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Effective thermodynamic potentials and internal variables: linear viscoelastic composites

Noel Lahellec[a], Renaud Masson[b], Pierre Suquet[a,*]

[a] *Aix Marseille Univ, CNRS, Centrale Marseille, Laboratoire de Mécanique et d'Acoustique, 4 impasse Nikola Tesla, CS 40006, 13453 Marseille Cedex 13, France.*
[b] *CEA, DEN, DEC, Cadarache, 13108, Saint-Paul-Lez Durance, France.*

Abstract

New theoretical relations in linear viscoelasticity are derived by combining two different points of view. On the one hand, the general thermodynamic framework makes it possible to define the energy stored and the energy dissipated in linearly viscoelastic composites. On the other hand, the correspondence principle permits to express the macroscopic strain-stress relation as ordinary differential equations for a set of effective internal variables. A finite and small number of internal variables is rigorously sufficient in several cases of interest, including in particular particulate composites. Interpreting the macroscopic response as a rheological generalized Maxwell model allows us to compute the macroscopic free and dissipated energy of the composite. This interpretation is proved to be exact in several cases of interest. Coupled with Hashin-Shtrikman estimates, these thermodynamic functions provide additional information on the statistics of the field within each individual phase of the composite.

Keywords:
Homogenization, Linear Viscoelasticity, Thermodynamics, Correspondence principle, Internal variables, Higher-order statistics, Full field computations

1. Introduction

This study is devoted to the local and effective response of heterogeneous materials made from linearly viscoelastic individual phases where the total (infinitesimal) strain is a combination of two different contributions

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}_{\mathsf{e}} + \boldsymbol{\varepsilon}_{\mathsf{v}}, \tag{1}$$

$\boldsymbol{\varepsilon}_{\mathsf{e}}$ is the (reversible) elastic strain linearly related to the stress and $\boldsymbol{\varepsilon}_{\mathsf{v}}$ is the (dissipative) viscous strain whose rate is a function of the stress. Attention is focused here on Maxwellian viscoelasticity. However it is worth noting that most of our results, and in particular the long-memory effects captured through internal variables and their accompanying dissipation, have a more general character and that some of the conclusions of our study apply to more general viscoelastic behaviors of the constituents corresponding to Kelvin-Voigt, Burger or generalized

*Corresponding author
Email address: {suquet}@lma.cnrs-mrs.fr (Pierre Suquet)

Maxwell models which describe more accurately the behaviour of other materials, such as polymers (Suarez Afanador et al., 2022).

The composite under consideration is made of R different phases and the different contributions to the total strain in phase $r, r = 1, ..., R$ are written as

$$\boldsymbol{\varepsilon}_{\mathsf{e}} = \boldsymbol{M}_{\mathsf{e}}^{(r)} : \boldsymbol{\sigma}, \quad \dot{\boldsymbol{\varepsilon}}_{\mathsf{v}} = \boldsymbol{M}_{\mathsf{v}}^{(r)} : \boldsymbol{\sigma}. \tag{2}$$

where $\boldsymbol{M}_{\mathsf{e}}^{(r)}$ and $\boldsymbol{M}_{\mathsf{v}}^{(r)}$are respectively the elastic compliance and the viscous compliance of phase r.

The present paper is organized as follows. In section 3 the classical Correspondence Principle (CP), which consists in applying the Laplace-Carson transform to the constitutive relations, is recalled and applied to transform the mathematical problem of finding the local and effective time-dependent response of a linear viscoelastic composite into an "elasticity-like" equilibrium problem for a composite with complex moduli (Hashin, 1965; Christensen, 1969). Homogenization methods developed for elastic composites can then be applied to obtain the effective complex moduli of the composite in the Laplace domain.

When these complex moduli are available in closed form in the Laplace domain, the corresponding effective relaxation function in the time domain can be obtained by inverting the Laplace-Carson transform. As noted in Sanchez-Hubert and Sanchez-Palencia (1978) and Laws and Mc Laughlin (1978) and further explored in Francfort and Suquet (1986), Suquet (1987) (among others), even when the individual phases have a *short memory* (such as the Maxwell model (1)), the effective behavior of composites made from different such phases is no more with short memory but may exhibit a *long memory*, which manifests itself in the effective constitutive relations through an integral kernel (the relaxation or creep function). For heterogeneous materials such as polycrystals, whose effective elastic properties are well captured by one of the variants of the self-consistent model, the relaxation spectrum has indeed a non trivial continuous component (Rougier et al., 1993; Beurthey and Zaoui, 2000; Masson et al., 2012) with support in a whole interval (or several intervals) of relaxation times. In other words an infinite number of relaxation times are needed to express in full rigor the effective viscoelastic properties of the composite.

