} = \boldsymbol{A}^{(2)}(c^{(2)}, w_1^{\mathrm{i}}, w_2^{\mathrm{i}}, w_1^{\mathrm{d}}, w_2^{\mathrm{d}}; \mu^{(1)}, \mu^{(2)})$.

Finally, the evolution of the aspect ratios of the spatial distribution of the inclusions is assumed to be controlled by the relations

$$\dot{w}_1^{\mathrm{d}} = w_1^{\mathrm{d}}(\bar{D}_{33} - \bar{D}_{11}) \quad \text{and} \quad \dot{w}_2^{\mathrm{d}} = w_2^{\mathrm{d}}(\bar{D}_{33} - \bar{D}_{22}), \tag{3.8}$$

which follow from the assumption that the inclusions are transported by the mean flow. That this should be the case has been demonstrated rigorously for composites with periodic microstructures. Thus, Levy & Sanchez Palencia (1983) have shown, through an asymptotic analysis of the problem, that the positions of rigid inclusions in a three-dimensional, periodic flow of an incompressible, viscous fluid is indeed controlled by the bulk flow (i.e. by $\boldsymbol{v} = \bar{\boldsymbol{D}}\boldsymbol{x}$). For composites with more general (random) microstructures, it is plausible that the same result should hold, even if the inclusions are allowed to deform.

The ordinary differential equations (3.6), (3.7) and (3.8), together with the effective constitutive equation (2.9), define the evolution of $c^{(2)}$, w_1^{i}, w_2^{i}, w_1^{d}, w_2^{d} and $\bar{\boldsymbol{D}}$ if $\boldsymbol{\sigma n} = \bar{\boldsymbol{\sigma}}\boldsymbol{n}$ is prescribed on the boundary (or, equivalently, of $\bar{\boldsymbol{\sigma}}$, if $\boldsymbol{v} = \bar{\boldsymbol{D}}\boldsymbol{x}$ is prescribed on the boundary). Note that the evolution of the microstructural variables may lead to the eventual overlap of the inclusions (and of their distributional ellipsoids), in which case the above model ceases to be valid (because the constitutive equation (2.9) would then no longer apply; see Ponte Castañeda & Willis (1995)). Because of the dependence of the microstructural variables on the average stress (or strain rate), it follows that the effective compliance and viscosity tensors will depend on the stress (or strain rate) and therefore the effective response of the two-phase, linearly viscous composites will be *nonlinear* and *anisotropic* (i.e. non-Newtonian). It is also noted that uniform boundary conditions have been assumed for the purpose of carrying out the homogenization procedure. Once this is done, however, the constitutive model can be used for general non-uniform boundary conditions provided that the scale of variation of the applied loading conditions is still large compared to the size of the typical heterogeneity. This would be the case, for example, in an extrusion process where the size of the dies is large compared to the size of the pores or inclusions in the material that is being extruded. For non-uniform boundary conditions, the equilibrium and compatibility conditions for the average stress $\bar{\boldsymbol{\sigma}}$ and the average strain rate $\bar{\boldsymbol{D}}$, respectively, must be taken together with the constitutive relations and the evolution equations for the now position-dependent microstructural variables. On the other hand, for uniform boundary conditions, the equilibrium and compatibility equations are automatically satisfied and hence the constitutive relations and the evolution equations for the microstructural variables are sufficient to describe the effective response of the composite.

4. Nonlinearly viscous composites

(*a*) *Effective constitutive relations*

In § 2, we saw that the effective behaviour of a composite is defined by the relation (2.3), together with the variational statement (2.4) for the effective potential of the composite. Ponte Castañeda (1991, 1992) has introduced an alternative variational representation by means of which estimates for the effective potential of nonlinear composites can be obtained from corresponding estimates for linearly viscous composites with the same microstructure as the nonlinear composites. Expressions for the effective compliance tensor of two-phase, linearly viscous composites were given in § 3 *a*. In this section, we demonstrate how these results may be utilized to obtain estimates for nonlinearly viscous composites.

For the class of two-phase composites, the variational statement of Ponte Castañeda gives the following estimate for the effective potential of the nonlinear composite:

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) \cong \max_{\mu^{(1)},\mu^{(2)}\geqslant 0}\left\{\tfrac{1}{2}\bar{\boldsymbol{\sigma}}\cdot(\tilde{\boldsymbol{M}}\bar{\boldsymbol{\sigma}}) - \sum_{r=1}^{2} c^{(r)}V^{(r)}(\mu^{(r)})\right\}, \tag{4.1}$$

where $\tilde{\boldsymbol{M}}$ denotes any estimate for the effective viscous compliance tensor for the two-phase linear composite with viscosities $\mu^{(1)}$ and $\mu^{(2)}$ prescribed in the volume fraction $c^{(1)}$ and $c^{(2)}$, respectively, and for fixed bulk viscosities $k^{(1)}$, $k^{(2)}$. In this expression, the functions $V^{(r)}$ are defined as

$$V^{(r)}(\mu^{(r)}) = \max_{\boldsymbol{\sigma}}\left\{\frac{1}{6\mu^{(r)}}\sigma_{\mathrm{e}}^2 - \phi^{(r)}(\sigma_{\mathrm{e}})\right\}. \tag{4.2}$$

Note that the estimate given by the right-hand side of (4.1) can be shown to be a rigorous lower bound, under appropriate circumstances (see Ponte Castañeda 1991).

