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# Second-order estimates for the effective behaviour of viscoplastic polycrystalline materials

M. Bornert<sup>a</sup>, R. Masson<sup>a,b</sup>, P. Ponte Castañeda<sup>c</sup>, A. Zaoui<sup>a,\*</sup>

<sup>a</sup>*Laboratoire de Mécanique des Solides, Centre commun XIENSMP/ENPC/CNRS, Ecole Polytechnique, F-91128 Palaiseau Cedex, France*

<sup>b</sup>*EDF-Research and Development Division-77818 Moret sur Loing, France*

<sup>c</sup>*Department of Mechanical Engineering and Applied Mechanics, University of Pennsylvania, Philadelphia, PA 19104-6315, USA*

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## Abstract

This paper is concerned with the application of the “second-order” nonlinear homogenisation procedure (Ponte Castañeda, *J. Mech. Phys. Solids* 44 (6) (1996) 827) to generate estimates of the self-consistent type for the effective behaviour of fcc and hcp viscoplastic polycrystals. The method has the distinctive property that it leads to estimates that are exact to second-order in the heterogeneity contrast, and which are expected to be more accurate, particularly when compared to rigorous bounds, than those resulting from earlier homogenisation schemes such as the Hill “incremental” method or its “total” formulation (Hutchinson) for pure power-law viscous materials. Special attention is paid to large grain anisotropy leading to correspondingly large heterogeneity contrast, and to highly nonlinear behaviour. Comparisons are also carried out with estimates derived from other more recent homogenisation schemes such as the “tangent” and “affine” methods. The results, illustrated for zirconium- and ice-type polycrystals, show that the second-order procedure offers the potential for significantly improved results, at least relative to the Hill incremental formulation. © 2001 Elsevier Science Ltd. All rights reserved.

*Keywords:* B. Inhomogeneous material; B. Polycrystalline material; B. Viscoplastic material; Homogenisation

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## 1. Introduction

The prediction of the effective properties of viscoplastic polycrystals under small and finite deformation has been an area of intensive activity for almost 50 years, due to the numerous applications, such as the prediction of microstructure and texture

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\* Corresponding author. Tel.: +33-1-6933-3370; fax: +33-1-6933-3026.

*E-mail address:* zaoui@lms.polytechnique.fr (A. Zaoui).

development during the thermomechanical processing of metals as well as the simulation of forming processes, by means of physically based computer models. In spite of being classical, the problem is still open largely because of its complexity. Many approximate theories have been proposed, the first of which being the popular Taylor (1938) which assumes that the strain is uniform throughout the polycrystal. Such a crude hypothesis entirely neglects the local strain heterogeneity, which has been frequently observed in the laboratory (e.g. Allais et al., 1994), and leads to an upper bound (Hill, 1952) for the effective strain rate potential, which is usually too stiff to be used to estimate the overall stress by derivation of such a potential. The dual Reuss-type approach, based on the similarly unrealistic but easy to handle assumption of uniform local stresses, leads to a lower bound that underestimates the effective dissipation.

Many other more sophisticated but still approximate models have been proposed for the derivation of more realistic estimates for the effective properties and local strain heterogeneities of nonlinear polycrystals. Most of them are based on some type of nonlinear extension of the classical linear self-consistent model (Kröner, 1958, 1977), which is widely accepted as an adequate model for the prediction of the effective elastic moduli of polycrystals. Omitting earlier contributions that assumed purely elastic interactions at the local scale (Kröner, 1961), the earliest contribution in the field is Hill's (1965) pioneering work, which is based on a linearised *incremental formulation* of the elastoplastic local constitutive equations, using tangent moduli. This complex formulation was implemented several years later by Hutchinson (1970), who also adapted it to viscoplasticity (Hutchinson 1976, 1977) and noticed that, for the particular case of power-law creep, the incremental formulation could be simplified into a total one, which was based on the use of secant moduli instead of tangent ones and easier to implement. His numerical results also seemed to suggest that Hill's formulation leads to predictions tending to the Taylor estimate for highly nonlinear phases, and especially for perfect plasticity. The secant formulation was further simplified by Berveiller and Zaoui (1979) in the special case of isotropic polycrystals, by means of some isotropic simplification of the secant moduli. Such ideas have also been applied in the context of two-phase composites (Qiu and Weng, 1991; Hervé and Zaoui, 1990). This *isotropic secant formulation* leads in many cases to predictions somewhat softer than the incremental one. Another total formulation, due to Molinari et al. (1987) who implemented it in some approximate isotropic form, is based on a tangent approximation of the nonlinear stress/strain rate relation, which is expected to be more accurate than a secant one. The full implementation of this *tangent model* for viscous polycrystals is due to Lebensohn and Tomé (1993), who observed that this scheme tends to the Reuss-type lower bound for high nonlinearities. This scheme suffers from several limitations, primary among which is the fact that it is restricted to (a specific) self-consistent type of model and to power law creep. More recently, a more general approach based on the same tangent approximation of the nonlinear behaviour of the constitutive phases has been proposed by Masson et al. (2000). This *affine formulation*, which can be applied to any type of microstructure and to any form of stress/strain rate relation, leads to predictions intermediate between the secant and the tangent approaches, without recourse to any appropriately fitted parameter.

In addition to these approximate schemes, variational approaches have been developed (Talbot and Willis, 1985; Ponte Castañeda, 1991; Willis, 1994; deBotton and Ponte Castañeda, 1995; see also Ponte Castañeda and Suquet, 1998, for a general review) and provide rigorous bounds for the effective dissipation potential, which, for the kind of nonlinearities exhibited by viscous materials, are upper bounds. These approaches are in general more difficult to implement but provide rigorous results. They have been compared to the above estimates in the simpler case of viscous two-phase materials (Gilormini, 1996) as well as for two-dimensional model polycrystals (Bornert and Ponte Castañeda, 1998) and it has been shown that all approximate schemes may violate the bounds, at least for some particular combination of the parameters.

Following up on the work of Bornert and Ponte Castañeda (1998) for model 2D polycrystals, this paper is concerned with the application of the second-order procedure (Ponte Castañeda, 1996) to estimate the effective behaviour of general 3D viscoplastic polycrystals. The method is based on second-order Taylor expansions of the relevant potentials for the constituent phases allowing the expression of the effective potential for the nonlinear material in terms of that of a suitably chosen linear comparison thermoelastic material. The method has the distinctive advantage that it leads to estimates that are exact to second-order in the heterogeneity contrast (Suquet and Ponte Castañeda, 1993), but has the disadvantage that it exhibits a duality gap (Ponte Castañeda, 1996). It has recently been given a variational interpretation (Ponte Castañeda and Willis, 1999) and has already been applied in the context of two-phase systems, including several important special cases, where the resulting estimates appear to be more accurate, particularly when compared to rigorous bounds, than earlier estimates such as those that are based on the Hill “incremental” or the Hutchinson “total” formulations. This work aims to investigate the case of viscoplastic polycrystals in some detail by applying the second-order procedure to generate estimates of the self-consistent type for the overall response of fcc as well as hcp polycrystals, with special attention being paid to large grain anisotropy and correspondingly large heterogeneity contrast, and to highly nonlinear behaviour.

The approach adopted is introduced in Sections 2 to 4, with detailed derivations given in Appendix A for the required per-phase average strain rates in a thermoelastic multiphase composite according to the Hashin–Shtrikman variational procedure and including estimates of self-consistent type for this class of linear materials. Section 5 then reports on a number of applications for different kinds of polycrystals, with illustrations for fcc and hcp zirconium- and ice-type polycrystals. Computational details are reported in Appendix B. The results clearly show that the second-order procedure offers the potential for significantly improved estimates for the effective response of viscoplastic polycrystals, at least relative to the Hill incremental method.

## **2. Effective properties for viscoplastic polycrystals**

Here a polycrystal is defined as a random aggregate of perfectly bonded single-crystal grains with a prescribed orientation distribution function (ODF) and set of two-point correlation functions (see, for example, Adams and Olson, 1998). The ODF determines

the *crystallographic texture* and the corresponding two-point statistics correlate with *grain shape*, so that the two-point correlation functions serve to characterise in an approximate fashion the *morphological texture*. For simplicity, it will be assumed here that the grain orientations take on a set of discrete values, characterised by rotation tensors  $\mathbf{Q}^{(r)}$  ( $r = 1, \dots, N$ ), and that the two-point correlation functions exhibit “ellipsoidal” symmetry with the same shape for all crystal orientations, characterised by a symmetric tensor  $\mathbf{Z}$  (correspondingly, the grains are assumed to be on average ellipsoidal in shape with identical aspect ratios and orientations for all lattice orientations). It will also be assumed that the polycrystal occupies a region  $\Omega$ , while all the grains of a given orientation  $\mathbf{Q}^{(r)}$  occupy subregions  $\Omega^{(r)}$  ( $r = 1, \dots, N$ ). The characteristic functions  $\chi^{(r)}$  describing the location of the various orientations are defined to be equal to 1 if the position vector  $\mathbf{x}$  is in  $\Omega^{(r)}$  and zero otherwise. Volume averages over  $\Omega$  are denoted by  $\langle \cdot \rangle$ , so that the scalars  $c^{(r)} = \langle \chi^{(r)} \rangle$  characterise the crystallographic texture of the polycrystal.

For a given stress  $\boldsymbol{\sigma}$ , the *local* constitutive response is then defined by

$$\boldsymbol{\varepsilon} = \frac{\partial u}{\partial \boldsymbol{\sigma}}, \quad u(\mathbf{x}, \boldsymbol{\sigma}) = \sum_{r=1}^N \chi^{(r)}(\mathbf{x}) u^{(r)}(\boldsymbol{\sigma}), \quad u^{(r)}(\boldsymbol{\sigma}) = \sum_{k=1}^K \phi_{(k)}(\boldsymbol{\tau}_{(k)}^{(r)}), \quad (1)$$

where  $\boldsymbol{\varepsilon}$  is the Eulerian strain rate, and  $u$  and  $u^{(r)}$  are the stress potentials for the polycrystal and orientation  $r$ , respectively. The convex functions  $\phi_{(k)}$  ( $k = 1, \dots, K$ ) characterise the response of the  $K$  slip systems in each crystal and depend on the resolved shear stresses

$$\boldsymbol{\tau}_{(k)}^{(r)} = \boldsymbol{\sigma} \cdot \boldsymbol{\mu}_{(k)}^{(r)}, \quad (2)$$

where the dot denotes the inner product of two second-order tensors (e.g.  $\boldsymbol{\sigma} \cdot \boldsymbol{\mu} = \sigma_{ij} \mu_{ij}$ ) and the set

$$\boldsymbol{\mu}_{(k)}^{(r)} = \frac{1}{2} (\mathbf{n}_{(k)}^{(r)} \otimes \mathbf{m}_{(k)}^{(r)} + \mathbf{m}_{(k)}^{(r)} \otimes \mathbf{n}_{(k)}^{(r)}) \quad (3)$$

are second-order tensors with  $\mathbf{n}_{(k)}^{(r)}$  and  $\mathbf{m}_{(k)}^{(r)}$  denoting the unit vectors normal to the slip plane and along the slip direction in the  $k$ th system, respectively, for each orientation  $r$ .

It is known (e.g. Hutchinson, 1976) that the *effective* viscous response for the polycrystal may be written in the form

$$\bar{\boldsymbol{\varepsilon}} = \frac{\partial \tilde{U}}{\partial \bar{\boldsymbol{\sigma}}}, \quad e\tilde{U}(\bar{\boldsymbol{\sigma}}) = \min_{\boldsymbol{\tau} \in \mathcal{S}(\bar{\boldsymbol{\sigma}})} \langle u(\mathbf{x}, \boldsymbol{\tau}) \rangle. \quad (4)$$

Here  $\tilde{U}$  is the *effective stress potential* for the polycrystal,  $\bar{\boldsymbol{\sigma}} = \langle \boldsymbol{\sigma} \rangle$  and  $\bar{\boldsymbol{\varepsilon}} = \langle \boldsymbol{\varepsilon} \rangle$  are the *average stress* and *strain rate*, and  $\mathcal{S}(\boldsymbol{\mathcal{S}}) = \{ \boldsymbol{\tau}, \operatorname{div} \boldsymbol{\tau} = 0 \text{ in } \Omega, \boldsymbol{\tau} \mathbf{n} = \mathbf{S} \mathbf{n} \text{ on } \partial \Omega \}$  denotes the set of statically admissible stresses.