However, this number can be reduced for the class of composites we are interested in, which are mostly particulate composites, *i.e.* composites where one phase plays the role of a matrix in which the other phases are dispersed, often (but not always) in the form of ellipsoidal inclusions. Indeed a typical application motivating our study is the creep of nuclear fuels under irradiation which may be particulate composites in the case of mixed uranium-plutonium fuels (Largenton et al., 2014) or in the case of accident-tolerant fuel concepts such as those with metallic additives (Kim et al., 2015). These nuclear fuels are submitted to cycles of thermal and mechanical loadings along which the contrast in elastic, viscous or thermal properties of the phases may induce significant internal stresses along the cycles leading to a potential degradation of the material.

From a theoretical point of view it has been noted by Ricaud and Masson (2009) and Berbenni et al. (2015) that for particulate and isotropic composites whose effective elastic properties are accurately captured by the Hashin-Shtrikman relation, the number of relaxation times is *rigorously* finite. In addition, it has been noticed that each relaxation time can be asso-

ciated with an internal variable and that the effective constitutive relations can be formulated as a set of ordinary differential equations for these internal variables, generalizing the Maxwell relation (2). This is a considerable simplification of the constitutive relations compared to an integral relation between the stress history and the strain-rate history. For microstructures which cannot be handled by a Hashin-Shtrikman type of relation, and therefore for which the number of internal variables is not guaranteed to be finite, a common practice consists in approximating the relaxation spectrum by a Prony series (a series of exponentials) with a finite number of relaxation times (Schapery, 1967; Turner and Tomé, 1993; Rekik and Brenner, 2011).

For this reason, the standpoint of a finite number of internal variables is adopted in section 4 onwards.

The formulation of the effective constitutive relations with internal variables suggests that the problem of homogenization and viscoelasticity should be considered from the point of view of thermodynamics. The seminal work of Halphen and Nguyen Quoc Son (1975) and Nguyen Quoc Son (2000) has evidenced a class of constitutive relations (at the level of individual constituents) called *generalized standard materials*, which can be derived from two potentials, the Helmholtz free-energy and the dissipation potential. These notions are recalled in section 2 together with averaging relations valid for all additive quantities which are useful to define the effective free-energy and the effective dissipation which plays a key-role in understanding the full coupling of all effects.

The three-dimensional effective constitutive relations which are obtained in section 3 are that of a generalized Maxwell model. It is therefore quite natural to identify the effective free-energy and the effective dissipation in the composite as that of the one-dimensional rheological model associated with the constitutive equations. This equality, which can be proved rigorously in certain cases of interest (see Appendix A, Appendix B) is formulated in section 4. These effective potentials are not only useful to derive the effective constitutive equations, they can also be useful to get a deeper insight into the *statistics* of the local fields. The correspondence principle can only be used to compute the first moment of the fields per phase (computed in the Laplace domain first and then inverted to the time domain) but is of no help for the intra-phase fluctuations. To the authors' knowledge the only available *exact* result is an explicit expression of the second moment of the equivalent von Mises stress in a two-phase particulate matrix made from isotropic incompressible constituents (Badulescu et al., 2015). Here, using the average relations between the effective and local dissipation as well as between the effective and the local free-energy of a composite, two exact relations for the second moments of the fields in the individual phases are derived which contain and extend the previous result of Badulescu et al. (2015).

Other approaches can be followed to get information about higher-order statistics in viscoelastic composites by using time-discretized variational principles, as initially proposed in Lahellec and Suquet (2007a,b), extended in dual form in Lahellec and Suquet (2013), Agoras et al. (2016) and Cotelo et al. (2020). These approaches, although more general in scope (not based on the CP therefore not limited to linear viscoelasticity) only deliver *approximations* of the field statistics (in general). By contrast, the present study delivers *two exact* relations for the second-order moments (section 4) at the expense of restricting the class of microstructures to those leading to a finite number of relaxation times. This is in particular sufficient to access *exactly* the fluctuations of the mean stress and of the deviatoric stress in particulate composites

made of compressible phases as illustrated in section 4.

2. Thermodynamics and effective constitutive relations

In the present paper we limit ourselves to isothermal evolutions of the composite under consideration. Therefore changes in temperature are not taken into account.

2.1. Individual constituents and thermodynamic potentials

The main ingredients needed in constitutive relations are threefold[1]

a) A finite number of *internal variables* $\boldsymbol{\xi}$ which, in addition to the observable strain $\boldsymbol{\varepsilon}$, describe the internal state of the material. The internal variables usually monitor the changes taking place in the material (plastic strains, damage, void growth etc...).