The Hashin–Shtrikman estimates (3.4) that were obtained for the effective compliance tensor of the linearly viscous composites can now be used to develop estimates for the effective potential of the nonlinearly viscous composites. The optimal values of $\mu^{(1)}$ and $\mu^{(2)}$ in the estimate (4.1) for the potential are denoted by $\hat{\mu}^{(1)}$ and $\hat{\mu}^{(2)}$, respectively, and are determined by the relations

$$\tfrac{1}{2}\bar{\boldsymbol{\sigma}}\cdot\left[\frac{\partial\tilde{\boldsymbol{M}}}{\partial\mu^{(s)}}(\hat{\mu}^{(r)})\bar{\boldsymbol{\sigma}}\right] - c^{(s)}\frac{\partial V^{(s)}}{\partial\mu^{(s)}}(\hat{\mu}^{(s)}) = 0, \quad (s = 1, 2). \tag{4.3}$$

The effective constitutive response of the nonlinear composite may then be written (see Ponte Castañeda & Zaidman 1996) in the form

$$\bar{\boldsymbol{D}} = \tilde{\boldsymbol{M}}\bar{\boldsymbol{\sigma}}, \tag{4.4}$$

where $\tilde{\boldsymbol{M}} = \tilde{\boldsymbol{M}}(c^{(2)}, w_1^{\mathrm{i}}, w_2^{\mathrm{i}}, w_1^{\mathrm{d}}, w_2^{\mathrm{d}}; \hat{\mu}^{(1)}, \hat{\mu}^{(2)})$. It is emphasized that this relation, despite its appearance, is nonlinear because of the dependence of $\hat{\mu}^{(1)}$ and $\hat{\mu}^{(2)}$ on $\bar{\boldsymbol{\sigma}}$ (directly) and on the microstructural variables (indirectly). Relation (4.4) suggests that the nonlinearly viscous composites may be considered as linearly viscous composites with stress-dependent viscosities. Later, we will make use of estimates (3.4) for the effective compliance of two-phase linearly viscous composites to obtain effective constitutive equations for the nonlinearly viscous and perfectly plastic, two-phase composites.

(*b*) *Evolution of the microstructure under triaxial loading conditions*

The evolution law for the volume fraction of the inclusions (3.6) is a purely kinematical relation and hence remains unchanged for nonlinear constitutive behaviour. Ponte Castañeda & Zaidman (1994) have argued that equations (3.7) can also be used to determine the (average) change in the shape of the inclusions for the nonlinear composites, provided that appropriate expressions are used for the average strain rate in the inclusions. As we have seen, for the linearly viscous composites, the average strain rate in the inclusions can be calculated from the strain-rate concentration tensor through the relation $\boldsymbol{D}^{(2)} = \boldsymbol{A}^{(2)}\bar{\boldsymbol{D}}$. It was also mentioned in the previous section that the nonlinearly viscous composites may be thought of as linearly viscous composites with viscosities that depend on the average stress (and the state of the microstructure). This suggests that the average strain-rate in the inclusions of the nonlinearly viscous composites can be obtained from $\boldsymbol{D}^{(2)} = \boldsymbol{A}^{(2)}\bar{\boldsymbol{D}}$ with expression (3.2) for $\boldsymbol{A}^{(2)}$, evaluated at the optimal values of the viscosities $\hat{\mu}^{(1)}$ and $\hat{\mu}^{(2)}$ resulting from (4.3). Then, the evolution equations for the aspect ratios of the inclusions in the nonlinearly viscous composites are given by (3.7) with the only difference being that the 'nonlinear' strain-rate concentration tensor $\boldsymbol{A}^{(2)}$ is to be used instead of the 'linear' one. On the other hand, equation (3.8) for the evolution of the distributional aspect ratios does not depend on any of the material parameters, being purely kinematical in nature. This allows the use of the same expression (3.8) for the evolution of the distribution of the inclusions in the nonlinearly viscous composites also. As a partial test of the proposed constitutive model, some comparisons with numerical simulations of porous materials with periodic microstructures will be given in part II of this work.

5. Application to linearly viscous composites and axisymmetric loading conditions

(a) *Porous composites*

In this section, we specialize the above results for general two-phase linear composites to porous composites whose microstructure initially consists of an isotropic distribution of spherical voids in a linearly viscous, incompressible matrix. Expressions (3.2) and (3.4) for the strain-rate concentration tensor and the effective compliance tensor can be simplified by noting that $\boldsymbol{L}^{(2)} \equiv \boldsymbol{0}$. We then have that

$$\boldsymbol{A}^{(2)} = [\boldsymbol{I} - (\boldsymbol{P}^{\mathrm{i}} - c^{(2)}\boldsymbol{P}^{\mathrm{d}})\boldsymbol{L}^{(1)}]^{-1} \tag{5.1}$$

and

$$\tilde{\boldsymbol{M}} = [\boldsymbol{I} + c^{(2)}(\boldsymbol{S}^{\mathrm{i}} - c^{(2)}\boldsymbol{S}^{\mathrm{d}} - \boldsymbol{I})^{-1}]^{-1}\boldsymbol{M}^{(1)}, \tag{5.2}$$

which, using the fact that $\boldsymbol{M}^{(1)} = (0, 1/(2\mu^{(1)}))$, may be rewritten as

$$\tilde{\boldsymbol{M}} = \frac{1}{3\mu^{(1)}}\tilde{\boldsymbol{m}}. \tag{5.3}$$