Dually, in terms of the dissipation potentials

$$w^{(r)}(\boldsymbol{\varepsilon}) = (u^{(r)})^*(\boldsymbol{\varepsilon}) = \max_{\boldsymbol{\sigma}} \{ \boldsymbol{\sigma} \cdot \boldsymbol{\varepsilon} - u^{(r)}(\boldsymbol{\sigma}) \} \quad (5)$$

such that

$$\boldsymbol{\sigma} = \frac{\partial w}{\partial \boldsymbol{\varepsilon}}, \quad w(\mathbf{x}, \boldsymbol{\varepsilon}) = \sum_{r=1}^N \lambda^{(r)}(\mathbf{x}) w^{(r)}(\boldsymbol{\varepsilon}), \quad (6)$$

the effective response of the polycrystal may be written in the form

$$\bar{\boldsymbol{\sigma}} = \frac{\partial \tilde{W}}{\partial \bar{\boldsymbol{\varepsilon}}}, \quad \tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \min_{\boldsymbol{\varepsilon} \in \mathcal{H}(\bar{\boldsymbol{\varepsilon}})} \langle w(\mathbf{x}, \boldsymbol{\varepsilon}) \rangle, \quad (7)$$

where  $\tilde{W}$  is the *effective dissipation potential* for the polycrystal, and  $\mathcal{H}$  is the set of kinematically admissible strain rates:

$$\mathcal{H}(\bar{\boldsymbol{\varepsilon}}) = \left\{ \boldsymbol{\varepsilon} \left| \text{there is } \mathbf{v} \text{ with } \boldsymbol{\varepsilon} = \frac{1}{2}[\nabla \mathbf{v} + (\nabla \mathbf{v})^T] \text{ in } \Omega, \mathbf{v} = \bar{\boldsymbol{\varepsilon}} \mathbf{x} \text{ on } \partial\Omega \right. \right\}. \quad (8)$$

### 3. Second-order estimates

As already mentioned, the second-order procedure was developed to generate estimates for the effective behaviour of nonlinear composites that are *exact to second-order* in the heterogeneity contrast. Thus, such estimates should be quite accurate for materials such as fcc polycrystals, which exhibit fairly small levels of heterogeneity, as a consequence of their relatively low degree of grain anisotropy. However, as already shown in the context of two-phase composites by Ponte Castañeda (1996), the second-order procedure also delivers fairly accurate results at high contrast, including the extreme cases of porous and rigidly reinforced composites. In this section, an abbreviated formulation of the second-order procedure is given for completeness. Further details may be obtained from Ponte Castañeda (1996), Ponte Castañeda and Suquet (1998) and Ponte Castañeda and Willis (1999).

The procedure is based on the use of a Taylor-type approximation for the phase potentials  $w^{(r)}$ . Introducing reference strain rates  $\bar{\boldsymbol{\varepsilon}}^{(r)}$ , the Taylor formula for  $w^{(r)}$  about  $\bar{\boldsymbol{\varepsilon}}^{(r)}$  is given by

$$w^{(r)}(\boldsymbol{\varepsilon}) = w^{(r)}(\bar{\boldsymbol{\varepsilon}}^{(r)}) + \boldsymbol{\rho}^{(r)} \cdot (\boldsymbol{\varepsilon} - \bar{\boldsymbol{\varepsilon}}^{(r)}) + \frac{1}{2}(\boldsymbol{\varepsilon} - \bar{\boldsymbol{\varepsilon}}^{(r)}) \cdot \mathbf{L}^{(r)}(\boldsymbol{\varepsilon} - \bar{\boldsymbol{\varepsilon}}^{(r)}), \quad (9)$$

where the symbols  $\boldsymbol{\rho}^{(r)}$ , defined by

$$\boldsymbol{\rho}^{(r)} = \frac{\partial w^{(r)}}{\partial \boldsymbol{\varepsilon}}(\bar{\boldsymbol{\varepsilon}}^{(r)}), \quad (10)$$

are used to denote stress-like variables in each phase, and the  $\mathbf{L}^{(r)}$ , which are as yet undetermined, serve to denote constant viscous moduli tensors in the phases.

It follows from Eq. (7) that the effective potential  $\tilde{W}$  of the nonlinear composite may be estimated as

$$\tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \sum_{r=1}^N c^{(r)} \left[ w^{(r)}(\bar{\boldsymbol{\varepsilon}}^{(r)}) - \boldsymbol{\rho}^{(r)} \cdot \bar{\boldsymbol{\varepsilon}}^{(r)} + \frac{1}{2} \bar{\boldsymbol{\varepsilon}}^{(r)} \cdot \mathbf{L}^{(r)} \bar{\boldsymbol{\varepsilon}}^{(r)} \right] + \tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}), \quad (11)$$

where reinterpreting  $\boldsymbol{\varepsilon}$  as a strain field,  $\tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \mathbf{I})$  can be thought of as the free energy of a *linear thermoelastic comparison* multiphase composite with moduli tensors  $\mathbf{L}^{(r)}$  and thermal stresses  $-\mathbf{I}^{(r)}$ , defined by

$$\mathbf{I}^{(r)} = \mathbf{L}^{(r)} \bar{\boldsymbol{\varepsilon}}^{(r)} - \boldsymbol{\rho}^{(r)}. \quad (12)$$

It is given by

$$\tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \mathbf{I}) = \min_{\boldsymbol{\varepsilon} \in \mathcal{H}(\bar{\boldsymbol{\varepsilon}})} \left\langle \frac{1}{2} \boldsymbol{\varepsilon} \cdot \mathbf{L} \boldsymbol{\varepsilon} - \mathbf{I} \cdot \boldsymbol{\varepsilon} \right\rangle, \quad (13)$$

where  $\mathbf{I}(\mathbf{x}) = \sum_{r=1}^N \chi^{(r)}(\mathbf{x}) \mathbf{I}^{(r)}$  and  $\mathbf{L}(\mathbf{x}) = \sum_{r=1}^N \chi^{(r)}(\mathbf{x}) \mathbf{L}^{(r)}$ . Note that neither the moduli tensors  $\mathbf{L}^{(r)}$  nor the pre-stresses  $\mathbf{I}^{(r)}$  are rotated versions of some reference values, so that the linear thermoelastic comparison composite cannot be considered to be a linear polycrystal, but must be thought of as a more general  $N$ -phase system.

The approximate expression (11) for the effective potential  $\tilde{W}$  of the nonlinear polycrystal is easier to compute than the original, exact expression (7). This is because it requires the solution of a linear problem (i.e., Eq. (13)) instead of a nonlinear one. In this connection, it is noted that estimates for linear-thermoelastic heterogeneous materials can be obtained by appropriate extension of the corresponding methods for linear-elastic composites; further details are given in appendix A. At this stage, it is only noted that, given an estimate for  $\tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \mathbf{I})$ , the expression (11) provides a corresponding estimate for  $\tilde{W}$ , for all choices of the  $\bar{\boldsymbol{\varepsilon}}^{(r)}$  and  $\mathbf{L}^{(r)}$ , which are thus far unspecified. Using the notation  $\langle \cdot \rangle^{(r)}$  to denote volume averages over phase  $r$ , the optimal choice for the variables  $\bar{\boldsymbol{\varepsilon}}^{(r)}$  follows by setting the quantity

$$\frac{\partial \tilde{W}}{\partial \bar{\boldsymbol{\varepsilon}}^{(r)}} = c^{(r)} \left[ \mathbf{L}^{(r)} - \frac{\partial^2 w^{(r)}}{\partial \boldsymbol{\varepsilon} \partial \boldsymbol{\varepsilon}}(\bar{\boldsymbol{\varepsilon}}^{(r)}) \right] (\bar{\boldsymbol{\varepsilon}}^{(r)} - \langle \boldsymbol{\varepsilon} \rangle^{(r)}) \quad (14)$$

identically equal to zero, which leads to the prescription that

$$\bar{\boldsymbol{\varepsilon}}^{(r)} = \langle \boldsymbol{\varepsilon} \rangle^{(r)}, \quad (15)$$

where it is emphasised that  $\boldsymbol{\varepsilon}$  is the strain in the thermoelastic comparison composite. This prescription was originally proposed by Ponte Castañeda (1996), based on the physical intuition that the strain  $\boldsymbol{\varepsilon}$  in phase  $r$  would oscillate about its average  $\bar{\boldsymbol{\varepsilon}}^{(r)}$  in phase  $r$  in such a way that large deviations would only be expected in regions of relatively small measure. The more rigorous derivation given here follows from the work of Ponte Castañeda and Suquet (1998) and Ponte Castañeda and Willis (1999).

Prescription (15) allows further simplification of estimate (11) for  $\tilde{W}$ . Thus, the Hill condition for the thermoelastic comparison solid ensures that

$$\langle \boldsymbol{\varepsilon} \cdot \mathbf{L} \boldsymbol{\varepsilon} \rangle = \langle \mathbf{I} \cdot \boldsymbol{\varepsilon} \rangle + \bar{\boldsymbol{\varepsilon}} \cdot \langle \mathbf{L} \boldsymbol{\varepsilon} - \mathbf{I} \rangle, \quad (16)$$

which, using definition (12) for the polarisations  $\mathbf{I}^{(r)}$ , combined with prescription (15), leads to

$$\tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \mathbf{I}) = \frac{1}{2} \sum_{r=1}^N c^{(r)} [\boldsymbol{\rho}^{(r)} \cdot (\bar{\boldsymbol{\varepsilon}} + \bar{\boldsymbol{\varepsilon}}^{(r)}) - \bar{\boldsymbol{\varepsilon}}^{(r)} \cdot \mathbf{L}^{(r)} \bar{\boldsymbol{\varepsilon}}^{(r)}]. \quad (17)$$

This result can then be used to rewrite estimate (11) in the simpler form (Ponte Castañeda and Suquet, 1998)

$$\tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \sum_{r=1}^N c^{(r)} \left[ w^{(r)}(\bar{\boldsymbol{\varepsilon}}^{(r)}) + \frac{1}{2} \boldsymbol{\rho}^{(r)} \cdot (\bar{\boldsymbol{\varepsilon}} - \bar{\boldsymbol{\varepsilon}}^{(r)}) \right], \quad (18)$$

where the  $\bar{\boldsymbol{\varepsilon}}^{(r)}$  are defined by prescription (15) and the  $\boldsymbol{\rho}^{(r)}$  are determined by expression (10) in terms of the  $\bar{\boldsymbol{\varepsilon}}^{(r)}$ .

The above estimates for  $\tilde{W}$  are valid for any choice of the moduli tensors in the linear comparison composite  $\mathbf{L}^{(r)}$ . The question then arises as to the best choice for these. However, it appears that stationarity of estimate (11), together with Eq. (13) or (18) does not lead to a useful result. It has been proposed to choose the  $\mathbf{L}^{(r)}$  from the prescription (Ponte Castañeda, 1996)

$$\mathbf{L}^{(r)} = \frac{\partial^2 w^{(r)}}{\partial \boldsymbol{\varepsilon} \partial \boldsymbol{\varepsilon}}(\bar{\boldsymbol{\varepsilon}}^{(r)}), \quad (19)$$

which has the interesting property (Ponte Castañeda and Willis, 1999) that it implies that

$$\frac{\partial^2 \tilde{W}}{\partial \bar{\boldsymbol{\varepsilon}}^{(r)} \partial \bar{\boldsymbol{\varepsilon}}^{(r)}} = 0 \quad (20)$$

or *second-order stationarity* with respect to the variables  $\bar{\boldsymbol{\varepsilon}}^{(r)}$ . There are other possible choices for  $\mathbf{L}^{(r)}$ , but they have not yet been explored in full detail.

It is noted that a similar expression may be obtained for the effective stress potential  $\tilde{U}$ , as defined by Eq. (4), by proceeding analogously with a Taylor expansion of the stress potentials  $u^{(r)}$  about appropriate reference stresses  $\bar{\boldsymbol{\sigma}}^{(r)}$ . The result may be written as

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \sum_{r=1}^N c^{(r)} \left[ u^{(r)}(\bar{\boldsymbol{\sigma}}^{(r)}) + \frac{1}{2} \frac{\partial u^{(r)}}{\partial \boldsymbol{\sigma}}(\bar{\boldsymbol{\sigma}}^{(r)}) \cdot (\bar{\boldsymbol{\sigma}} - \bar{\boldsymbol{\sigma}}^{(r)}) \right], \quad (21)$$

where the  $\bar{\boldsymbol{\sigma}}^{(r)}$  has been identified with the average stresses in the phases of the corresponding linear comparison composite. Unfortunately, the estimate for  $\tilde{U}$  is not the Legendre dual of estimate (18) for  $\tilde{W}$ , and is therefore not equivalent to it. A clear explanation of this fact, as well as of the connections with the affine procedure (Zaoui and Masson, 2000) has been given by Masson et al. (2000). However, as suggested in Ponte Castañeda (1996), the expansion based on the dissipation potentials is preferred in plasticity, as a consequence of the fact that, for the physical nonlinearity present in plasticity and creep, the Taylor expansions on the dissipation potentials are expected to be more accurate than the corresponding expansions on the stress potentials.