b) A *Helmholtz free-energy function* $w(\boldsymbol{\varepsilon}, \boldsymbol{\xi})$ which gives, for each possible state $(\boldsymbol{\varepsilon}, \boldsymbol{\xi})$ of the material the energy available to trigger its evolution. The thermodynamic forces associated with the state variables are :

$$\boldsymbol{\sigma}^{\mathsf{rev}} = \frac{\partial w}{\partial \boldsymbol{\varepsilon}}(\boldsymbol{\varepsilon}, \boldsymbol{\xi}), \quad \boldsymbol{\mathcal{A}} = -\frac{\partial w}{\partial \boldsymbol{\xi}}(\boldsymbol{\varepsilon}, \boldsymbol{\xi}). \tag{3}$$

c) *Complementary laws* describing the change in the state variables under the effect of thermodynamic forces

$$(\dot{\boldsymbol{\varepsilon}}, \dot{\boldsymbol{\xi}}) = \mathcal{F}(\boldsymbol{\sigma}^{\mathsf{irr}}, \boldsymbol{\mathcal{A}}), \text{ where } \boldsymbol{\sigma}^{\mathsf{irr}} = \boldsymbol{\sigma} - \boldsymbol{\sigma}^{\mathsf{rev}}. \tag{4}$$

The energy dissipated in a thermodynamic evolution of the material, called the intrinsic dissipation (in the wording of Germain et al., 1983), also called internal dissipation in Rosakis et al. (2000), can be expressed along an isothermal process in terms of the evolution of the internal variables $\boldsymbol{\xi}$ as (Germain et al., 1983),

$$d = \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}} - \dot{w} = \boldsymbol{\sigma}^{\mathsf{irr}} : \dot{\boldsymbol{\varepsilon}} + \boldsymbol{\mathcal{A}} : \dot{\boldsymbol{\xi}}. \tag{5}$$

In other words the (intrinsic) dissipation is the work of the forces $(\boldsymbol{\sigma}^{\mathsf{irr}}, \boldsymbol{\mathcal{A}})$ in the change of the state variables $\boldsymbol{\varepsilon}, \boldsymbol{\xi}$. The second principle of Thermodynamics requires that this dissipation remains positive along all possible evolutions of the material and this requirement imposes limitations on the form of the function $\mathcal{F}$.

Remark 1. Let us note that the dissipation d is a scalar which is the rate of energy dissipated in the *actual* evolution of the system which may depend on all the quantities involved in the evolution of the system, $\boldsymbol{\sigma}^{\mathsf{irr}}$, $\boldsymbol{\mathcal{A}}$, $\dot{\boldsymbol{\varepsilon}}$, $\dot{\boldsymbol{\xi}}$ through relation (5). Using the complementary laws (4), the dissipation can be expressed as a function of the strain-rate and of the rate of internal variables and this function is sometimes called the dissipation *function*, Fischer et al. (2014).

[1] Attention is limited to infinitesimal strains, so that the strain is the symmetric part of the gradient of the displacement field and changes in mass density can be neglected.

Throughout this study it will be assumed that $\boldsymbol{\sigma} = \boldsymbol{\sigma}^{\mathsf{rev}}$, so that d does not depend on $\dot{\boldsymbol{\varepsilon}}$ but only on $\dot{\boldsymbol{\xi}}$ (hence we favor Maxwell materials over Kelvin-Voigt materials), although most of the developments here can be extended, at least qualitatively, to the more general case. It is readily checked that the viscoelastic model (1), (2) fits into the above framework of generalized standard material. For this purpose let us define $\boldsymbol{\xi} = \boldsymbol{\varepsilon}_{\mathsf{v}}$, the free-energy function w and the complementary laws as,

$$w(\boldsymbol{\varepsilon}, \boldsymbol{\xi}) = \frac{1}{2}(\boldsymbol{\varepsilon} - \boldsymbol{\xi}) : \boldsymbol{L}_{\mathsf{e}} : (\boldsymbol{\varepsilon} - \boldsymbol{\xi}), \quad \dot{\boldsymbol{\xi}} = \boldsymbol{M}_{\mathsf{v}} : \boldsymbol{\mathcal{A}} \tag{6}$$

where $\boldsymbol{L}_{\mathsf{e}} = \boldsymbol{M}_{\mathsf{e}}^{-1}$. The thermodynamic forces associated with $\boldsymbol{\varepsilon}$ and $\boldsymbol{\xi} = \boldsymbol{\varepsilon}_{\mathsf{v}}$ through (3) are respectively

$$\boldsymbol{\sigma} = \boldsymbol{L}_{\mathsf{e}} : (\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}_{\mathsf{v}}), \quad \boldsymbol{\mathcal{A}} = \boldsymbol{\sigma}.$$

The actual dissipation along a deformation path reads as

$$d = \boldsymbol{\mathcal{A}} : \dot{\boldsymbol{\xi}} = \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}}_{\mathsf{v}} = \boldsymbol{\sigma} : \boldsymbol{M}_{\mathsf{v}} : \boldsymbol{\sigma} = \dot{\boldsymbol{\varepsilon}}_{\mathsf{v}} : \boldsymbol{L}_{\mathsf{v}} : \dot{\boldsymbol{\varepsilon}}_{\mathsf{v}},$$

with $\boldsymbol{L}_{\mathsf{v}} = \boldsymbol{M}_{\mathsf{v}}^{-1}$.