We then note that, in this case, both $\boldsymbol{A}^{(2)}$ and $\tilde{\boldsymbol{m}}$ are independent of the material properties of the phases, which allows us to express them in the form

$$\boldsymbol{A}^{(2)} = \boldsymbol{A}^{(2)}(c^{(2)}, w_1^{\mathrm{i}}, w_2^{\mathrm{i}}, w_1^{\mathrm{d}}, w_2^{\mathrm{d}}) \quad \text{and} \quad \tilde{\boldsymbol{m}} = \tilde{\boldsymbol{m}}(c^{(2)}, w_1^{\mathrm{i}}, w_2^{\mathrm{i}}, w_1^{\mathrm{d}}, w_2^{\mathrm{d}}). \tag{5.4}$$

As the composite undergoes deformation, the microstructural variables evolve and hence both $\boldsymbol{A}^{(2)}$ and $\tilde{\boldsymbol{M}}$ change at every stage of the deformation process. We also note, for future reference, that the effective potential of the linearly viscous composite may be written in the form

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \frac{1}{6\mu^{(1)}}\bar{\boldsymbol{\sigma}} \cdot (\tilde{\boldsymbol{m}}\bar{\boldsymbol{\sigma}}). \tag{5.5}$$

When a composite with an initially isotropic distribution of spherical voids is subjected to axisymmetric loading conditions, the voids and their distribution both

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become spheroidal ($w_1^{\mathrm{i}} = w_2^{\mathrm{i}} = w^{\mathrm{i}}$ and $w_1^{\mathrm{d}} = w_2^{\mathrm{d}} = w^{\mathrm{d}}$) and the composite exhibits transverse isotropy. It is then noted that, in this case, we have only three microstructural variables to deal with: $c^{(2)}$, w^{i} and w^{d}. Due to the transversely isotropic nature of $\boldsymbol{A}^{(2)}$ and $\tilde{\boldsymbol{m}}$, they can conveniently be expressed (Walpole 1981) as $\tilde{\boldsymbol{m}} = (m_1, m_2, m_3, m_4, m_5, m_6)$ and $\boldsymbol{A}^{(2)} = (a_1, a_2, a_3, a_4, a_5, a_6)$, where the expressions for the m_i and a_i are dependent on the relevant microstructural variables ($c^{(2)}$, w^{i} and w^{d}) and are given in the appendix. The effective potential of the composite is given by (5.5) with

$$\bar{\boldsymbol{\sigma}} \cdot (\tilde{\boldsymbol{m}}\bar{\boldsymbol{\sigma}}) = m_1\bar{\sigma}_{\mathrm{p}}^2 + \tfrac{1}{2}m_2\bar{\sigma}_{\mathrm{n}}^2 + m_3\bar{\tau}_{\mathrm{p}}^2 + m_4\bar{\tau}_{\mathrm{n}}^2 + 2m_5\bar{\sigma}_{\mathrm{p}}\bar{\sigma}_{\mathrm{n}}, \tag{5.6}$$

where $\bar{\sigma}_{\mathrm{p}} = \frac{1}{2}[\bar{\sigma}_{11} + \bar{\sigma}_{22}]$, $\bar{\sigma}_{\mathrm{n}} = \bar{\sigma}_{33}$, $\bar{\tau}_{\mathrm{p}} = [\bar{\sigma}_{12}^2 + \frac{1}{4}(\bar{\sigma}_{11} - \bar{\sigma}_{22})^2]^{1/2}$, $\bar{\tau}_{\mathrm{n}} = [\bar{\sigma}_{13}^2 + \bar{\sigma}_{23}^2]^{1/2}$ (see figure 1) are an appropriate set of transversely isotropic invariants of $\bar{\boldsymbol{\sigma}}$, corresponding to in-plane hydrostatic tensile, normal tensile, transverse shear and longitudinal shear stresses, respectively (see deBotton & Ponte Castañeda 1992). The instantaneous constitutive relation for the composite is then obtained from relation (2.3).

The evolution equations for the relevant microstructural variables given in § 3 *b* can be simplified using the fact that, for axisymmetric loading, $\bar{D}_{11} = \bar{D}_{22}$. This results in the following law for the evolution of the porosity $c^{(2)}$:

$$c^{(2)} = (1 - c^{(2)})(2\bar{D}_{11} + \bar{D}_{33}), \tag{5.7}$$

while those for the two aspect ratios are given by

$$\dot{w}^{\mathrm{i}} = w^{\mathrm{i}}[(2a_5 - a_1)\bar{D}_{11} + (a_2 - a_6)\bar{D}_{33}] \tag{5.8}$$

and

$$\dot{w}^{\mathrm{d}} = w^{\mathrm{d}}[\bar{D}_{33} - \bar{D}_{11}]. \tag{5.9}$$