#### 4. Estimates of the self-consistent type

Given the simplified form (18) for  $\tilde{W}$ , the implementation of the second-order procedure now only requires estimates for the phase averages  $\bar{\boldsymbol{\varepsilon}}^{(r)}$  of the strain field in the



variational thermoelastic problem (13). Formally, this can be accomplished by means of the identity

$$\bar{\boldsymbol{\varepsilon}}^{(r)} = -\frac{1}{c^{(r)}} \frac{\partial \tilde{\Psi}}{\partial \mathbf{I}^{(r)}}(\bar{\boldsymbol{\varepsilon}}; \mathbf{I}). \quad (22)$$

Alternatively, because the problem is linear, the principle of superposition ensures that (Laws, 1973)

$$\bar{\boldsymbol{\varepsilon}}^{(r)} = \mathbf{A}^{(r)} \bar{\boldsymbol{\varepsilon}} - \mathbf{a}^{(r)}. \quad (23)$$

The fourth-order tensor  $\mathbf{A}^{(r)}$  is the linear localisation tensor which depends on the phase distribution of the composite and has to be estimated by an appropriate linear homogenisation scheme. For the class of polycrystals under consideration here, an appropriate choice is the classical self-consistent (SC) model (Kröner, 1958; Hill, 1965), which leads to the following estimate for  $\mathbf{A}^{(r)}$ :

$$\mathbf{A}^{(r)} \approx \mathbf{A}_{\text{SC}}^{(r)} = [\mathbf{L}^{(r)} + \mathbf{L}_{\text{SC}}^{\star}]^{-1} \mathbf{P}_{\text{SC}}^{-1}, \quad (24)$$

where  $\mathbf{L}_{\text{SC}}^{\star} = \mathbf{P}_{\text{SC}}^{-1} - \tilde{\mathbf{L}}_{\text{SC}}$  is the constraint tensor defined by Hill (1965) and  $\tilde{\mathbf{L}}_{\text{SC}}$  is the self-consistent estimate for the effective modulus tensor  $\tilde{\mathbf{L}}$ , which is obtained as the solution of the implicit equation

$$[\tilde{\mathbf{L}}_{\text{SC}} + \mathbf{L}_{\text{SC}}^{\star}]^{-1} = \sum_{s=1}^N c^{(s)} [\mathbf{L}^{(s)} + \mathbf{L}_{\text{SC}}^{\star}]^{-1} \quad (25)$$

to be solved iteratively (see Appendix B). In these relations,  $\mathbf{P}_{\text{SC}}$  is a microstructural tensor, depending on  $\tilde{\mathbf{L}}_{\text{SC}}$  and on the “shape” of the two-point correlation functions  $\langle \chi^{(r)}(\cdot) \chi^{(s)}(\cdot + \mathbf{h}) \rangle$  for the distribution of the grain orientations within the polycrystal (see Appendix A). More generally, for a given modulus tensor  $\mathbf{L}^{(0)}$ ,

$$\mathbf{P}^{(0)} = \frac{1}{4\pi \det \mathbf{Z}} \int_{|\boldsymbol{\xi}|=1} \mathbf{H}^{(0)}(\boldsymbol{\xi}) |\mathbf{Z}^{-1} \boldsymbol{\xi}|^{-3} dS, \quad (26)$$

where  $H_{ijkh}^{(0)}(\boldsymbol{\xi}) = N_{ik}^{(0)} \xi_j \xi_h |(ij)(kh)|$ ,  $\mathbf{N}^{(0)} = \mathbf{K}^{(0)-1}$ ,  $K_{ik}^{(0)} = L_{ijkh}^{(0)} \xi_j \xi_h$ , and  $\mathbf{Z}$  is a symmetric, second-order tensor serving to characterise the “shape” of the two-point correlation functions, under the “ellipsoidal symmetry” hypothesis (Willis, 1977). For a polycrystal in which the orientations of two adjacent grains are uncorrelated, the shape of the two-point correlation functions is known to correlate with the “average” shape of the grains. In particular,  $\mathbf{Z} = \mathbf{I}$  corresponds to the special case of statistical isotropy for the two-point correlation functions, which is similar to the so-called “equiaxed” grain hypothesis. More generally, the tensor  $\mathbf{Z}$  takes account of change in the average shape of the grains, when the polycrystal is subjected to finite deformation histories, such as rolling or extrusion. It should be emphasized that the above expression for  $\tilde{\mathbf{L}}$  would apply not just for the case of one-phase polycrystals, where the tensors  $\mathbf{L}^{(r)}$  are “rotated” versions of a reference single crystal, but for more general situations where the  $\mathbf{L}^{(r)}$  could be unrelated to each other.

The second-order tensors  $\mathbf{a}^{(r)}$  give the average strains in the phases induced by the thermal stresses  $\mathbf{I}^{(r)}$ . While they can be computed exactly from the tensors  $\mathbf{A}^{(r)}$  in the case of two-phase materials (Levin, 1967), there is no general relation for multiphase

composites. However, when  $\mathbf{A}^{(r)}$  is obtained from a Hashin and Shtrikman (1963) type of estimate, among which the self-consistent model is a particular case, there is a “natural” estimate for the tensors  $\mathbf{a}^{(r)}$ , the derivation of which relies on exactly the same assumption on the phase distribution as the one that was used to derive the estimates for  $\mathbf{A}^{(r)}$ . The justification of this property can be found in Willis (1981). For convenience and completeness, the derivation is given in Appendix A, in a slightly different form and with explicit relations. As a consequence, any two-phase linear model and any Hashin–Shtrikman type of estimate can be extended to nonlinear behaviour by means of the second-order procedure. In particular, for self-consistent estimates one gets

$$\mathbf{a}^{(r)} \approx \mathbf{a}_{\text{SC}}^{(r)} = [\mathbf{L}^{(r)} + \mathbf{L}_{\text{SC}}^{\star}]^{-1} [\tilde{\mathbf{I}}_{\text{SC}} - \mathbf{l}^{(r)}] \quad \text{with} \quad \tilde{\mathbf{I}}_{\text{SC}} = \sum_{s=1}^N c^{(s)} \mathbf{l}^{(s)} \mathbf{A}_{\text{SC}}^{(s)}. \quad (27)$$

The reference strains  $\bar{\boldsymbol{\varepsilon}}^{(r)}$  required by the second-order procedure are then determined by the set of nonlinear equations (10), (12), (19),(23)–(25) and (27), which can be rewritten as follows:

$$\begin{aligned} \mathbf{L}^{(r)} &= \frac{\partial^2 w^{(r)}}{\partial \boldsymbol{\varepsilon} \partial \boldsymbol{\varepsilon}}(\bar{\boldsymbol{\varepsilon}}^{(r)}), \\ \tilde{\mathbf{L}}_{\text{SC}} &\text{ solution of Eq. (25) and } \mathbf{L}_{\text{SC}}^{\star} = \mathbf{P}_{\text{SC}}^{-1} - \tilde{\mathbf{L}}_{\text{SC}}, \\ \boldsymbol{\rho}^{(r)} - \bar{\boldsymbol{\rho}} &= -\mathbf{L}_{\text{SC}}^{\star} [\bar{\boldsymbol{\varepsilon}}^{(r)} - \bar{\boldsymbol{\varepsilon}}], \\ \boldsymbol{\rho}^{(r)} &= \frac{\partial w^{(r)}}{\partial \boldsymbol{\varepsilon}}(\bar{\boldsymbol{\varepsilon}}^{(r)}), \\ \bar{\boldsymbol{\varepsilon}} &= \sum_s c_s \bar{\boldsymbol{\varepsilon}}^{(s)}, \\ \bar{\boldsymbol{\rho}} &= \sum_s c_s \boldsymbol{\rho}^{(s)}. \end{aligned} \quad (28)$$

Note that the four last equations are identical to those that determine the (uniform) strain in a nonlinear ellipsoidal inclusion of phase ( $r$ ) embedded in an infinite linear thermoelastic matrix with tensor of moduli  $\tilde{\mathbf{L}}_{\text{SC}}$  and subjected to homogeneous strain  $\bar{\boldsymbol{\varepsilon}}$  and stress  $\bar{\boldsymbol{\rho}}$  at infinity. Appendix B provides some details on how to solve this nonlinear system of equations iteratively for any applied deformation  $\bar{\boldsymbol{\varepsilon}}$ . The connections between these nonlinear equations and those related to the derivation of nonlinear incremental (Hutchinson, 1976), tangent (Lebensohn and Tomé, 1993) or affine (Masson et al., 2000) estimates are discussed as well.

## 5. Applications

In the following applications, the equiaxed grain assumption (i.e. isotropic two-point correlation functions) is adopted and the slip potentials are assumed to be of the

power-law type

$$\phi_{(k)}(\boldsymbol{\tau}_{(k)}^{(r)}) = \frac{\gamma^0 \boldsymbol{\tau}_{(k)}^{(0)}}{n_{(k)} + 1} \left| \frac{\boldsymbol{\tau}_{(k)}^{(r)}}{\boldsymbol{\tau}_{(k)}^{(0)}} \right|^{n_{(k)}+1}, \quad (29)$$

where  $\gamma^0$  is some reference shear strain rate and *both* the power  $n_{(k)}$  and the flow stress  $\boldsymbol{\tau}_{(k)}^{(0)}$  are allowed to be different for different slip-system families, so that, in general, the macroscopic behaviour of the polycrystal will not be of the power-law type. As mentioned earlier, the second-order procedure leads to two different results depending on the choice of the formulation (i.e. dissipation versus stress potential). The former being more appropriate for plastic behaviour ( $n_{(k)} \geq 1$ ), all the results given hereafter are based on the dissipation potential formulation.

The following applications are concerned with isotropic or transversely isotropic polycrystals. For the latter more general case, let  $(\mathbf{e}_1, \mathbf{e}_2)$  and  $\mathbf{e}_3$  denote the unit base vectors in the transverse plane and along the longitudinal direction, respectively. The creep overall potential must have the form (Aravas et al., 1995)

$$\tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \tilde{W}(I_1, I_2, I_3, I_4; s), \quad (30)$$

where  $s$  denotes a set of material parameters and  $I_1, I_2, I_3$  and  $I_4$  are four independent transversely isotropic invariants of the traceless symmetric second-order tensor  $\bar{\boldsymbol{\varepsilon}}$ , as defined by deBotton and Ponte Castañeda (1993)

$$\begin{aligned} I_1 &= \bar{\varepsilon}_{33}, & I_2 &= \sqrt{\bar{\varepsilon}_{12}^2 + \frac{(\bar{\varepsilon}_{11} - \bar{\varepsilon}_{22})^2}{4}} \\ I_3 &= \sqrt{\bar{\varepsilon}_{13}^2 + \bar{\varepsilon}_{23}^2}, & I_4 &= \sqrt[3]{\bar{\varepsilon}_{ij} \bar{\varepsilon}_{jk} \bar{\varepsilon}_{ki}}. \end{aligned} \quad (31)$$

In the following examples, polycrystals are subjected to an overall isochoric tensile strain rate along the symmetry axis  $\bar{\boldsymbol{\varepsilon}}^0 = \bar{\varepsilon}_{33}^0 [\mathbf{e}_3 \otimes \mathbf{e}_3 - \frac{1}{2}(\mathbf{e}_1 \otimes \mathbf{e}_1 + \mathbf{e}_2 \otimes \mathbf{e}_2)]$ . For such a load, the above invariants and their derivatives with respect to the tensor  $\bar{\boldsymbol{\varepsilon}}$  are given by

$$\begin{aligned} I_1(\bar{\boldsymbol{\varepsilon}}^0) &= \bar{\varepsilon}_{33}^0, & \frac{\partial I_1}{\partial \bar{\boldsymbol{\varepsilon}}}(\bar{\boldsymbol{\varepsilon}}^0) &= \mathbf{e}_3 \otimes \mathbf{e}_3, \\ I_2(\bar{\boldsymbol{\varepsilon}}^0) &= 0, & \frac{\partial I_2}{\partial \bar{\boldsymbol{\varepsilon}}}(\bar{\boldsymbol{\varepsilon}}^0) &= 0, \\ I_3(\bar{\boldsymbol{\varepsilon}}^0) &= 0, & \frac{\partial I_3}{\partial \bar{\boldsymbol{\varepsilon}}}(\bar{\boldsymbol{\varepsilon}}^0) &= 0, \\ I_4(\bar{\boldsymbol{\varepsilon}}^0) &= \sqrt[3]{\frac{3}{4} \bar{\varepsilon}_{33}^0}, & \frac{\partial I_4}{\partial \bar{\boldsymbol{\varepsilon}}}(\bar{\boldsymbol{\varepsilon}}^0) &= \frac{\bar{\varepsilon}_{33}^0}{(I_4(\bar{\boldsymbol{\varepsilon}}^0))^2} \left[ \mathbf{e}_3 \otimes \mathbf{e}_3 + \frac{1}{4}(\mathbf{e}_1 \otimes \mathbf{e}_1 + \mathbf{e}_2 \otimes \mathbf{e}_2) \right]. \end{aligned} \quad (32)$$

so that the macroscopic stress reads as

$$\begin{aligned} \bar{\boldsymbol{\sigma}} &= p\mathbf{i} + \sum_{i=1}^4 \frac{\partial \tilde{W}}{\partial I_i} \frac{\partial I_i}{\partial \bar{\boldsymbol{\varepsilon}}} = p\mathbf{i} + \frac{\partial \tilde{W}}{\partial I_1} \mathbf{e}_3 \otimes \mathbf{e}_3 \\ &+ \frac{\partial \tilde{W}}{\partial I_4} \frac{\bar{\varepsilon}_{33}^0}{(I_4(\bar{\boldsymbol{\varepsilon}}^0))^2} \left[ \mathbf{e}_3 \otimes \mathbf{e}_3 + \frac{1}{4}(\mathbf{e}_1 \otimes \mathbf{e}_1 + \mathbf{e}_2 \otimes \mathbf{e}_2) \right], \end{aligned} \quad (33)$$

where  $\mathbf{i}$  is the second-order identity tensor and  $p$  is the overall hydrostatic stress. If it is such that the overall stress is a uniaxial tension along  $\mathbf{e}_3$ , the tensile stress is

$$\bar{\sigma}_{33} = \frac{\partial \tilde{W}}{\partial I_1} + \sqrt[3]{\frac{3}{4}} \frac{\partial \tilde{W}}{\partial I_4}. \quad (34)$$