2.2. Heterogeneous materials and averaging

The first step in an homogenization process, *i.e.* in the derivation of the effective (or macroscopic) behavior of small-scale heterogeneous media, is to define a representative volume element (r.v.e.), which contains the relevant statistical information on the microstructure. The definition of such a volume is unambiguous in the case of a periodic medium (a unit cell is sufficient to generate the whole microstructure and is therefore representative), but is more delicate in the case of a random medium. Without loss of generality attention is restricted to periodic composites[2]. The periodicity of the microstructure allows one to consider the homogenization as an asymptotic problem where a small parameter (usually considered to be the size of the unit-cell reproduced by periodicity to generate the whole body) goes to 0 and to derive limits of all the above quantities (stress, strain, temperature, free-energy, dissipation, entropy etc...).

The unit-cell, or representative volume element, V is comprised of R phases occupying domains $V^{(r)}$ with characteristic functions $\chi^{(r)}$ and volume fraction $c^{(r)}$. Averaging over $V^{(r)}$ and V are respectively denoted as $\langle . \rangle^{(r)}$ and $\langle . \rangle$ with

$$\langle . \rangle = \sum_{r=1}^{R} c^{(r)} \langle . \rangle^{(r)} .$$

Then, in order to define the relations between microscopic (at small scale) and macroscopic (at a larger scale) quantities it is essential to note that additive quantities are averaged in the change

[2]The justification is exactly the same as in Gibiansky and Milton (1993) who write "because a composite which is not periodic, but say statistically homogeneous, can be replaced by a periodic one with negligible change in its effective properties: one can take a sufficiently large cubic representative sample of the statistically homogeneous composite and extend it periodically."

of scales in the following sense (Francfort et al., 1983). As a prototypical example consider the mass density ρ. The total mass of the volume V is $m = \int_V \rho(\boldsymbol{x}) d\boldsymbol{x}$. If a homogeneous material, with a homogeneous density $\widetilde{\rho}$ is substituted to the heterogeneous material in V, it should have mass m as well and therefore its density must be

$$\widetilde{\rho} = \frac{m}{|V|} = \frac{1}{|V|} \int_V \rho(\boldsymbol{x}) d\boldsymbol{x} = \langle \rho \rangle \, .$$

In other words $\widetilde{\rho}$ is the average of the local density field $\rho(\boldsymbol{x})$.

The same reasoning applies to *all* additive quantities, *i.e.* quantities which, for a material system, are equal to the sum over all material points of that quantity at each material point (an integral for a continuum). Clearly the temperature of a material system for instance is not additive, whereas its total internal energy $\mathcal{E}$, its total Helmholtz free-energy $\mathcal{W}$, its dissipation $\mathcal{D}$ can be written as an integral over the volume of a density and are additive quantities

$$\mathcal{E} = \int_V e(\boldsymbol{x}) \, \mathrm{d}\boldsymbol{x}, \quad \mathcal{W} = \int_V w(\boldsymbol{x}) \, \mathrm{d}\boldsymbol{x}, \quad \mathcal{D} = \int_V d(\boldsymbol{x}) \, \mathrm{d}\boldsymbol{x}.$$

Following the same reasoning as for the mass density, we find that, in the change of scales, the effective value of an additive quantity is the average of that quantity over the r.v.e.,

$$\widetilde{e} = \langle e \rangle \, , \quad \widetilde{w} = \langle w \rangle \, , \quad \widetilde{d} = \langle d \rangle \, . \tag{7}$$

As is well-known, the averaging rule applies as well to the stress and the strain tensors, although they are not additive quantities. Indeed, when the stress field is statically admissible, when the strain is kinematically admissible and when appropriate boundary conditions are applied to the boundary of the r.v.e. V[3], the overall stress and strain read as

$$\overline{\boldsymbol{\sigma}} = \langle \boldsymbol{\sigma} \rangle \, , \quad \overline{\boldsymbol{\varepsilon}} = \langle \boldsymbol{\varepsilon} \rangle \, .$$

In addition, the average of the power of internal forces at small scale is equal to the power of internal forces at the larger scale (so-called Hill-Mandel relation)

$$\langle \boldsymbol{\sigma}' : \boldsymbol{\varepsilon}' \rangle = \langle \boldsymbol{\sigma}' \rangle : \langle \boldsymbol{\varepsilon}' \rangle \tag{8}$$

for any periodic divergence-free stress field $\boldsymbol{\sigma}'$ at the microscopic scale and any periodic kinematically compatible strain field $\boldsymbol{\varepsilon}'$.

Averaging the expression (5) of the dissipation and taking into account (7) and (8), yields

$$\widetilde{d} + \dot{\widetilde{w}} = \overline{\boldsymbol{\sigma}} : \dot{\overline{\boldsymbol{\varepsilon}}}, \tag{9}$$

which can be interpreted as the split of the power supplied to the r.v.e., $\overline{\boldsymbol{\sigma}} : \dot{\overline{\boldsymbol{\varepsilon}}}$, into a part which is stored, $\dot{\widetilde{w}}$, and a part which is dissipated, $\widetilde{d}$.