(*b*) *Two-phase incompressible composites*

Here, we specialize the relations for the effective compliance tensor (3.4) and the evolution laws for the microstructural variables to the case where the microstructure of the composite initially consists of an isotropic distribution of incompressible, spherical inclusions in an incompressible matrix. Here again, we may write $\tilde{\boldsymbol{m}} = (3\mu^{(1)})\tilde{\boldsymbol{M}}$, so that expression (5.5) for the effective potential still holds. However, here, unlike in the case of the porous composites, $\boldsymbol{A}^{(2)}$ and $\tilde{\boldsymbol{m}}$ both depend on the properties of the inclusion and matrix materials through the ratio $y = \mu^{(2)}/\mu^{(1)}$. Also, in this case, the proportions of the two phases remain fixed due to incompressibility, so that $c^{(2)}$ will be taken to be a constant. This allows us to express $\boldsymbol{A}^{(2)}$ and $\tilde{\boldsymbol{m}}$ as

$$\boldsymbol{A}^{(2)} = \boldsymbol{A}^{(2)}(w_1^{\mathrm{i}}, w_2^{\mathrm{i}}, w_1^{\mathrm{d}}, w_2^{\mathrm{d}}; y) \quad \text{and} \quad \tilde{\boldsymbol{m}} = \tilde{\boldsymbol{m}}(w_1^{\mathrm{i}}, w_2^{\mathrm{i}}, w_1^{\mathrm{d}}, w_2^{\mathrm{d}}; y). \tag{5.10}$$

If, in addition, we consider axisymmetric loading, the microstructure will evolve in such a way that the composite exhibits transverse isotropy (as discussed in the previous section). In this case, the only relevant microstructural variables are w^{i} and w^{d} ($c^{(2)}$ is a constant). As mentioned earlier, for transversely isotropic symmetry, we can use the representation $\tilde{\boldsymbol{m}} = (m_1, m_2, m_3, m_4, m_5, m_6)$ and $\boldsymbol{A}^{(2)} = (a_1, a_2, a_3, a_4, a_5, a_6)$, where the explicit expressions for m_i and a_i ($i = 1, \ldots, 6$) (which are dependent on w^{i}, w^{d} and y) are given in the appendix. This allows us to obtain the effective potential for the composite from expression (5.5) using the following relation:

$$\bar{\boldsymbol{\sigma}} \cdot (\tilde{\boldsymbol{m}}\bar{\boldsymbol{\sigma}}) = 3m_1\bar{\tau}_{\mathrm{d}}^2 + m_3\bar{\tau}_{\mathrm{p}}^2 + m_4\bar{\tau}_{\mathrm{n}}^2, \tag{5.11}$$

where $\bar{\tau}_{\rm d} = (1/\sqrt{3})[\bar{\sigma}_{33} - \frac{1}{2}(\bar{\sigma}_{11} + \bar{\sigma}_{22})]$, $\bar{\tau}_{\rm p}$ and $\bar{\tau}_{\rm n}$ are the quadratic, incompressible, transversely isotropic invariants of $\bar{\boldsymbol{\sigma}}$ corresponding to the axisymmetric shear stress, the transverse shear stress and the longitudinal shear stress.

As mentioned earlier, we have only two microstructural variables to contend with; $w^{\rm i}$ and $w^{\rm d}$, the evolution equations for which are simplified by fact that $\bar{D}_{11} = \bar{D}_{22} = -\frac{1}{2}\bar{D}_{33}$ and are given by

$$\dot{w}^{\rm i} = w^{\rm i}[\tfrac{1}{2}a_1 + a_2 - a_5 - a_6]\bar{D}_{33} \tag{5.12}$$

and

$$\dot{w}^{\rm d} = \tfrac{3}{2}w^{\rm d}\bar{D}_{33}. \tag{5.13}$$

6. Application to nonlinearly viscous composites and axisymmetric loading

(*a*) *Porous composites*

In this section, we allow the inclusion phase to be vacuous so that

$$U^{(2)}(\boldsymbol{\sigma}) = \begin{cases} 0, & \text{if } \boldsymbol{\sigma} = \boldsymbol{0}, \\ \infty, & \text{otherwise}, \end{cases} \tag{6.1}$$

and the matrix to be nonlinearly viscous and incompressible with a potential defined by (2.5). Upon utilizing expression (5.2) for the effective viscous compliance tensor of the linearly viscous, porous composite in the variational representation (4.1), we obtain the following estimate for the effective potential of the nonlinearly viscous, porous composite:

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) \cong c^{(1)}\phi^{(1)}\left\{\left[\frac{\bar{\boldsymbol{\sigma}}\cdot(\tilde{\boldsymbol{m}}\bar{\boldsymbol{\sigma}})}{c^{(1)}}\right]^{1/2}\right\}, \tag{6.2}$$

where $\tilde{\boldsymbol{m}} = (3\mu^{(1)})\tilde{\boldsymbol{M}}$ depends on all the microstructural variables identified in § 5 *a* and is independent of material properties. Special cases of this result were given by Ponte Castañeda (1991), Willis (1991), Suquet (1992) and Talbot & Willis (1992). This result may be specialized further for power-law materials; in the limiting case of rigid–perfectly plastic behaviour, we obtain the following expression for the effective yield function of the composite:

$$\tilde{\Phi}(\bar{\boldsymbol{\sigma}}) = \frac{\bar{\boldsymbol{\sigma}}\cdot(\tilde{\boldsymbol{m}}\bar{\boldsymbol{\sigma}})}{c^{(1)}} - (\sigma_y^{(1)})^2. \tag{6.3}$$

The instantaneous constitutive equation for the composite is then obtained through the use of expression (2.14). It is noted that $\dot{\Lambda}$ is obtained from the consistency condition

$$\dot{\tilde{\Phi}} = 0$$

and depends on the evolution of the microstructure.