It is easy to check that it is equal to the derivative of the function  $\tilde{W}^0(\bar{\varepsilon}_{33}^0) = \tilde{W}(\bar{\boldsymbol{\varepsilon}}^0) = \tilde{W}(\bar{\varepsilon}_{33}^0, 0, 0, \sqrt[3]{\frac{3}{4}} \bar{\varepsilon}_{33}^0; s)$  with respect to its (scalar) argument

$$\bar{\sigma}_{33} = \frac{d\tilde{W}^0}{d\bar{\varepsilon}_{33}^0}. \quad (35)$$

When, in addition, the power-law exponents  $n_{(k)}$  are uniform and equal to  $n$  for all slip systems, the effective dissipation potential is a homogeneous function of degree  $n + 1$ , and so is  $\tilde{W}^0$ , which then reads as

$$\tilde{W}^0(\bar{\varepsilon}_{33}^0) = \frac{n}{n+1} \gamma^0 \bar{\tau}^0 \left( \frac{\bar{\varepsilon}_{33}^0}{\gamma^0} \right)^{1+1/n}, \quad (36)$$

where  $\bar{\tau}^0$  denotes the overall reference stress of the polycrystal for uniaxial tension; it depends only on  $n$ , the shear reference stresses  $\tau_{(k)}^0$  and the microstructural parameters  $s$ . Finally, relations (35) and (36) ensure that  $\bar{\sigma}_{33}$  reads as

$$\bar{\sigma}_{33} = \bar{\tau}^0 \left( \frac{\bar{\varepsilon}_{33}^0}{\gamma^0} \right)^{1/n} \quad (37)$$

for a transversely isotropic polycrystal strained along the longitudinal direction or an isotropic polycrystal subjected to any uniaxial tensile test. Therefore, the reference stress  $\bar{\tau}^0$  governs both the overall strain potential and the overall stress–strain rate response.

In the following, second-order estimates for the overall response are compared to incremental estimates of the Hill-Hutchinson (1976) type, as well as to the tangent (Molinari et al., 1987; Lebensohn and Tomé, 1993) and the affine (Zaoui and Masson, 2000) ones. While these more “classical” schemes generate stress–strain relations, the second-order procedure produces estimates for the dissipation potential. The above property ensures that, for uniaxial tension along the symmetry axis, it is sufficient to compare the reference stresses  $\bar{\tau}^0$  as predicted by all these schemes. Details on the numerical derivation of these estimates can be found in Appendix B. All these estimates are also compared to the more rigorous variational estimates recently derived for similar situations by Nebozhyn et al. (2000, 2001). Although the latter are not rigorous upper bounds, they may be considered as such for the special class of phase distributions accurately described by the linear self-consistent scheme.

### 5.1. fcc polycrystals

In this section, untextured fcc polycrystals are considered for which inelastic deformation is assumed to take place by power-law glide with identical exponent  $n$  and reference shear stress  $\tau^0$  on all twelve octahedral slip systems.

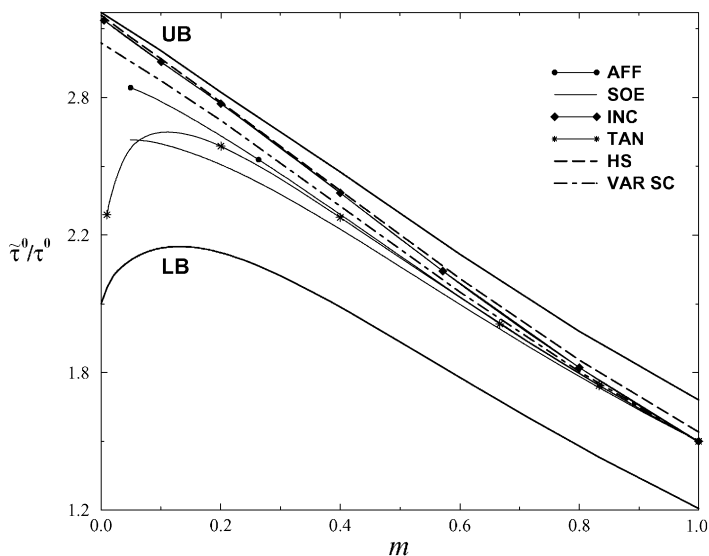


Fig. 1. Evolution of the normalised overall flow stress  $\bar{\tau}^0/\tau^0$  of an untextured fcc polycrystal with the strain-rate sensitivity  $m=1/n$ . Predictions of the self-consistent type according to second-order (SOE), affine (AFF), incremental (INC), tangent (TAN) and variational (VAR) procedures, as well as Hashin–Shtrikman (HS) and Taylor (UB) upper bounds and Reuss (LB) lower bound, are plotted for uniaxial tension.

Fig. 1 shows second-order estimates of the self-consistent type for the effective flow stress  $\bar{\tau}^0$  of these fcc polycrystals as a function of the strain-rate sensitivity  $m=1/n$  and compares them against various bounds and other self-consistent estimates. It is seen that the second-order estimates lie slightly higher than half way between the classical Taylor upper and Reuss lower bounds. This is consistent with what would be expected based on the corresponding results for linear polycrystals, where the SC estimate is well established. The second-order estimates are found to satisfy also the Hashin–Shtrikman bounds of deBotton and Ponte Castañeda (1995), which is not surprising in view of the fact that these remain quite close to the classical Taylor upper bound, especially with decreasing values of  $m$ . With respect to the various nonlinear SC estimates, there is wide discrepancy, especially with increasing nonlinearity, in spite of the fact that they all derive from the same classical linear SC estimates, with which they all agree for  $m=1$ . Consistent with earlier observations, the classical “incremental” SC estimates appears to be too stiff and to tend to the upper bound in the rate-insensitive limit. On the other hand, the tangent SC estimates, which rely on a linearisation scheme that is similar to that used for the second-order and affine estimates, are seen to tend to the Reuss lower bound in this limit (Masson et al., 2000). Finally, it is seen that while the affine estimates improve on the classical “incremental” estimates, they remain stiffer than the second-order predictions; both stay below the recently proposed “variational” estimates (Nebozhyn et al., 2001). It should be emphasised that slight differences for the overall flow stress may correspond to significant differences for the overall strain rates: according to (37), a 5% deviation between the affine and second-order predictions

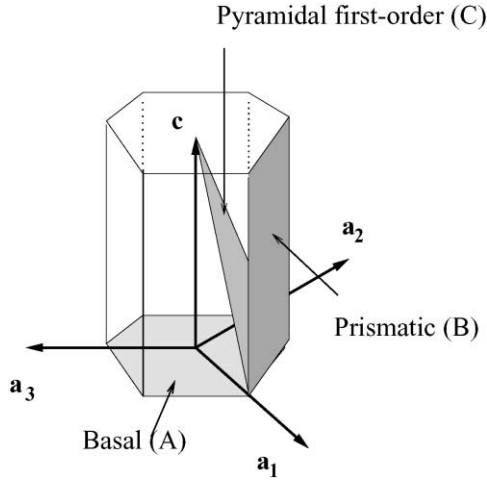


Fig. 2. Basal, prismatic and pyramidal first-order  $\langle c + a \rangle$  slip systems in an HCP crystal.

on the flow stress ( $n = 5$ ) corresponds to a 30% deviation on the predictions for the strain rates.

## 5.2. hcp polycrystals

Following Hutchinson's (1977) analysis of hcp and ionic fcc (Hutchinson, 1976) polycrystals and recent results obtained with use of the variational self-consistent method (Nebozhyn et al., 2000), in this section, the focus is on the overall response of polycrystals with highly anisotropic hcp grains.

### 5.2.1. Isotropic aggregates with the same power-law exponent

Referring to Fig. 2 for an hcp single crystal,  $\mathbf{a}$  and  $\mathbf{c}$  are used to denote the lattice vectors in the basal plane and along the hexagonal axis, respectively. Unlike fcc crystals, hcp crystals possess several different slip system families. The dominant mechanisms are generally prismatic or basal glide roughly depending on whether the value of the  $c/a$  ratio is lower (as in zirconium and titanium) or higher (as in ice) than the ideal value ( $=2\sqrt{2/3}$ ). In addition, the possibility of pyramidal first-order  $\langle c + a \rangle$  slip is also considered here. Following Hutchinson (1977), it will be assumed initially that glide on all systems obeys the power-law relation (29) with the same exponent  $n$ , but with generally different values of their flow shear stresses  $\tau_A^0$ ,  $\tau_B^0$  and  $\tau_C^0$ , corresponding to the basal, prismatic and pyramidal systems, respectively.

Figs. 3 and 4 show the evolution of the overall flow stress  $\tilde{\tau}^0$  as a function of the relevant anisotropy parameter  $M$  for isotropic zirconium ( $n = 5$ ,  $c/a = 1.59$ ,  $\tau_A^0 = \tau_B^0$ ,  $M = \tau_C^0/\tau_A^0$ ) and ice ( $n = 3$ ,  $c/a = 2\sqrt{2/3}$ ,  $\tau_C^0 = \infty$ ,  $M = \tau_B^0/\tau_A^0$ ) polycrystals. As previously, the second-order SC estimates are compared with the incremental, tangent and affine SC estimates, as well as with the lower and upper bounds. As noticed by Hutchinson (1977), the basal and prismatic families provide only four independent slip systems.

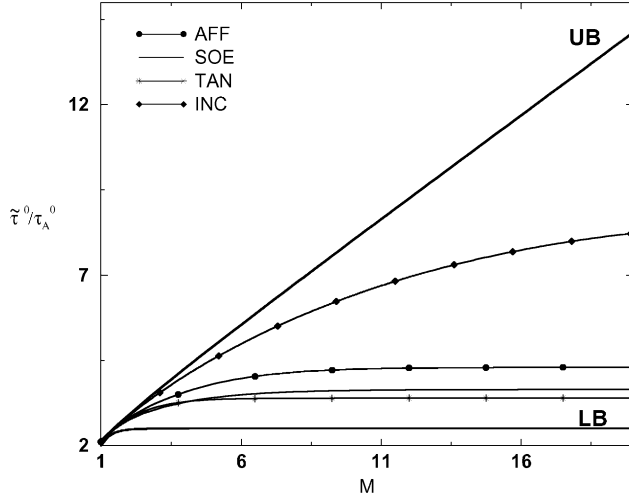


Fig. 3. Evolution, for a zirconium-type polycrystal, of the normalised overall flow stress  $\tilde{\tau}^0/\tau_A^0$  with the grain anisotropy parameter  $M = \tau_C^0/\tau_A^0$ . Predictions of the self-consistent type according to the second-order (SOE), affine (AFF), incremental (INC) and tangent (TAN) procedures, as well as Taylor (UB) and Reuss (LB) bounds, are plotted for uniaxial tension ( $\tau_B^0 = \tau_A^0$ ,  $n = 5$ ).

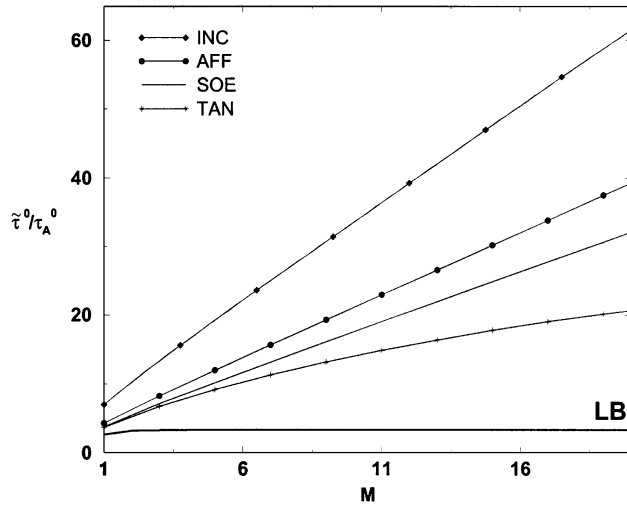


Fig. 4. Evolution, for an ice-type polycrystal, of the normalised overall flow stress  $\tilde{\tau}^0/\tau_A^0$  with the anisotropy parameter  $M = \tau_B^0/\tau_A^0$ . Predictions of the self-consistent type according to the second-order (SOE), affine (AFF), incremental (INC) and tangent (TAN) procedures, as well as Reuss lower bound (LB), are plotted for uniaxial tension ( $\tau_C^0 = \infty$ ,  $n = 3$ ).