[3]Here we have chosen periodicity conditions but other well-known admissible boundary conditions are affine displacements, or uniform tractions applied on the boundary of V (Hashin, 1983; Suquet, 1987).

3. Effective stress-strain relations in linear viscoelasticity

It is a well-known fact that the effective behavior of heterogeneous viscoelastic materials can exhibit *long-range memory* effects even when the individual (linear viscoelastic) constituents have short memory (Sanchez-Hubert and Sanchez-Palencia, 1978; Suquet, 1987; Francfort and Suquet, 1986).

The effective properties of the composite are analyzed by examining the response to external loadings of a representative volume element V of the composite containing the relevant statistical information on the microstructure. V is subjected to a prescribed loading consisting of a time-dependent average strain history $\overline{\boldsymbol{\varepsilon}}(t) = \langle \boldsymbol{\varepsilon}(t) \rangle$ over a time interval $[0, T]$. For simplicity, it is assumed throughout the paper that the initial conditions for the local stress and strain fields (and therefore for their averages) are identically zero. The local stress and strain fields are determined by solving the boundary value problem consisting of the constitutive relations (1)-(2), equilibrium, compatibility conditions and periodic boundary conditions. The effective constitutive relations are the relations between the overall stress at time t, $\overline{\boldsymbol{\sigma}}(t) = \langle \boldsymbol{\sigma}(t) \rangle$, and the history of overall strain $\overline{\boldsymbol{\varepsilon}}(s),\ 0 \leq s \leq t$, or vice-versa.

3.1. Long memory effects arising from homogenization

3.1.1. Creep function and retardation spectrum

The so-called "correspondence principle" transforms a linear viscoelastic problem (without ageing) into an "elasticity-like" problem by means of the Laplace-Carson (LC) transform and the Stieljes convolution defined as

$$f^*(p) = p \int_0^{+\infty} e^{-pt} f(t)\, \mathrm{d}t,\ f \star g(t) = \frac{\mathrm{d}}{\mathrm{d}t}(f * g)(t) = \int_0^t f(t-s)\, \dot{g}(s)\, \mathrm{d}s,\ (f \star g)^*(p) = f^*(p) g^*(p).$$

The constitutive relations (1) (2) in phase r can be re-written as

$$\boldsymbol{\varepsilon} = \boldsymbol{M}^{(r)} \star \boldsymbol{\sigma}, \quad \text{with} \quad \boldsymbol{M}^{(r)}(t) = \boldsymbol{M}_{\mathsf{e}}^{(r)} + t\, \boldsymbol{M}_{\mathsf{v}}^{(r)}. \tag{10}$$

Upon application of the LC transform, the constitutive equations of the phases in Laplace space read as

$$\boldsymbol{\varepsilon}^*(p) = \boldsymbol{M}^{(r)*}(p) : \boldsymbol{\sigma}^*(p), \quad \text{with} \quad \boldsymbol{M}^{(r)*}(p) = \boldsymbol{M}_{\mathsf{e}}^{(r)} + \frac{1}{p} \boldsymbol{M}_{\mathsf{v}}^{(r)}. \tag{11}$$

Note that

$$\boldsymbol{M}^{(r)*}(p) \sim \boldsymbol{M}_{\mathsf{e}}^{(r)} \text{ when } p \to +\infty, \quad \boldsymbol{M}^{(r)*}(p) \sim \frac{1}{p} \boldsymbol{M}_{\mathsf{v}}^{(r)} \text{ when } p \to 0. \tag{12}$$

Then, upon homogenization, the effective constitutive relations read as

$$\overline{\boldsymbol{\varepsilon}}^*(p) = \widetilde{\boldsymbol{M}}^*(p) : \overline{\boldsymbol{\sigma}}^*(p), \tag{13}$$

where, unfortunately, $\widetilde{\boldsymbol{M}}^*(p)$ is different from $\widetilde{\boldsymbol{M}}_{\mathsf{e}} + \frac{1}{p}\widetilde{\boldsymbol{M}}_{\mathsf{v}}$ (homogenization is not an additive process) but, taking into account (12), can be written formally as

$$\widetilde{\boldsymbol{M}}^*(p) = \widetilde{\boldsymbol{M}}_{\mathsf{e}} + \frac{1}{p}\widetilde{\boldsymbol{M}}_{\mathsf{v}} + \int_0^{+\infty} \widehat{\boldsymbol{M}}(\tau)\, \frac{1}{p + \frac{1}{\tau}}\, \mathrm{d}\tau. \tag{14}$$