We shall consider a composite whose microstructure initially consists of an isotropic distribution of spherical voids and is subjected to triaxial loading conditions. Then, the relevant microstructural variables are $c^{(2)}$, $w_1^{\rm i}$, $w_2^{\rm i}$, $w_1^{\rm d}$ and $w_2^{\rm d}$. As discussed in § 4 *b*, we can make use of the equations (3.6), (3.7) and (3.8) for the evolution of these variables for nonlinearly viscous composites, provided the optimal values, $\hat{\mu}^{(1)}$ and $\hat{\mu}^{(2)}$, of the viscosities are utilized to obtain the relevant strain-rate

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concentration tensors $\boldsymbol{A}^{(2)}$. It was also mentioned in § 5 *a* that the strain-rate concentration tensors for porous composites are independent of the matrix properties. This has the implication that the evolution equations for the microstructural variables are identical for the linear and the nonlinear porous composites. It is emphasized, however, that these evolution equations must be solved in conjunction with the instantaneous constitutive equations and therefore the evolution of the microstructure will depend on the material nonlinearity, in general.

We now proceed to determine $\dot{\Lambda}$ from the consistency condition requiring that

$$\dot{\tilde{\Phi}} = \frac{\partial\tilde{\Phi}}{\partial\bar{\sigma}_{ij}}\dot{\bar{\sigma}}_{ij} + \frac{\partial\tilde{\Phi}}{\partial c^{(2)}}\dot{c}^{(2)} + \frac{\partial\tilde{\Phi}}{\partial w_1^{\mathrm{i}}}\dot{w}_1^{\mathrm{i}} + \frac{\partial\tilde{\Phi}}{\partial w_2^{\mathrm{i}}}\dot{w}_2^{\mathrm{i}} + \frac{\partial\tilde{\Phi}}{\partial w_1^{\mathrm{d}}}\dot{w}_1^{\mathrm{d}} + \frac{\partial\tilde{\Phi}}{\partial w_2^{\mathrm{d}}}\dot{w}_2^{\mathrm{d}} = 0. \tag{6.4}$$

Here, we have made use of the fact that, for the loading conditions considered, the principal axes of the applied stress $\bar{\boldsymbol{\sigma}}$ do not rotate, and hence we do not distinguish between the Jaumann and the standard time derivatives. The evolution equations for the microstructural variables are now used in the above expression to obtain the following expression for the instantaneous behaviour of the composite:

$$\bar{D}_{ij} = \frac{1}{H}\frac{\partial\tilde{\Phi}}{\partial\bar{\sigma}_{ij}}\frac{\partial\tilde{\Phi}}{\partial\bar{\sigma}_{kl}}\dot{\bar{\sigma}}_{kl}, \tag{6.5}$$

where H is the effective hardening rate and is given by

$$H = -\left[(1-c^{(2)})\frac{\partial\tilde{\Phi}}{\partial c^{(2)}}\frac{\partial\tilde{\Phi}}{\partial\bar{\sigma}_{kk}} + w_1^{\mathrm{i}}\frac{\partial\tilde{\Phi}}{\partial w_1^{\mathrm{i}}}(A_{33ij}^{(2)} - A_{11ij}^{(2)})\frac{\partial\tilde{\Phi}}{\partial\bar{\sigma}_{ij}} + w_2^{\mathrm{i}}\frac{\partial\tilde{\Phi}}{\partial w_2^{\mathrm{i}}}(A_{33ij}^{(2)} - A_{22ij}^{(2)})\frac{\partial\tilde{\Phi}}{\partial\bar{\sigma}_{ij}} \right.$$
$$\left. + w_1^{\mathrm{d}}\frac{\partial\tilde{\Phi}}{\partial w_1^{\mathrm{d}}}\left(\frac{\partial\tilde{\Phi}}{\partial\bar{\sigma}_{33}} - \frac{\partial\tilde{\Phi}}{\partial\bar{\sigma}_{11}}\right) + w_2^{\mathrm{d}}\frac{\partial\tilde{\Phi}}{\partial w_2^{\mathrm{d}}}\left(\frac{\partial\tilde{\Phi}}{\partial\bar{\sigma}_{33}} - \frac{\partial\tilde{\Phi}}{\partial\bar{\sigma}_{22}}\right)\right]. \tag{6.6}$$

We emphasize that even though the matrix phase has been assumed to be perfectly plastic in the derivation of (6.6), the effective hardening rate for the porous composite is seen to be non-vanishing ($H \neq 0$), in general, corresponding to non-perfectly plastic behaviour for the composite. Of course, as pointed out earlier in the context of the linearly viscous composites, this is a direct consequence of the finite changes in geometry associated with the evolution of the microstructure.