Consequently, the overall strain rate as predicted by the Taylor upper bound tends to zero when the anisotropy parameter  $M = \tau_C^0/\tau_A^0$  tends to infinity (see Fig. 3). On the other hand, all the self-consistent estimates (and the lower bound), which allow grain-to-grain heterogeneity in the strain rate, predict a non-zero overall creep strain rate as the value of  $\tau_C^0$  tends to infinity. However, if, in addition to letting  $\tau_C^0 = \infty$ , the anisotropy parameter  $M = \tau_B^0/\tau_A^0$  is now allowed to become large, the effective flow stress of the polycrystal is predicted to become large by all SC estimates (see Fig. 4), although apparently at different rates.

In this connection, it is mentioned that based on recent estimates of the SC type (Nebozhyn et al., 2000, 2001) using the variational procedure of deBotton and Ponte Castañeda (1995), the following scaling law has been conjectured for the growth of the effective flow stress with increasing values of  $M$ :

$$\bar{\tau}^0 \approx M^{(4-i)/2}, \quad (38)$$

where  $i$  denotes the number of independent slip systems that are left in the limit as  $M$  tends to infinity. Figs. 3 and 4, which correspond to the values of  $i = 4$  and 2, respectively, show that classical bounds and tangent estimates do not obey this scaling law, whereas both the affine and second-order estimates do exhibit the expected zero growth (Fig. 3) and linear variation (Fig. 4) for large values of  $M$ , in agreement with relation (38). Note that previous simulations (Nebozhyn et al., 2000) have already shown that the incremental estimates are in disagreement with this scaling law at sufficiently high values of  $n$  for values of  $i$  greater than 2.

Fig. 5 shows the effect of the strain-rate sensitivity  $m$  on the various predictions for the overall properties of high contrast ( $M = \tau_B^0/\tau_A^0 = 10$ ;  $\tau_C^0 = \infty$ ) ice-type polycrystals. This case was also investigated recently (Nebozhyn et al., 2000) by means of the variational self-consistent method. In agreement with earlier observations in the context of fcc polycrystals, it is seen that the various SC estimates give widely diverging predictions as the level of nonlinearity increases ( $m \rightarrow 0$ ). At opposite extremes, the incremental<sup>1</sup> and tangent SC estimates are seen to become unbounded (and thus tend to the Taylor bound) and tend to the Reuss lower bound, respectively, in the rate-insensitive limit. On the other hand, the second-order and affine SC estimates remain distinct from either bound, with the second-order estimates again being somewhat softer than the affine estimates.

Experiments on both ice single crystals and polycrystals (see, for instance, Duval et al., 1983) show that inelastic deformation is mostly due to basal slip and, to a smaller extent, to prismatic slip. Estimates for the relevant grain anisotropy parameter  $M = \tau_B^0/\tau_A^0$  range from a value of about 10 to much higher values. It is also found experimentally that the polycrystal steady-creep response is generally well approximated by a power-law with an exponent equal to 3. With the assumption that the basal and prismatic slip systems only differ by their reference stresses, it has been found here (see Figs. 4 and 5) that the various SC models lead to relatively large differences in

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<sup>1</sup> For  $M = 10$ , previous calculations of incremental estimates (Hutchinson, 1977) turn out to be too stiff. In accordance with our simulations as well as results computed independently (Nebozhyn et al., 2000, Fig. 2b),  $\bar{\tau}^0/\tau_A^0 \approx 34$  is the accurate value of the incremental estimate when  $M = 10$ ,  $n = 3$ ,  $\tau_C^0 = \infty$ .



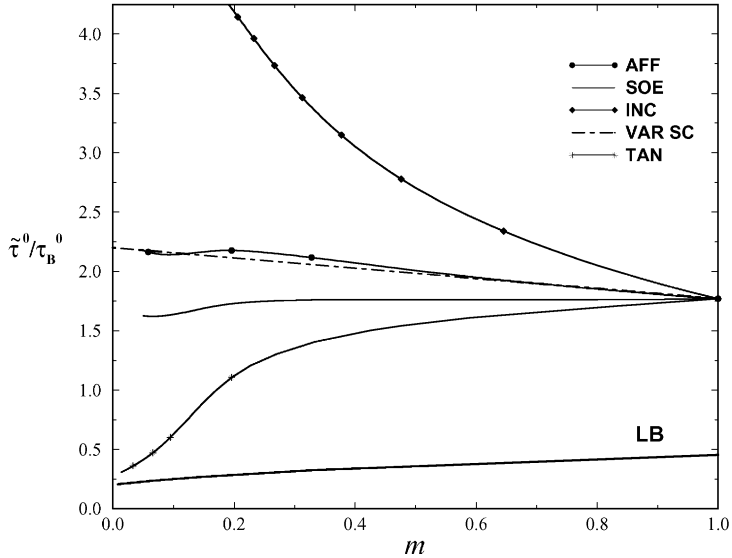


Fig. 5. Evolution, for an ice-type polycrystal ( $M = \tau_B^0/\tau_A^0 = 10$ ,  $\tau_C^0 = \infty$ ), of the normalised overall flow stress  $\tilde{\tau}^0/\tau_B^0$  with the strain rate sensitivity  $m = 1/n$ . Predictions of the self-consistent type according to the second-order (SOE), affine (AFF), incremental (INC), tangent (TAN) and variational (VAR) procedures, as well as Reuss lower bound, are plotted for uniaxial tension.

their predictions for the overall response of ice polycrystals at these large values of the grain anisotropy parameter. Based on the information that is available at the moment, it appears that the incremental and tangent estimates are, respectively, too stiff and too soft and must be discarded in favour of more accurate estimates. The second-order and affine estimates seem to offer the most accurate predictions to date, although the work of Bornert and Ponte Castañeda (1998) on model 2D polycrystals, for which there is a sharp bound, suggests that the second-order estimates should be the more accurate of the two, since the affine model can violate this sharp bound for extreme values of the grain anisotropy and nonlinearity parameters.

### 5.2.2. Anisotropic strain-rate sensitivity of polycrystalline ice

Inelastic glide in hcp crystals frequently derives from different mechanisms on different slip families. Therefore, shear glide on different families does not generally obey the same flow law. For polycrystalline ice, the basal power-law exponent is known to be close to 2 (Duval et al., 1983) while the prismatic is about 3. Unlike the tangent model and like the affine model, the second-order procedure is not limited to power-law behaviour with identical exponents on the different slip systems. Nevertheless, the second-order treatment of this situation is less straightforward than with the affine model because of the fact that relation (37) breaks down and the overall stress has to be determined numerically (see Appendix B).

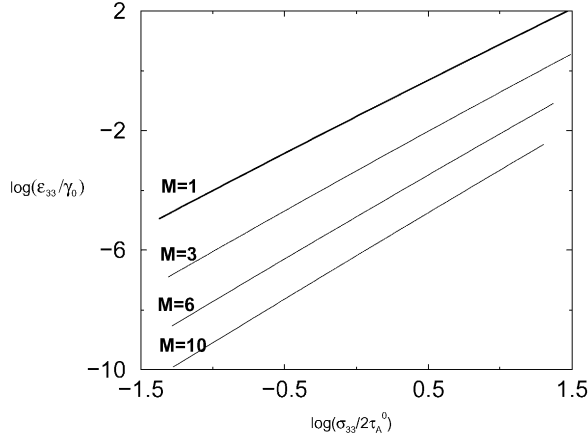


Fig. 6. Second-order predictions of the self-consistent type for the stress–strain overall response (in logarithmic scales) of untextured ice polycrystals with increasing values of the anisotropy parameter  $M$  ( $M = \tau_B^0/\tau_A^0$ , basal and prismatic strain-rate sensitivities equal to 2 and 3, respectively).

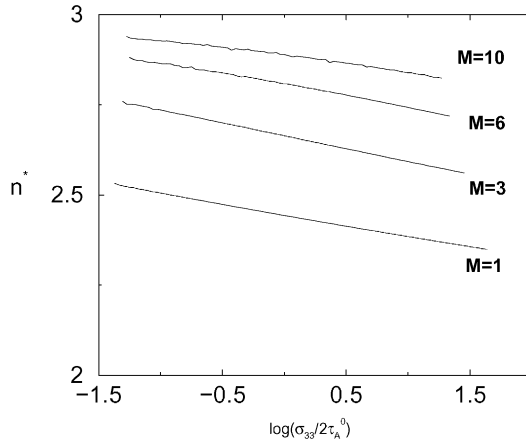


Fig. 7. Second-order predictions of the self-consistent type for the overall instantaneous power-law exponent  $n^*$  with increasing values of the anisotropy parameter  $M$  ( $M = \tau_B^0/\tau_A^0$  basal and prismatic power-law exponents equal to 2 and 3, respectively).

In Fig. 6, we have reported various stress–strain rate responses as predicted by the second-order procedure (logarithmic scale) for different values of the anisotropy parameter  $M$ . As reported in Fig. 7, the overall instantaneous power-law exponent  $n^*$  decreases slightly with increasing stresses while the mean slope ranges from 2.5 to 3 with increasing contrast. The choice of an overall power law with an exponent of 3, first proposed by Hutchinson (1977), is therefore quite relevant for an anisotropy

parameter larger than 10. But a lower exponent associated with basal slip undoubtedly affects the corresponding overall reference stress: for  $M = 10$ ,  $\tilde{\tau}^0$  drops from  $17\tau_A^0$  (identical power-law exponents) to approximately  $14\tau_A^0$  (this calculation).

### 5.2.3. Texture effects

As a result of the anisotropy at the single crystal level, hexagonal materials often display crystallographic textures. For instance, the  $\langle c \rangle$  axes of ice single crystals in low-dome and grip-ice core polycrystal specimens (Castelnau et al., 1997) are concentrated around the vertical direction. In this last section, the effect of such an initial texture on the steady-creep response of ice is considered. To represent texture effects in low-dome or grip-ice core specimens of ice, an isotropic distribution of the  $\langle c \rangle$  axes is weighted by

$$p_w(\phi) = \frac{2\alpha_w}{\pi} \exp\left(-4w \left(\frac{\phi}{\pi} - \frac{1}{2}\right)^2\right) \quad \text{for } w \geq 0, \quad (39)$$

$$p_w(\phi) = \frac{2\alpha_w}{\pi} \exp\left(4w \left(\frac{\phi}{\pi}\right)^2\right) \quad \text{for } w \leq 0, \quad (40)$$

where  $\phi$  denotes the angle between the tensile direction and the  $\langle c \rangle$  axis ( $0 \leq \phi \leq \pi/2$ ),  $w$  is the texture parameter and  $\alpha_w$  is a normalisation factor such that

$$\int_0^{\pi/2} p_w(\phi) d\phi = 1. \quad (41)$$

High values of the texture parameter correspond to the  $\langle c \rangle$  axes located mainly in the transverse plane while decreasing  $w$  leads to make more  $\langle c \rangle$  axes parallel to the tensile axis.

The second-order predictions for the overall reference stress with respect to this texture parameter, as well as the incremental, tangent, affine estimates and the lower bound, have been reported in Fig. 8 (the slight inflexion displayed by these curves at  $w = 0$  is due to the fact that the derivative of  $p_w(\phi)$  with respect to  $w$  is not continuous at  $w = 0$ ). Schematic pole figures associated with different values of the texture parameter ( $w = -10, -5, 5$  and  $10$ ) have been reported in this figure too: dotted circles correspond to average orientations while dashed areas contain up to 80% of orientations around these averages. When the  $\langle c \rangle$  axes concentrate around the tensile axis, accommodation of the overall inelastic strain rate throughout the polycrystal becomes harder and the predictions of the overall reference stress increase for decreasing values of the texture parameter. Low negative values of this parameter, which correspond to realistic situations (e.g. low dome or grip ice core specimens), lead to significant deviations between estimates due to the high contrast between the phases.

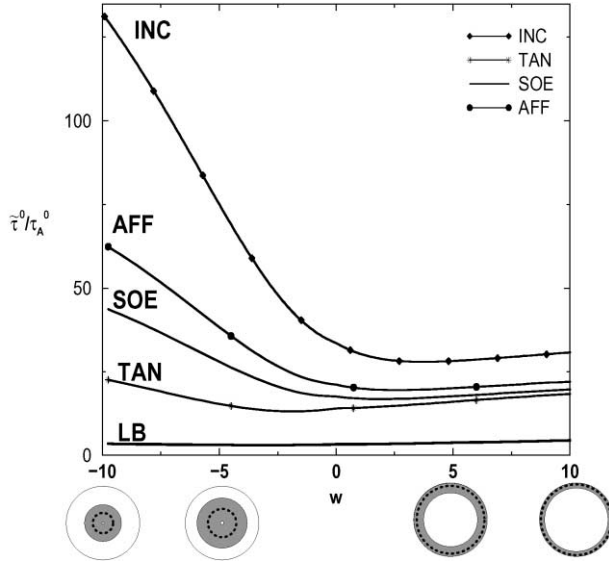


Fig. 8. Evolution, for a textured ice-type polycrystal, of the normalised overall flow stress  $\bar{\tau}^0/\tau_A^0$  with the texture parameter  $w$  (anisotropy parameter  $M = 10$ ,  $n = 3$ ). Predictions of the self-consistent type according to the second-order (SOE), affine (AFF), incremental (INC) and tangent (TAN) procedures, as well as Reuss lower bound (LB), are plotted for uniaxial tension along the symmetry axis. The dotted circles in the schematic pole figures along the symmetry axis under each associated  $w$  value correspond to average orientations, whereas dashed areas contain up to 80% of orientations around these averages.