The interpretation of $\widehat{\boldsymbol{M}}(\tau)$ is easier in time space where, after inverse Laplace transform, the constitutive relation reads as

$$\overline{\boldsymbol{\varepsilon}}(t) = \widetilde{\boldsymbol{M}} \star \overline{\boldsymbol{\sigma}}(t), \tag{15}$$

where $\widetilde{\boldsymbol{M}}(t)$ is the overall *creep function* of the composite (response of the composite to a creep test),

$$\widetilde{\boldsymbol{M}}(t) = \widetilde{\boldsymbol{M}}_{\mathsf{e}} + t\, \widetilde{\boldsymbol{M}}_{\mathsf{v}} + \int_0^{+\infty} \widehat{\boldsymbol{M}}(\tau)\, (1 - \mathrm{e}^{-t/\tau})\, \mathrm{d}\tau. \tag{16}$$

$\widetilde{\boldsymbol{M}}_{\mathsf{e}} + t\, \widetilde{\boldsymbol{M}}_{\mathsf{v}}$ is the Maxwellian component of $\widetilde{\boldsymbol{M}}(t)$ (corresponding to "short memory" effects as in (10)) and $\widehat{\boldsymbol{M}}(\tau)$ is the *retardation spectrum* associated with "long memory" effects.

The effective constitutive relation can be alternatively expressed with a continuous distribution of internal variables by noting that (13) (14) in Laplace space can be alternatively written as

$$\overline{\boldsymbol{\varepsilon}}^*(p) = \widetilde{\boldsymbol{M}}_{\mathsf{e}} : \overline{\boldsymbol{\sigma}}^*(p) + \frac{1}{p}\widetilde{\boldsymbol{M}}_{\mathsf{v}} : \overline{\boldsymbol{\sigma}}^*(p) + \int_0^{+\infty} \boldsymbol{\alpha}_\tau^*(p)\, \mathrm{d}\tau, \quad \text{with} \quad \boldsymbol{\alpha}_\tau^*(p) = \frac{\widehat{\boldsymbol{M}}(\tau) : \overline{\boldsymbol{\sigma}}^*(p)}{p + 1/\tau},$$

or, in time domain,

$$\dot{\overline{\boldsymbol{\varepsilon}}}(t) = \widetilde{\boldsymbol{M}}_{\mathsf{e}} : \dot{\overline{\boldsymbol{\sigma}}}(t) + \widetilde{\boldsymbol{M}}_{\mathsf{v}} : \overline{\boldsymbol{\sigma}}(t) + \int_0^{+\infty} \dot{\boldsymbol{\alpha}}_\tau(t)\, \mathrm{d}\tau, \text{ with } \quad \dot{\boldsymbol{\alpha}}_\tau(t) + \frac{1}{\tau}\boldsymbol{\alpha}_\tau(t) = \widehat{\boldsymbol{M}}(\tau) : \overline{\boldsymbol{\sigma}}(t). \tag{17}$$

The definition (17) of the effective constitutive relations through a continuous distribution of internal variables $\boldsymbol{\alpha}_\tau$ (where τ is a continuous parameter) is of little help in practice and it is indeed easier to work with the integral formulation (16). However, as will be seen in section 3.2, the situation is totally different when the retardation spectrum consists of a finite number of Dirac masses.

3.1.2. Relaxation function and relaxation spectrum

In the above version of the effective constitutive relations, the strain at time t is expressed in terms of the stress history. However it is often convenient to invert these relations to express the stress at time t in terms of the strain history. This is done by inverting the relations (11), (13) and (15),

$$\overline{\boldsymbol{\sigma}}^*(p) = \widetilde{\boldsymbol{L}}^*(p) : \overline{\boldsymbol{\varepsilon}}^*(p), \quad \overline{\boldsymbol{\sigma}}(t) = \widetilde{\boldsymbol{L}} \star \overline{\boldsymbol{\varepsilon}}(t) = \int_0^t \widetilde{\boldsymbol{L}}(t-s) : \dot{\overline{\boldsymbol{\varepsilon}}}(s)\, \mathrm{d}s. \tag{18}$$

where, again, $\widetilde{\boldsymbol{L}}^*(p) \neq \left(\widetilde{\boldsymbol{M}}_{\mathsf{e}} + \frac{1}{p}\widetilde{\boldsymbol{M}}_{\mathsf{v}}\right)^{-1}$. The inverse LC transform of $\widetilde{\boldsymbol{L}}^*$ is the effective relaxation modulus $\widetilde{\boldsymbol{L}}(t)$ of the composite which can be expressed in terms of the relaxation

spectrum of the composite $\overline{\boldsymbol{L}}(\tau)$ defined as

$$\widetilde{\boldsymbol{L}}(t) = \int_0^{+\infty} \overline{\boldsymbol{L}}(\tau) e^{-\frac{t}{\tau}} \, \mathrm{d}\tau, \quad \widetilde{\boldsymbol{L}}^*(p) = \int_0^{+\infty} \overline{\boldsymbol{L}}(\tau) \frac{p}{p+1/\tau} \, \mathrm{d}\tau. \tag{19}$$