For the case where the composite is subjected to axisymmetric loads, we have already mentioned that the composite exhibits transverse isotropy. The corresponding yield function for this case can be obtained from (6.3), where $\bar{\boldsymbol{\sigma}} \cdot (\tilde{\boldsymbol{m}}\bar{\boldsymbol{\sigma}})$ is given by expression (5.6). Also, the expression for the hardening rate can be simplified by observing that $w_1^{\mathrm{i}} = w_2^{\mathrm{i}} = w^{\mathrm{i}}$ and $w_1^{\mathrm{d}} = w_2^{\mathrm{d}} = w^{\mathrm{d}}$. The relevant microstructural variables in this case are $c^{(2)}$, w^{i} and w^{d}, the evolution equations for which are the same as in the linear case ((5.7), (5.8) and (5.9)), where the fact that $\boldsymbol{A}^{(2)}$ is independent of material properties has been utilized.

In summary, expression (6.5), along with the suitable expression for the hardening rate, provides the instantaneous constitutive equation for the composite, which can be solved in combination with the evolution equations for the relevant microstructural variables to complete the constitutive description of the composite. Finally, it is noted that the results of Ponte Castañeda & Zaidman (1994) are recovered when $w_1^{\mathrm{i}} = w_1^{\mathrm{d}}$ and $w_2^{\mathrm{i}} = w_2^{\mathrm{d}}$.

(*b*) *Incompressible composites*

In this section, we consider nonlinearly viscous composites where both phases—the inclusions and the matrix—are incompressible. We make use of the result (3.4) for linearly viscous composites, specialized to the case where the constituents are isotropic and incompressible, in the variational statement (4.1) to obtain corresponding estimates for the effective potential of the nonlinearly viscous composite. When both the constituents are allowed to have the pure-power law form (2.6) with $n^{(1)} = n^{(2)} = n$, the general expression (4.1) may be simplified to give (see, also, Suquet 1993)

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) \cong \max_{y \geqslant 0} \left\{ \left[c^{(1)} + c^{(2)} \frac{z^{2n/(n+1)}}{y^{(n+1)/(n-1)}} \right] \phi^{(1)}(\hat{\sigma}_{\mathrm{e}}) \right\}, \tag{6.7}$$

where

$$\hat{\sigma}_{\mathrm{e}}^2 = [\bar{\boldsymbol{\sigma}} \cdot (\tilde{\boldsymbol{m}}\bar{\boldsymbol{\sigma}})] \left[c^{(1)} + c^{(2)} \frac{z^{2n/(n+1)}}{y^{(n+1)/(n-1)}} \right]^{-1} \quad \text{and} \quad z = \frac{\sigma_y^{(2)}}{\sigma_y^{(1)}}.$$

The optimal value of y resulting from the optimization in (6.7) is denoted $\hat{y}$; it is related to the optimal viscosities $\hat{\mu}^{(1)}$ and $\hat{\mu}^{(2)}$ through the relation, $\hat{y} = \hat{\mu}^{(2)}/\hat{\mu}^{(1)}$.

For the limiting case where the phases are rigid–perfectly plastic, we obtain the estimate for the effective yield function as

$$\tilde{\Phi}(\bar{\boldsymbol{\sigma}}) = \max_{y \geqslant 0} \left\{ [\bar{\boldsymbol{\sigma}} \cdot (\tilde{\boldsymbol{m}}\bar{\boldsymbol{\sigma}})] \left[c^{(1)} + c^{(2)} \frac{z^2}{y} \right]^{-1} - (\boldsymbol{\sigma}_y^{(1)})^2 \right\}. \tag{6.8}$$

While (6.7), along with (2.3), provides the instantaneous constitutive equation for the nonlinearly viscous composite with pure-power law potentials, for the rigid–perfectly plastic case $\dot{\Lambda}$ has to be obtained from the consistency condition which, as mentioned earlier, depends on the evolution of the microstructure.

In this case also, we shall consider a composite whose microstructure initially consists of spherical inclusions distributed isotropically and is subjected to triaxial loading conditions. The relevant microstructural variables in this case are w_1^{i}, w_2^{i}, w_1^{d} and w_2^{d} (the relative proportions of phases remains unchanged due to incompressibility, so that the microstructural variable $c^{(2)}$ is fixed in this case). The microstructural variables and the evolution equations in this case are the same as the ones for the linearly viscous, incompressible composites with the difference that (as mentioned in § 4 *b*) the optimal values of the viscosities must be used in the expression for the strain-rate concentration tensors. From (5.10), it is seen that strain-rate concentration tensor $\boldsymbol{A}^{(2)}$ depends on the viscosities only through their ratio y. This has the implication that the same evolution equations ((3.7) and (3.8)) may be used for the nonlinear case as well, provided that the optimal ratio $\hat{y} = \hat{\mu}^{(2)}/\hat{\mu}^{(1)}$ is used in the expression for the strain-rate concentration tensor in (3.7).