## 6. Conclusion

The above developments show that the second-order procedure, which has already been shown to lead to accurate estimates for the effective behaviour of two-phase materials, can also be easily combined with the linear Hashin–Shtrikman estimates in order to model the overall response of a wide variety of multiphase nonlinear elastic or viscoplastic materials, and especially with the linear self-consistent scheme which is more appropriate for polycrystalline materials. Extensions to finite strains, which were not fully detailed here, can also be easily developed. As illustrated by the above results, the corresponding estimates compare very satisfactorily with all known estimates and bounds. The known limitation of the procedure, namely the “duality gap” between stress and strain approaches, can be considered, from a practical point of view, as a minor one if one is careful to choose the appropriate formulation (stress versus strain) for the intended application. In conclusion, it can be considered, at the moment, as the most general and accurate homogenisation method for heterogeneous materials whose constitutive equations derive from one single potential. The need for a similar method for more complex constitutive behaviours such as rate-dependent or rate-independent elastoplasticity is still satisfied by the less accurate “affine” formulation, where variational approaches are not available.

## Acknowledgements

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## Appendix A. Hashin–Shtrikman estimates for a thermoelastic multiphase composite

The full classical variational formulation of the linear thermoelastic local problem (13) reads as

$$\tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \boldsymbol{l}) = \min_{\boldsymbol{\varepsilon} \in \mathcal{H}(\bar{\boldsymbol{\varepsilon}})} \left\langle \frac{1}{2} \boldsymbol{\varepsilon} \cdot \boldsymbol{L} \boldsymbol{\varepsilon} - \boldsymbol{l} \cdot \boldsymbol{\varepsilon} \right\rangle = -\min_{\boldsymbol{\sigma} \in \mathcal{S}} \left\langle \frac{1}{2} (\boldsymbol{\sigma} + \boldsymbol{l}) \cdot \boldsymbol{M} (\boldsymbol{\sigma} + \boldsymbol{l}) - \boldsymbol{\sigma} \cdot \bar{\boldsymbol{\varepsilon}} \right\rangle, \quad (\text{A.1})$$

where  $\mathcal{S}$  is the set of statically admissible fields on  $\Omega$  and  $\boldsymbol{M} = \boldsymbol{L}^{-1}$ . Given the linearity of this problem, its solution takes the form  $\boldsymbol{\varepsilon} = \boldsymbol{A} : \bar{\boldsymbol{\varepsilon}} - \boldsymbol{a}$  where  $\boldsymbol{A}$  (resp.  $\boldsymbol{a}$ ) is a fourth- (resp. second-) order tensor field, so that the average strains in the phases, required to generate second-order estimates, take the form

$$\langle \boldsymbol{\varepsilon} \rangle^{(r)} = \boldsymbol{A}^{(r)} \bar{\boldsymbol{\varepsilon}} - \boldsymbol{a}^{(r)} \quad (\text{A.2})$$

with  $\boldsymbol{A}^{(r)} = \langle \boldsymbol{A} \rangle^{(r)}$  and  $\boldsymbol{a}^{(r)} = \langle \boldsymbol{a} \rangle^{(r)}$ . The relations  $\langle (\boldsymbol{\varepsilon} - \boldsymbol{A} \bar{\boldsymbol{\varepsilon}}) \cdot (\boldsymbol{L} \boldsymbol{\varepsilon} - \boldsymbol{l}) \rangle = 0$  and  $\langle \boldsymbol{L} \boldsymbol{A} \boldsymbol{\varepsilon} \cdot \boldsymbol{\varepsilon} \rangle = \langle \boldsymbol{L} \boldsymbol{A} \rangle \bar{\boldsymbol{\varepsilon}} \cdot \bar{\boldsymbol{\varepsilon}}$  result from two applications of Hill’s macrohomogeneity condition and allow us to rewrite the overall potential as

$$\tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \boldsymbol{l}) = \frac{1}{2} \bar{\boldsymbol{\varepsilon}} \cdot \boldsymbol{L} \bar{\boldsymbol{\varepsilon}} - \bar{\boldsymbol{\varepsilon}} \cdot \tilde{\boldsymbol{l}} + \frac{1}{2} \tilde{\boldsymbol{f}} \quad \text{with} \quad \begin{cases} \tilde{\boldsymbol{L}} = \langle \boldsymbol{L} \boldsymbol{A} \rangle = \sum_r c_r \boldsymbol{L}^{(r)} \boldsymbol{A}^{(r)}, \\ \tilde{\boldsymbol{l}} = \langle \boldsymbol{l} \boldsymbol{A} \rangle = \sum_r c_r \boldsymbol{l}^{(r)} \boldsymbol{A}^{(r)}, \\ \tilde{\boldsymbol{f}} = \langle \boldsymbol{l} \cdot \boldsymbol{a} \rangle = \sum_r c_r \boldsymbol{l}^{(r)} \cdot \boldsymbol{a}^{(r)}. \end{cases} \quad (\text{A.3})$$

$\tilde{\boldsymbol{L}}$  is the tensor of effective moduli,  $-\tilde{\boldsymbol{l}}$  is the effective thermal stress and  $-\frac{1}{2} \tilde{\boldsymbol{f}}$  is the overall potential under vanishing overall deformation, related to thermal strain incompatibilities. The effective stress–strain relation is then given by  $\bar{\boldsymbol{\sigma}} = \tilde{\boldsymbol{L}} \bar{\boldsymbol{\varepsilon}} - \tilde{\boldsymbol{l}}$  and its derivation requires only the knowledge of the linear localisation tensors  $\boldsymbol{A}^{(r)}$ . The computation of the effective potential or the per phase average strain requires in addition the determination of the tensors  $\boldsymbol{a}^{(r)}$ , which give the strain induced by the thermal stresses under vanishing overall strain. It is the purpose of this appendix to generate Hashin–Shtrikman and, as a particular case, self-consistent-type estimates for these quantities. The procedure has been outlined in Willis (1981). Here it is recalled in a slightly different manner and explicit expressions are given. In particular, it is shown that such estimates are based on exactly the same assumptions on the statistics of the phase distribution of the considered multiphase composite as the linear classical Hashin–Shtrikman estimates, so that such linear estimates have “natural” extensions for

thermoelasticity, and so that any Hashin–Shtrikman type linear estimate can be used to generate second-order estimates for nonlinear behaviours. The results obtained by Laws (1973) in the different context of Eshelby-type considerations are recovered for the classical self-consistent scheme.

For any distribution of second-order tensors  $\boldsymbol{\eta}$  on  $\Omega$  and any reference tensor of elastic moduli  $\mathbf{L}^0 = [\mathbf{M}^0]^{-1}$ , let  $\boldsymbol{\varepsilon}'(\boldsymbol{\eta})$  and  $\boldsymbol{\sigma}'(\boldsymbol{\eta})$  be the strain and stress field solution of the following auxiliary thermoelastic problem:

$$\boldsymbol{\sigma}' = \mathbf{L}^0 \boldsymbol{\varepsilon}' + [\mathbf{L} - \mathbf{L}^0] \boldsymbol{\eta} - \mathbf{l}, \quad \boldsymbol{\varepsilon}' \in \mathcal{H}(\bar{\boldsymbol{\varepsilon}}) \quad \text{and} \quad \boldsymbol{\sigma}' \in \mathcal{S}. \quad (\text{A.4})$$

These fields can be used as trial fields in the variational formulation (A.2) for the initial thermoelastic problem on the heterogeneous body

$$-\Delta^- + \text{HS}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}; \boldsymbol{\eta}) \leq \tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}) \leq \text{HS}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}; \boldsymbol{\eta}) + \Delta^+ \quad (\text{A.5})$$

$$\text{with} \quad \begin{cases} 2\Delta^+ = \langle (\boldsymbol{\varepsilon}' - \boldsymbol{\eta}) \cdot (\mathbf{L} - \mathbf{L}^0)(\boldsymbol{\varepsilon}' - \boldsymbol{\eta}) \rangle \\ 2\Delta^- = \langle \mathbf{L}^0(\boldsymbol{\varepsilon}' - \boldsymbol{\eta}) \cdot (\mathbf{M} - \mathbf{M}^0)\mathbf{L}^0(\boldsymbol{\varepsilon}' - \boldsymbol{\eta}) \rangle \end{cases} \quad (\text{A.6})$$

and where  $\text{HS}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}; \boldsymbol{\eta})$  is the Hashin–Shtrikman functional given by

$$\begin{aligned} 2\text{HS}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}; \boldsymbol{\eta}) &= \bar{\boldsymbol{\varepsilon}} \cdot \mathbf{L}^0 \bar{\boldsymbol{\varepsilon}} + \langle (\bar{\boldsymbol{\varepsilon}} + \boldsymbol{\varepsilon}' - \boldsymbol{\eta}) \cdot (\mathbf{L} - \mathbf{L}^0) \boldsymbol{\eta} \rangle - \langle (\bar{\boldsymbol{\varepsilon}} + \boldsymbol{\varepsilon}') \cdot \mathbf{l} \rangle \\ &= \langle (\mathbf{L} - \mathbf{L}^0) \boldsymbol{\eta} \cdot (2\boldsymbol{\varepsilon}' - \boldsymbol{\eta}) + \boldsymbol{\varepsilon}' \cdot \mathbf{L}^0 \boldsymbol{\varepsilon}' - 2\mathbf{l} \cdot \boldsymbol{\varepsilon}' \rangle. \end{aligned} \quad (\text{A.7})$$

Inequalities (A.5) are derived from (A.1) by some simple algebraic manipulations using Hill's relation  $\langle (\mathbf{L}^0 \boldsymbol{\varepsilon}' + [\mathbf{L} - \mathbf{L}^0] \boldsymbol{\eta} - \mathbf{l}) \cdot (\bar{\boldsymbol{\varepsilon}} - \boldsymbol{\varepsilon}') \rangle = 0$ . Note that the classical expressions of HS as a function of the so-called polarisation  $\boldsymbol{\tau} = [\mathbf{L} - \mathbf{L}^0] \boldsymbol{\eta} - \mathbf{l}$  are recovered when  $\mathbf{l} = 0$ . The present ones do not involve  $[\mathbf{L} - \mathbf{L}^0]^{-1}$  and so do also apply to situations with  $\mathbf{L} - \mathbf{L}^0$  singular.

They show that  $\text{HS}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}; \boldsymbol{\eta})$  is an upper or lower bound for  $\tilde{\Psi}(\bar{\boldsymbol{\varepsilon}})$  when the reference medium  $\mathbf{L}^0$  is chosen such that  $\mathbf{L} - \mathbf{L}^0 \leq 0$  or  $\mathbf{M} - \mathbf{M}^0 \leq 0$ , respectively. Optimal bounds are obtained when  $\text{HS}(\bar{\boldsymbol{\varepsilon}}; \boldsymbol{\eta})$  is minimised or maximised with respect to  $\boldsymbol{\eta}$ . The variation of  $\text{HS}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}; \boldsymbol{\eta})$  is computed as

$$\frac{\partial \text{HS}}{\partial \boldsymbol{\eta}}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}; \boldsymbol{\eta}) \cdot \delta \boldsymbol{\eta} = \langle (\mathbf{L} - \mathbf{L}^0)(\boldsymbol{\varepsilon}' - \boldsymbol{\eta}) \cdot \delta \boldsymbol{\eta} \rangle + \left\langle (\mathbf{L}^0 \boldsymbol{\varepsilon}' + (\mathbf{L} - \mathbf{L}^0) \boldsymbol{\eta} - \mathbf{l}) \cdot \frac{\partial \boldsymbol{\varepsilon}'}{\partial \boldsymbol{\eta}} \delta \boldsymbol{\eta} \right\rangle.$$

The second term of the last expression is identically zero since  $(\partial \boldsymbol{\varepsilon}' / \partial \boldsymbol{\eta}) \delta \boldsymbol{\eta}$  is an admissible strain field compatible with zero displacement on  $\partial \Omega$  and  $\boldsymbol{\sigma}' = \mathbf{L}^0 \boldsymbol{\varepsilon}' + (\mathbf{L} - \mathbf{L}^0) \boldsymbol{\eta} - \mathbf{l}$  is equilibrated. The optimal field  $\boldsymbol{\eta}^0$  is thus characterised by

$$\forall \delta \boldsymbol{\eta} \quad \langle (\mathbf{L} - \mathbf{L}^0)(\boldsymbol{\varepsilon}' - \boldsymbol{\eta}^0) \cdot \delta \boldsymbol{\eta} \rangle = 0. \quad (\text{A.8})$$