The constitutive relation (18) can be formulated using a continuous distribution of internal variables. For this purpose, (19) is written as

$$\overline{\boldsymbol{\sigma}}^*(p) = \int_0^{+\infty} \frac{p}{p+1/\tau} \overline{\boldsymbol{L}}(\tau) : \overline{\boldsymbol{\varepsilon}}^*(p) \, \mathrm{d}\tau = \int_0^{+\infty} \overline{\boldsymbol{L}}(\tau) : (\overline{\boldsymbol{\varepsilon}}^*(p) - \boldsymbol{\beta}_\tau^*(p)) \, \mathrm{d}\tau,$$

with

$$\text{with} \quad \boldsymbol{\beta}_\tau^*(p) = \frac{1}{\tau p + 1} \overline{\boldsymbol{\varepsilon}}^*(p),$$

or in time domain

$$\overline{\boldsymbol{\sigma}}(t) = \int_0^{+\infty} \overline{\boldsymbol{L}}(\tau) : (\overline{\boldsymbol{\varepsilon}}(t) - \boldsymbol{\beta}_\tau(t)) \, \mathrm{d}\tau, \quad \text{with} \quad \tau \dot{\boldsymbol{\beta}}_\tau(t) + \boldsymbol{\beta}_\tau(t) = \overline{\boldsymbol{\varepsilon}}(t), \quad \boldsymbol{\beta}_\tau(0) = 0. \tag{20}$$

Again, the benefit of a formulation based on internal variables will appear in the next section.

3.2. Prony series and finite number of internal variables

Let us assume for the moment that the relaxation spectrum consists of a finite sum of Dirac masses at discrete times $\overline{\tau}_i$ (the relaxation times),

$$\overline{\boldsymbol{L}}(\tau) = \sum_{i=1}^{N} \overline{\boldsymbol{L}}_i \delta_{\overline{\tau}_i}(\tau), \ \overline{\boldsymbol{L}}_i \geq 0, \ (\overline{L}_i)_{pqrs} = (\overline{L}_i)_{rspq} = (\overline{L}_i)_{qprs}, \ \forall p, q, r, s, \tag{21}$$

where the inequality $\overline{\boldsymbol{L}}_i \geq 0$ is to be understood in the sense of quadratic forms and the last relations meaning that $\overline{\boldsymbol{L}}_i$ has major and minor symmetries. Equivalently

$$\widetilde{\boldsymbol{L}}(t) = \sum_{i=1}^{N} \overline{\boldsymbol{L}}_i e^{-\frac{t}{\overline{\tau}_i}}, \quad \widetilde{\boldsymbol{L}}^*(p) = \sum_{i=1}^{N} \overline{\boldsymbol{L}}_i \frac{p}{p+1/\overline{\tau}_i}. \tag{22}$$

Then the constitutive relations (20) reduce to,

$$\overline{\boldsymbol{\sigma}}(t) = \sum_{i=1}^{N} \overline{\boldsymbol{\sigma}}_i, \quad \overline{\boldsymbol{\sigma}}_i = \overline{\boldsymbol{L}}_i : (\overline{\boldsymbol{\varepsilon}}(t) - \boldsymbol{\beta}_i(t)), \quad \text{with} \quad \overline{\tau}_i \dot{\boldsymbol{\beta}}_i(t) + \boldsymbol{\beta}_i(t) = \overline{\boldsymbol{\varepsilon}}(t), \ \boldsymbol{\beta}_i(0) = \mathbf{0}. \tag{23}$$

This last relation shows the benefit of internal variables as it is much easier to store the value at time t of N internal variables $\boldsymbol{\beta}_i(t)$, obtained by integration of N decoupled first-order differential equations, rather than to store the whole history $\overline{\boldsymbol{\varepsilon}}(s)|_{0 \leq s \leq t}$ of the macroscopic strain from 0 to t.

The dual formulation (17) of the constitutive relations simplifies as well. When the relaxation spectrum is discrete (finite sum of Dirac masses), the retardation spectrum is a sum of a

finite number of Dirac masses at times $\widehat{\tau}_k$ (the retardation times)

$$\widehat{\boldsymbol{M}}(\tau) = \sum_{k=1}^{M} \widehat{\boldsymbol{M}}_i \delta_{\widehat{\tau}_k}(\tau). \tag{24}$$

Note that the number M of retardation times is in general less than the number N of relaxation times, since additional retardation times are already present in the first two terms of (17). How the retardation times $\hat{\tau}_k$ and the corresponding weights $\hat{\boldsymbol{M}}_k$ are related to the relaxation times $\overline{\tau}_i$ and weights $\overline{\boldsymbol{L}}_i$ is discussed (in simple cases) in Appendix C.

3.3. Dependence of the number of internal variables on the microstructure

There is in general an infinite number of internal variables involved in the formulations (17) or (20) of the effective relations. However in several cases of interest this number can be reduced to a finite number.