Proceeding as in the previous section, the consistency condition takes the form

$$\dot{\tilde{\Phi}} = \frac{\partial \tilde{\Phi}}{\partial \bar{\sigma}_{ij}} \dot{\bar{\sigma}}_{ij} + \frac{\partial \tilde{\Phi}}{\partial w_1^{\mathrm{i}}} \dot{w}_1^{\mathrm{i}} + \frac{\partial \tilde{\Phi}}{\partial w_2^{\mathrm{i}}} \dot{w}_2^{\mathrm{i}} + \frac{\partial \tilde{\Phi}}{\partial w_1^{\mathrm{d}}} \dot{w}_1^{\mathrm{d}} + \frac{\partial \tilde{\Phi}}{\partial w_2^{\mathrm{d}}} \dot{w}_2^{\mathrm{d}} = 0. \tag{6.9}$$

The evolution equations (3.7) and (3.8) are then used in the above expression in order to obtain an instantaneous relation of the form (6.5), where the hardening rate is given by

$$\begin{aligned} H = -\Bigg[& w_1^{\mathrm{i}} \frac{\partial \tilde{\Phi}}{\partial w_1^{\mathrm{i}}} (A_{33ij}^{(2)} - A_{11ij}^{(2)}) \frac{\partial \tilde{\Phi}}{\partial \bar{\sigma}_{ij}} + w_2^{\mathrm{i}} \frac{\partial \tilde{\Phi}}{\partial w_2^{\mathrm{i}}} (A_{33ij}^{(2)} - A_{22ij}^{(2)}) \frac{\partial \tilde{\Phi}}{\partial \bar{\sigma}_{ij}} \\ & + w_1^{\mathrm{d}} \frac{\partial \tilde{\Phi}}{\partial w_1^{\mathrm{d}}} \left(\frac{\partial \tilde{\Phi}}{\partial \bar{\sigma}_{33}} - \frac{\partial \tilde{\Phi}}{\partial \bar{\sigma}_{11}} \right) + w_2^{\mathrm{d}} \frac{\partial \tilde{\Phi}}{\partial w_2^{\mathrm{d}}} \left(\frac{\partial \tilde{\Phi}}{\partial \bar{\sigma}_{33}} - \frac{\partial \tilde{\Phi}}{\partial \bar{\sigma}_{22}} \right) \Bigg]. \end{aligned} \tag{6.10}$$

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For the case where the composite is subjected to axisymmetric loads, there are only two relevant microstructural variables (w^{i} and w^{d}) and the composite exhibits transverse isotropy. In that case, $\bar{\boldsymbol{\sigma}} \cdot (\tilde{\boldsymbol{m}}\bar{\boldsymbol{\sigma}})$ is given explicitly by (5.11) and can be used in (6.8) to obtain the corresponding yield function. It must also be noted that the expression for the hardening rate is further simplified by the fact that there are only two microstructural variables in this case. The evolution equations for the microstructural variables are the same as in the linear case ((5.12) and (5.13)), with the difference that the optimal value $\hat{y}$ is used in the expression for the strain-rate concentration tensor. As described earlier, the evolution equations are solved in combination with the instantaneous constitutive equations to complete the constitutive description of the nonlinearly viscous composite.

7. Concluding remarks

This part of the work has dealt with the development of constitutive models for composites which take into account the evolution of the microstructure as the composites are subjected to finite deformations. In particular, the distribution of the inclusions and its evolution in two-phase composites with particulate microstructures have been accounted for. More specifically, estimates have been developed for the effective potentials and yield surfaces of two-phase composites with allowance for the fact that the distribution of the centres of the inclusions may have a different shape from that of the inclusions. These estimates for the instantaneous response of the nonlinearly viscous composites were obtained by making use of the recent Hashin–Shtrikman estimates of Ponte Castañeda & Willis (1995) for linearly viscous composites in conjunction with the variational representation of Ponte Castañeda (1991).

The aspect ratios of the distribution of the inclusions (or voids) were identified as independent microstructural variables and evolution equations were developed for these variables. These equations, along with those for the volume fraction and the shape of the inclusions, when used in combination with the instantaneous constitutive relations, provide a full description of the behaviour of the composite under finite deformation. The model incorporates the ability to account for the effect of the distribution of the inclusions and its evolution on the overall response of the composite, thus generalizing the earlier work of Ponte Castañeda & Zaidman (1994) which neglected distributional effects. In an attempt to understand the implications of the constitutive model developed in this part of the work, part II will be concerned with a detailed study of the behaviour of certain model composite systems subjected to axisymmetric loading conditions.

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Appendix A. Results for spheroidal inclusions distributed with spheroidal symmetry

(*a*) *Porous composites*

$$m_1 = 1 + c^{(2)} \frac{8he_5 - 4e_5 - 2e_4 - c^{(2)}(8ge_5 - 4e_5 - 2e_3)}{\Delta},$$

$$m_2 = 2 + c^{(2)}\frac{2he_5 - 4e_5 + e_4 - c^{(2)}(2ge_5 - 4e_5 + e_3)}{\varDelta},$$
$$m_3 = 3 + \frac{4c^{(2)}e_3}{2e_5 - e_4 - c^{(2)}(2e_5 - e_3)},$$
$$m_4 = 3 - \frac{2c^{(2)}e_3}{4e_5 - 3ge_5 + 2e_3 - c^{(2)}(4e_5 - 3he_5 + 2e_4)},$$
$$m_5 = -1 + c^{(2)}\frac{e_4 - he_5 + 2e_5 - c^{(2)}(e_3 - ge_4 + 2e_4)}{\varDelta},$$
$$m_6 = m_5 \qquad \text{(A 1)}$$