Assuming that  $\boldsymbol{\eta}$  varies in a vectorial space, it can be substituted for  $\delta \boldsymbol{\eta}$  in this relation and the following expression of the optimal value of HS is obtained:

$$2\text{HS}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}; \boldsymbol{\eta}^0) = \bar{\boldsymbol{\varepsilon}} \cdot \mathbf{L}^0 \bar{\boldsymbol{\varepsilon}} + \bar{\boldsymbol{\varepsilon}} \cdot \langle (\mathbf{L} - \mathbf{L}^0) \boldsymbol{\eta}^0 - \mathbf{l} \rangle - \langle \boldsymbol{\varepsilon}'(\boldsymbol{\eta}^0) \cdot \mathbf{l} \rangle. \quad (\text{A.9})$$

When  $\boldsymbol{\eta}$  is searched for in the whole space of symmetric second-order tensor fields on  $\Omega$ , and when  $\mathbf{L} - \mathbf{L}^0$  is regular, Eq. (A.8) leads simply to  $\boldsymbol{\varepsilon}' = \boldsymbol{\eta}^0$ , so that the fields generated in the auxiliary thermoelastic problem (A.4) coincide with the exact solution of the initial problem and  $\text{HS}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}; \boldsymbol{\eta}^0)$  is equal to  $\tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l})$ . The same property holds

when  $\mathbf{L} - \mathbf{L}^0$  is singular, except that  $\boldsymbol{\eta}$  is not defined uniquely: since only  $(\mathbf{L} - \mathbf{L}^0)\boldsymbol{\eta}$  is relevant for problem (A.4) one may choose to enforce the condition  $\boldsymbol{\varepsilon}' = \boldsymbol{\eta}^0$  among all possible optimal choices of  $\boldsymbol{\eta}$  which lead to the same fields  $\boldsymbol{\varepsilon}'$  and values of HS. As a consequence, one has the general stationarity property

$$\tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \mathbf{I}) = \underset{\boldsymbol{\eta}}{\text{Stat HS}}(\bar{\boldsymbol{\varepsilon}}; \mathbf{I}; \boldsymbol{\eta}), \quad (\text{A.10})$$

which, as already mentioned, specialises into a minimum or a maximum condition when  $\mathbf{L} - \mathbf{L}^0$  is positive or negative (not necessarily definite).

In practice, the optimisation of  $\boldsymbol{\eta}$  in the whole set of symmetric second-order tensor fields on  $\Omega$  is not possible and one performs it on a subset which is suited to the microstructure and the information that is available on it. In a classical description of multiphase materials, piecewise fields uniform over each constitutive phase make an appropriate choice (Willis, 1981). Optimality condition (A.9) then reads as  $\forall r \langle \boldsymbol{\varepsilon}' \rangle^{(r)} = \boldsymbol{\eta}^{(r)}$ , where the tensors  $\boldsymbol{\eta}^{(r)}$  are the values  $\boldsymbol{\eta}$  take on each phase. Note that this is the unique optimal choice for  $\boldsymbol{\eta}$  when  $\mathbf{L} - \mathbf{L}^0$  is regular and that, when not, it is one choice between several possible but all equivalent ones: all of them lead to the same value of the polarisation  $\boldsymbol{\tau} = [\mathbf{L} - \mathbf{L}^0]\boldsymbol{\eta} - \mathbf{I}$ , and thus of the local fields  $\boldsymbol{\sigma}'$  and  $\boldsymbol{\varepsilon}'$  in the auxiliary thermoelastic problem, which are used as Hashin–Shtrikman-type estimates of the local fields  $\boldsymbol{\sigma}$  and  $\boldsymbol{\varepsilon}$  in the heterogeneous thermoelastic material. This optimality condition, together with the equations of the auxiliary thermoelastic problem (A.4), determine completely the per-phase average strains that are sought in the context of the second-order procedure. At this stage it should be noted that the microstructural information that is involved to determine them is the one that is required to address problem (A.4), which is identical to the corresponding problem that would have been addressed in the purely linear elastic case: this is the fundamental reason which allows one to naturally extend any linear Hashin–Shtrikman-type bound or estimate to thermoelasticity and consecutively to generate second-order estimates for nonlinear behaviour.

Within the limit that the scale of the fluctuations of the phase distribution is small with respect to the overall dimensions of  $\Omega$ , problem (A.4) is classically solved by means of the modified strain Green's operator  $\Gamma^0$  associated with an infinite medium by

$$\boldsymbol{\varepsilon}'(\mathbf{x}) = \bar{\boldsymbol{\varepsilon}} - \int_{V(\mathbf{x})} \Gamma^0(\mathbf{x} - \mathbf{x}') [\boldsymbol{\tau}(\mathbf{x}') - \langle \boldsymbol{\tau} \rangle] dV', \quad (\text{A.11})$$

where  $V(\mathbf{x})$  is a sufficiently large neighbourhood of point  $\mathbf{x}$ . This general relation leads to the following expression of the per-phase average strains  $\langle \boldsymbol{\varepsilon} \rangle^{(r)}$ :

$$\langle \boldsymbol{\varepsilon} \rangle^{(r)} = \bar{\boldsymbol{\varepsilon}} - \frac{1}{c_r} \sum_s \mathcal{P}_{rs} \boldsymbol{\tau}^{(r)} \quad (\text{A.12})$$

with  $\boldsymbol{\tau}^{(r)} = (\mathbf{L}^{(r)} - \mathbf{L}^0)\boldsymbol{\eta}^{(r)} - \mathbf{I}^{(r)}$  and

$$\mathcal{P}_{rs} = \int_{V(0)} \Gamma^0(\mathbf{h})(C_{rs}(\mathbf{h}) - c_r c_s) dV_{\mathbf{h}}, \quad (\text{A.13})$$

where  $C_{rs}(\mathbf{h}) = \langle \chi^{(r)}(\cdot) \chi^{(s)}(\cdot + \mathbf{h}) \rangle$  is the two-point correlation function relative to phases  $r$  and  $s$  equal to  $c_r \delta_{rs}$  for  $\mathbf{h} = 0$  and tending to  $c_r c_s$  for large  $\mathbf{h}$ , assuming there is no

long range order in the phase distribution. These tensors  $\mathcal{P}_{rs}$  are the only quantities that involve microstructural information and are involved identically in both Hashin–Shtrikman-type formulations, relative to linear elasticity or to linear thermoelasticity. The average strain fields in the second case are thus completely determined by the following coupled system of tensorial equations:

$$\forall r \quad \langle \boldsymbol{\varepsilon} \rangle^{(r)} + \frac{1}{c_r} \sum_s \mathcal{P}_{rs} [(\mathbf{L}^{(s)} - \mathbf{L}^0) \langle \boldsymbol{\varepsilon} \rangle^{(s)} - \mathbf{l}^{(s)}] = \bar{\boldsymbol{\varepsilon}}. \quad (\text{A.14})$$

Assuming the operators  $\mathcal{P}_{rs}$  as known, this system could be solved numerically for any phase distribution. In practice, one assumes classically isotropic or more generally ellipsoidal (Willis, 1977) symmetry of the microstructure, such that  $C_{rs}(\mathbf{h}) = \phi(|\boldsymbol{\alpha}\mathbf{h}|)$ , where the symmetric second-order tensor  $\boldsymbol{\alpha}$  characterises the anisotropy of such a particular phase distribution. In this case, as a consequence of the particular properties of  $\mathbf{I}^0$ , the operators  $\mathcal{P}_{rs}$  can be computed exactly and only depend on  $\boldsymbol{\alpha}$ , not on  $\phi$ . One gets

$$\mathcal{P}_{rs} = c_r(\delta_{rs} - c_s)\mathbf{P}^0, \quad (\text{A.15})$$

where  $\mathbf{P}^0$  is Hill's tensor (Hill, 1965) associated with an ellipsoidal inclusion whose shape and orientation are determined by the tensor  $\boldsymbol{\alpha}$ . The system of equations is then much less coupled and can be rewritten as

$$\forall r \quad \langle \boldsymbol{\varepsilon} \rangle^{(r)} + \mathbf{P}^0 [(\mathbf{L}^{(r)} - \mathbf{L}^0) \langle \boldsymbol{\varepsilon} \rangle^{(r)} - \mathbf{l}^{(r)}] = \boldsymbol{\varepsilon}^1, \quad (\text{A.16})$$

$$\boldsymbol{\varepsilon}^1 = \bar{\boldsymbol{\varepsilon}} + \mathbf{P}^0 \langle (\mathbf{L} - \mathbf{L}^0) \boldsymbol{\varepsilon} - \mathbf{l} \rangle.$$

Note that the auxiliary strain  $\boldsymbol{\varepsilon}^1$  can also be conveniently determined by the condition  $\sum_r c_r \langle \boldsymbol{\varepsilon} \rangle^{(r)} = \bar{\boldsymbol{\varepsilon}}$ . The explicit computation of  $\langle \boldsymbol{\varepsilon} \rangle^{(r)}$  is then easy and leads to the estimates  $\mathbf{A}_{\text{HS}}^{(r)}$  and  $\mathbf{a}_{\text{HS}}^{(r)}$  of  $\mathbf{A}^{(r)}$  and  $\mathbf{a}^{(r)}$ :

$$\mathbf{A}_{\text{HS}}^{(r)} = [\mathbf{L}^{(r)} + \mathbf{L}_0^*]^{-1} \left\{ \sum_s c_s [\mathbf{L}^{(s)} + \mathbf{L}_0^*]^{-1} \right\}^{-1}, \quad (\text{A.17})$$

$$\mathbf{a}_{\text{HS}}^{(r)} = [\mathbf{L}^{(r)} + \mathbf{L}_0^*]^{-1} [\tilde{\mathbf{l}}_{\text{HS}} - \mathbf{l}^{(r)}] \quad (\text{A.18})$$

with

$$\tilde{\mathbf{l}}_{\text{HS}} = \sum_s c_s \mathbf{l}^{(s)} \mathbf{A}_{\text{HS}}^{(r)} \quad (\text{A.19})$$

and where  $\mathbf{L}_0^* = [\mathbf{P}^0]^{-1} - \mathbf{L}^0$  is Hill's constraint tensor, relative to  $\mathbf{C}^0$  and to the ellipsoid defined by  $\boldsymbol{\alpha}$ .

These relations hold for any choice of reference medium  $\mathbf{L}^0$ . Self-consistent-type estimates are generated when, in addition, the self-consistency condition

$$\mathbf{L}^0 = \tilde{\mathbf{L}}_{\text{HS}}(\mathbf{L}^0) = \sum_s c_s \mathbf{L}^{(s)} \mathbf{A}_{\text{HS}}^{(r)} \quad (\text{A.20})$$

is enforced. Such a choice is governed by the fact that the difference between  $\tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l})$  and  $\text{HS}(\bar{\boldsymbol{\varepsilon}}; \mathbf{l}; \boldsymbol{\eta}^0)$ , which is bounded by  $\text{Sup}(|\Delta^+|, |\Delta^-|)$  given by Eq. (A.6), might be



close to a minimum, since positive contributions might be counterbalanced by negative ones in the averages that define  $\Delta^+$  and  $\Delta^-$ , so that  $\text{HS}(\bar{\boldsymbol{\varepsilon}}; \boldsymbol{L}; \boldsymbol{\eta}^0)$  provides an accurate estimate of  $\tilde{\Psi}(\bar{\boldsymbol{\varepsilon}}; \boldsymbol{L})$  for this choice of reference medium. Note that this consideration holds both for linear and thermoelastic composites. Condition (A.20) is equivalent to  $\sum_s c_s [\boldsymbol{L}^{(s)} + \boldsymbol{L}_0^\star]^{-1} = \boldsymbol{P}^0$  which induces expression (24) for the tensors  $A_{\text{HS}}^{(r)}$ , now noted as  $A_{\text{SC}}^{(r)}$ , the self-consistent estimate  $\tilde{\boldsymbol{L}}_{\text{SC}}$  for the effective tensor of moduli being the solution of the implicit Eq. (25). The tensors  $\boldsymbol{a}_{\text{SC}}^{(r)}$  are still given by Eq. (A.18), with  $A_{\text{SC}}^{(r)}$  substituted for  $A_{\text{HS}}^{(r)}$  and  $\boldsymbol{L}_{\text{SC}}^\star$  for  $\boldsymbol{L}_0^\star$ , which leads to relation (27).

The above relations can be found in Laws (1973), where they had been obtained in a different context: it was postulated that, in extension of the classical interpretation of the self-consistent scheme in elasticity, the average strain in phase ( $r$ ) was identical to the uniform strain that is generated in a thermoelastic ellipsoidal inclusion, with uniform moduli  $\boldsymbol{L}^{(r)}$  and thermal stress  $\boldsymbol{l}^{(r)}$ , embedded in an infinite medium with moduli  $\tilde{\boldsymbol{L}}$  and thermal stress  $\tilde{\boldsymbol{l}}$ , and submitted at infinity to the macroscopic strain  $\bar{\boldsymbol{\varepsilon}}$ . Here this property appears as a consequence of the simplification of the local interactions in the heterogeneous thermoelastic composite by means of the Hashin–Shtrikman analysis, and of the particular assumption on the phase distribution. It is shown that the Eshelby-type inclusion to be considered is related to the overall ellipsoidal symmetry of the phase distribution, and should not be seen as a particular individual inclusion in the microstructure. However, in the case of a polycrystal in which the lattice orientation in each grain is uncorrelated with the orientation in the other grains, the shape of the inclusion can be understood as the average shape of the grains and may serve to characterise the morphological texture of the polycrystal.