and

$$a_1 = \frac{4e_5 - 6ge_5 - c^{(2)}(6e_5 - 6ge_5 + 2e_2)}{\varSigma},$$
$$a_2 = \frac{e_1 - c^{(2)}(e_2 + 2e_5)}{\varSigma},$$
$$a_3 = \frac{4e_5}{2e_5 - e_1 + c^{(2)}(2e_5 + e_2)},$$
$$a_4 = \frac{2e_5}{4e_5 - 3ge_5 + 2e_1 - c^{(2)}(2e_5 - 3he_5 + 2e_2)},$$
$$a_5 = \frac{e_1 - c^{(2)}e_2}{\varSigma},$$
$$a_6 = \frac{2e_5 - 3ge_5 + e_1 - c^{(2)}(2e_5 - 3he_5 + e_2)}{\varSigma}, \qquad \text{(A 2)}$$

where,

$$\varDelta = e_5(3h^2 - 8h + 4) + 2e_4 + c^{(2)}[e_5(8h + 8g - 8 + 6gh) - 2e_4 + 2e_3] + (c^{(2)})^2[e_5(3g^2 - 8g + 4) + 2e_3], \qquad \text{(A 3)}$$
$$\varSigma = c^{(2)}[3e_1 + 4e_5 - 6ge_5 - c^{(2)}(3e_2 + 6e_5 + 6he_5)]. \qquad \text{(A 4)}$$

(*b*) *Incompressible composites*

$$m_1 = \frac{2e_5 + (1-y)[6he_5 - 6e_5 - 3e_4 - c^{(2)}(6ge_5 - 6e_5 - 3e_3)]}{2e_5 + (1-y)[6he_5 - 6e_5 - 3e_4 - c^{(2)}(6ge_5 - 4e_5 - 3e_3)]},$$
$$m_2 = 2m_1,$$
$$m_3 = \frac{4e_5 + (1-y)[-2e_5 - e_4 + c^{(2)}(2e_5 + e_3)]}{4e_5 + (1-y)[-2e_5 - e_4 + c^{(2)}(-2e_5 + e_3)]},$$
$$m_4 = \frac{2e_5 + (1-y)[2e_4 - 3he_5 + 2e_5 - c^{(2)}(2e_3 - 3ge_5 + 2e_5)]}{2e_5 + (1-y)[2e_4 - 3he_5 + 2e_5 - c^{(2)}(2e_3 - 3ge_5 + 4e_5)]},$$
$$m_5 = -m_1,$$
$$m_6 = -m_1 \qquad \text{(A 5)}$$

and

$$a_1 = 2\frac{e_5 + (1-y)[2he_5 - 2e_5 - e_4 - c^{(2)}(2ge_5 - 2e_5 - e_3)]}{\varGamma},$$

$$
\begin{aligned}
a_2 &= \frac{2e_5 + (1-y)[2he_5 - 2e_5 - e_4 - c^{(2)}(2ge_5 - 2e_5 - e_3)]}{\Gamma}, \\
a_3 &= \frac{4e_5}{4e_5 + (1-y)[-2e_5 - e_4 + c^{(2)}(2e_5 + e_3)]}, \\
a_4 &= \frac{2e_5}{2e_5 + (1-y)[2e_5 - 3he_5 + 2e_4 - c^{(2)}(2e_5 - 3ge_5 + 2e_3)]}, \\
a_5 &= a_1 - a_2, \\
a_6 &= a_5,
\end{aligned} \tag{A 6}
$$

where

$$
\Gamma = 2e_5 + 3(1-y)[2he_5 - 2e_5 - e_4 - c^{(2)}(2ge_5 - 2e_5 - e_3)]. \tag{A 7}
$$

In the above expressions,

$$
\begin{aligned}
e_1 &= (3g-2)[1-(w^{\mathrm{d}})^2], \\
e_2 &= (3h-2)[1-(w^{\mathrm{i}})^2], \\
e_3 &= (3g-2)[1-(w^{\mathrm{i}})^2], \\
e_4 &= (3h-2)[1-(w^{\mathrm{d}})^2], \\
e_5 &= [1-(w^{\mathrm{i}})^2][1-(w^{\mathrm{d}})^2],
\end{aligned} \tag{A 8}
$$

$$
h(w^{\mathrm{i}}) = \begin{cases} \dfrac{w^{\mathrm{i}}[\cos^{-1}(w^{\mathrm{i}}) - w^{\mathrm{i}}(1-(w^{\mathrm{i}})^2)^{1/2}]}{(1-(w^{\mathrm{i}})^2)^{3/2}}, & \text{if } w^{\mathrm{i}} \leqslant 1, \\[2ex] \dfrac{w^{\mathrm{i}}[\cosh^{-1}(w^{\mathrm{i}}) + w^{\mathrm{i}}((w^{\mathrm{i}})^2-1)^{1/2}]}{((w^{\mathrm{i}})^2-1)^{3/2}}, & \text{if } w^{\mathrm{i}} \geqslant 1 \end{cases} \tag{A 9}
$$

and g is given by the same formula above, with the w^{i} replaced by w^{d}.

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