## Appendix B. Computational aspects

### B.1. Derivation of the reference strains

The computation of the second-order estimate for the overall potential  $\tilde{W}(\bar{\boldsymbol{\varepsilon}})$  requires the computation of the reference strains  $\bar{\boldsymbol{\varepsilon}}^{(r)}$  as the solutions of the system of equations (28), for any applied macroscopic strain  $\bar{\boldsymbol{\varepsilon}}$ . Since  $w^{(r)}$  is only known as the Legendre transform of  $u^{(r)}$  for the type of nonlinear behaviours under consideration here, no explicit relation is available for the computation of the quantities  $\boldsymbol{\rho}^{(r)}$  as a function of the  $\bar{\boldsymbol{\varepsilon}}^{(r)}$ . That is why it is more convenient to consider the stresses  $\boldsymbol{\rho}^{(r)}$  as main unknowns and to rewrite system (28) as follows:

$$\boldsymbol{M}^{(r)} = \frac{\partial^2 u^{(r)}}{\partial \boldsymbol{\sigma} \partial \boldsymbol{\sigma}}(\boldsymbol{\rho}^{(r)}),$$

$$\tilde{\boldsymbol{M}}_{\text{SC}} = \tilde{\boldsymbol{L}}_{\text{SC}}^{-1} = \sum c_s \boldsymbol{M}^{(s)} : \boldsymbol{B}_{\text{SC}}^{(s)},$$

$$\boldsymbol{B}_{\text{SC}}^{(r)} = (\boldsymbol{M}^{(r)} + \boldsymbol{M}_{\text{SC}}^\star)^{-1} (\boldsymbol{M}_{\text{SC}} + \boldsymbol{M}_{\text{SC}}^\star),$$

$$\boldsymbol{M}_{\text{SC}}^\star = [\boldsymbol{L}_{\text{SC}}^\star]^{-1} = [\boldsymbol{P}_{\text{SC}}^{-1} - \tilde{\boldsymbol{L}}_{\text{SC}}]^{-1},$$

$$\begin{aligned}
\bar{\boldsymbol{\varepsilon}}^{(r)} - \bar{\boldsymbol{\varepsilon}} &= -\mathbf{M}_{\text{SC}}^{\star}[\boldsymbol{\rho}^{(r)} - \bar{\boldsymbol{\rho}}], \\
\bar{\boldsymbol{\varepsilon}}^{(r)} &= \frac{\partial u^{(r)}}{\partial \boldsymbol{\sigma}}(\boldsymbol{\rho}^{(r)}), \\
\bar{\boldsymbol{\varepsilon}} &= \sum_s c_s \bar{\boldsymbol{\varepsilon}}^{(s)}, \\
\bar{\boldsymbol{\rho}} &= \sum_s c_s \boldsymbol{\rho}^{(s)}.
\end{aligned} \tag{B.1}$$

Affine estimates (Zaoui and Masson, 2000) rely on exactly the same system of equations. Incremental (or total) estimates for power-law type polycrystals (Hutchinson, 1976) as well as tangent ones (Molinari et al., 1987; Lebensohn and Tomé, 1993) require the resolution of a similar set of equations, with  $\mathbf{M}^{(s)}$  replaced by the anisotropic secant compliances  $m\mathbf{M}^{(r)}$  ( $m = 1/n$ ), or with  $\boldsymbol{\beta}_{\text{SC}}^{(r)} = (m\mathbf{M}^{(r)} + \mathbf{M}_{\text{SC}}^{\star})^{-1}(m\mathbf{M}_{\text{SC}} + \mathbf{M}_{\text{SC}}^{\star})$  substituted for  $\mathbf{B}_{\text{SC}}^{(r)}$ , respectively (see Masson et al., 2000 for details). For all these estimates, the overall stress is given by  $\bar{\boldsymbol{\sigma}} = \bar{\boldsymbol{\rho}}$ , which is not the case for the second-order estimate (see below).

To solve this nonlinear system of equations for a prescribed overall strain  $\bar{\boldsymbol{\varepsilon}}$ , we make use of the following iterative procedure adapted from the one proposed by Molinari et al. (1987) for the tangent approach: step ( $k$ ) starts with the given initial values  $\boldsymbol{\rho}_{(k)}^{(r)}$  from which we derive the corresponding creep compliance  $\mathbf{M}_{(k)}^{(r)}$ . Once the overall creep compliance  $\tilde{\mathbf{M}}_{\text{SC}(k)}$  has been found as the solution of Eq. (B.1) by means of a classical fixed point algorithm (see below), the  $\boldsymbol{\rho}_{(k+1)}^{(r)}$  are obtained by solving the  $N$  uncoupled nonlinear equations

$$\frac{\partial u^{(r)}}{\partial \boldsymbol{\sigma}}(\boldsymbol{\rho}_{(k+1)}^{(r)}) - \bar{\boldsymbol{\varepsilon}} = -\mathbf{M}_{\text{SC}(k)}^{\star}[\boldsymbol{\rho}_{(k+1)}^{(r)} - \langle \boldsymbol{\rho}_{(k)}^{(r)} \rangle] \tag{B.2}$$

through a classical Newton–Raphson procedure. Final convergence is achieved when  $(\boldsymbol{\rho}_{(k+1)}^{(r)} - \boldsymbol{\rho}_{(k)}^{(r)})^{\text{eq}}/(\boldsymbol{\rho}_{(k+1)}^{(r)})^{\text{eq}}$  is less than a prescribed small value (typically, 0.01%).

For the derivation of second-order estimates based on the expansion of the stress potentials  $u^{(r)}$ , a similar set of equations has to be solved but with a prescribed overall stress  $\bar{\boldsymbol{\sigma}}$  and with the reference stresses  $\bar{\boldsymbol{\sigma}}^{(r)}$  substituted for the unknowns  $\boldsymbol{\rho}^{(r)}$ . The same treatment can be applied except that the set of nonlinear uncoupled equations at iteration ( $k + 1$ ) is now

$$\frac{\partial u^{(r)}}{\partial \boldsymbol{\sigma}}(\bar{\boldsymbol{\sigma}}_{(k+1)}^{(r)}) - \left\langle \frac{\partial u^{(s)}}{\partial \boldsymbol{\sigma}}(\bar{\boldsymbol{\sigma}}_{(k)}^{(s)}) \right\rangle = -\mathbf{M}_{\text{SC}(k)}^{\star}[\bar{\boldsymbol{\sigma}}_{(k+1)}^{(r)} - \bar{\boldsymbol{\sigma}}]. \tag{B.3}$$

The initial values of the reference stresses  $\boldsymbol{\rho}_{(k)}^{(r)}$  have to be carefully chosen to achieve satisfactory convergence. Calculations for fcc polycrystals were carried out with increasing values of the power-law exponent (for the first step, e.g.,  $n = 1$ , we use the linear solution and for  $n + \delta n$ , we adopt as initial unknown quantities the final values obtained for  $n$ ). For hcp polycrystals, the same procedure was used for the power-law exponents. The anisotropy parameter  $M$  (or  $w$ ) was then increased in a similar way.

For the specific microstructures reported in Section 5, this general procedure was adapted as follows:

- Integration over the whole set of crystalline orientations was reduced to integration over one spherical triangle. Consequently, averaging operations obey the rules

$$\bar{\varepsilon}_{33} = \langle \varepsilon_{33} \rangle, \quad (\text{B.4})$$

$$\bar{\varepsilon}_{11} = \bar{\varepsilon}_{22} = \left\langle \frac{\varepsilon_{11} + \varepsilon_{22}}{2} \right\rangle,$$

where  $\langle \cdot \rangle$  now denotes an integration over one spherical triangle.

- The resolution of Eq. (B.1) requires the computation of the “accommodation tensor”  $\mathbf{M}_{\text{SC}}^\star$ . For the applications considered in Section 5,  $\tilde{\mathbf{M}}_{\text{SC}}$  exhibits transverse isotropy with respect to the tensile axis. The accommodation tensor can then be evaluated by means of ordinary integrals (see Hutchinson, 1976). However, Hutchinson’s formulae do not strictly apply for a traceless overall creep compliance. It is necessary to include a very small isotropic compressibility to the overall creep compliance tensor. While Hutchinson (1976) introduced this small compressibility only for the purpose of calculating  $\mathbf{M}_{\text{SC}}^\star$ , the penalty method applied throughout this paper consists in adding this compressibility uniformly throughout the polycrystal. Numerical tests yield the same results with these two penalty methods, but the second one leads to a better convergence for ice polycrystal calculations.
- At each step ( $k$ ), the self-consistent estimate of the overall creep compliance  $\tilde{\mathbf{M}}_{\text{SC}}$  was calculated by solving the equation

$$\tilde{\mathbf{M}}_{\text{SC}} = \langle (\mathbf{M}^{(s)} + \mathbf{M}_{\text{SC}}^\star)^{-1} \rangle^{-1} - \mathbf{M}_{\text{SC}}^\star, \quad (\text{B.5})$$

which is equivalent to (B.1)<sub>(2,3,4)</sub>. The reduced integration required to solve this equation for transversely isotropic invariants. Due to the penalty method, the following five invariants of  $\tilde{\mathbf{M}}_{\text{SC}}$  were needed (Voigt notations, with 3 along the tensile axis):

$$\begin{aligned} \tilde{\mathbf{M}}_{\text{SC}1} &= \tilde{\mathbf{M}}_{\text{SC}33}, \\ \tilde{\mathbf{M}}_{\text{SC}2} &= 2\tilde{\mathbf{M}}_{\text{SC}44} + 2\tilde{\mathbf{M}}_{\text{SC}55}, \\ \tilde{\mathbf{M}}_{\text{SC}3} &= \frac{1}{2}(\tilde{\mathbf{M}}_{\text{SC}11} + \tilde{\mathbf{M}}_{\text{SC}22} - 2\tilde{\mathbf{M}}_{\text{SC}12}) + 2\tilde{\mathbf{M}}_{\text{SC}66}, \\ \tilde{\mathbf{M}}_{\text{SC}4} &= \frac{1}{2}(\tilde{\mathbf{M}}_{\text{SC}11} + \tilde{\mathbf{M}}_{\text{SC}22} + 2\tilde{\mathbf{M}}_{\text{SC}12}), \\ \tilde{\mathbf{M}}_{\text{SC}5} &= \frac{1}{\sqrt{2}}(2\tilde{\mathbf{M}}_{\text{SC}13} + 2\tilde{\mathbf{M}}_{\text{SC}23}). \end{aligned} \quad (\text{B.6})$$

The corresponding set of equations was solved with the use of a fixed-point iterative procedure.

## B.2. Computation of second-order estimates

Once the reference strains  $\bar{\varepsilon}^{(r)}$  are found, the second-order estimate for the effective viscoplastic potential  $\tilde{W}(\bar{\varepsilon})$  can be computed from (18). But, as already mentioned, for the type of local potentials under consideration in this paper, there is no explicit

relation for  $w^{(r)}$ : it is only known as the Legendre transform of the stress potentials  $u^{(r)}$ , which is not easy to compute. A more convenient relation for  $\tilde{W}(\bar{\boldsymbol{\varepsilon}})$ , is then

$$\tilde{W}(\bar{\boldsymbol{\varepsilon}}) = - \sum_r c^{(r)} u^{(r)}(\bar{\boldsymbol{\rho}}^{(r)}) + \frac{1}{2} \sum_r c^{(r)} \bar{\boldsymbol{\rho}}^{(r)} : (\bar{\boldsymbol{\varepsilon}} + \bar{\boldsymbol{\varepsilon}}^{(r)}) \quad (\text{B.7})$$

which results from the duality relation  $u^{(r)}(\bar{\boldsymbol{\rho}}^{(r)}) + w^{(r)}(\bar{\boldsymbol{\varepsilon}}^{(r)}) = \bar{\boldsymbol{\rho}}^{(r)} \cdot \bar{\boldsymbol{\varepsilon}}^{(r)}$ .

The overall stress response of a polycrystal subjected to the overall strain rate  $\bar{\boldsymbol{\varepsilon}}$  is obtained by derivation of the strain rate potential according to

$$\bar{\boldsymbol{\sigma}}_{ij} = \frac{\partial \tilde{W}}{\partial \varepsilon_{ij}}(\bar{\boldsymbol{\varepsilon}}). \quad (\text{B.8})$$

Generally, this derivation has to be performed numerically for each component of the stress. For a polycrystal displaying a transversely isotropic texture strained along its longitudinal direction, these relations reduce to (35). Results reported in Fig. 6 were computed using the following approximate expression with  $h = 0.05$ :

$$\bar{\sigma}_{33} \approx \frac{1}{10h\bar{\varepsilon}_{33}^0} \sum_{p=1}^2 p [\tilde{W}^0((1+hp)\bar{\varepsilon}_{33}^0) - \tilde{W}^0((1-hp)\bar{\varepsilon}_{33}^0)]. \quad (\text{B.9})$$

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