It is readily seen that when the complex moduli $\boldsymbol{L}^{(r)*}(p)$ are rational fractions in p, and when the effective moduli $\widetilde{\boldsymbol{L}}$ are rational fractions of the moduli of the phases, then $\widetilde{\boldsymbol{L}}^*(p)$ are rational fractions in p which can be decomposed into partial fractions. In that case the decomposition (22) is an exact result.

When this decomposition is not an exact result, it is often a common approximation which can be made accurate by an optimization procedure.

The number of internal variables depends on the singularities of the function $p \to \widetilde{\boldsymbol{L}}^*(p)$ in the complex plane. As can be inferred from (21), the LC-transform of the relaxation function has poles when $p = -1/\overline{\tau}_i$ and reducing the model to a finite number of internal variables is equivalent to $\widetilde{\boldsymbol{L}}^*(p)$ having a finite number of isolated poles.

As is well known, the effective complex moduli are the average of the moduli of the phases, weighted by the strain localization tensor $\boldsymbol{A}^{(r)*}(p)$,

$$\widetilde{\boldsymbol{L}}^*(p) = \sum_{r=1}^{R} c^{(r)} \boldsymbol{L}^{(r)*}(p) : \boldsymbol{A}^{(r)*}(p).$$

This relation shows in particular that the poles of $\widetilde{\boldsymbol{L}}^*(p)$ can be caused either by the phases (poles of any of the $\boldsymbol{L}^{(r)*}(p)$) or by the microstructure (through $\boldsymbol{A}^{(r)*}(p)$).

A simple example sheds light on these two sources of singularities. Consider a two-phase composite composed of incompressible Maxwell phases. Then the complex moduli of the phases and their contrast read as

$$\mu^{(r)*} = \mu_{\mathsf{e}}^{(r)} \frac{p}{p + \frac{1}{\tau^{(r)}}}, \quad z = \frac{\mu^{(2)*}}{\mu^{(1)*}} = \frac{\mu_{\mathsf{e}}^{(2)}}{\mu_{\mathsf{e}}^{(1)}} \frac{p + \frac{1}{\tau^{(1)}}}{p + \frac{1}{\tau^{(2)}}}, \tag{25}$$

where $\mu_{\mathsf{e}}^{(r)}$ is the elastic shear modulus of phase r and $\tau^{(r)}$ is its time relaxation (its viscous shear modulus is $\mu_{\mathsf{v}}^{(r)} = \mu_{\mathsf{e}}^{(r)} \tau^{(r)}$). Assuming that the domains occupied by the phases are cylindrical parallel to $\boldsymbol{e}_3$, the anti-plane effective shear modulus of the composite (or equivalently its in-

plane conductivity) can be written as a function of the contrast between the two phases as

$$\widetilde{\mu}_L^*(\mu^{(1)*}(p), \mu^{(2)*}(p)) = \mu^{(1)*}(p)\widetilde{\mu}_L^*(1, z). \tag{26}$$

The singularities of $\widetilde{\mu}_L^*$ not already contained in that of the phases are the singularities of $\widetilde{\mu}_L^*(1, z)$ as a function of z. These singularities now depend only on the microstructure. Among the many possible arrangements of the phases, three classes of microstructures are typical of the relaxation spectra that can be expected.

The first class of microstructure corresponds to a classical fiber-matrix configuration with well-separated circular fibers isotropically distributed in the plane. As is classical, the effective elastic properties of such composites are accurately described by the Hashin-Shtrikman relation (phase 1 playing the role of the matrix),

$$\widetilde{\mu}_L^*(z) = \mu^{(1)*}\left(1 + c^{(2)}\frac{z-1}{1 + c^{(1)}\beta(z-1)}\right), \quad \beta = \frac{2}{2+d}, \tag{27}$$

where d is dimension of space ($d = 2$ for the two-dimensional problem considered here). The effective modulus $\widetilde{\mu}_L^*(z)$ becomes singular when

$$1 + c^{(1)}\beta(z-1) = 0, \quad \textit{i.e.} \text{ when} \quad z = z_c \stackrel{\text{def}}{=} -\frac{1-\beta c^{(1)}}{\beta c^{(1)}}.$$

The critical contrast z_c is a function of the microstructure only (depending on β and $c^{(1)}$ only). Through the relation between z and p (which now depends on the constitutive relations of the phases), it is found that the critical contrast is reached when p is equal to a critical value p_c defined as

$$p_c \stackrel{\text{def}}{=} -\frac{1}{\overline{\tau}}, \quad \text{where} \quad \overline{\tau} = \tau^{(1)}\tau^{(2)}\frac{\mu_{\mathsf{e}}^{(1)}(1-\beta c^{(1)}) + \mu_{\mathsf{e}}^{(2)}\beta c^{(1)}}{\tau^{(1)}\mu_{\mathsf{e}}^{(1)}(1-\beta c^{(1)}) + \tau^{(2)}\mu_{\mathsf{e}}^{(2)}\beta c^{(1)}}. \tag{28}$$

Coming back to (26) it is seen that the complex modulus $\widetilde{\mu}_L^*(p)$ has two isolated poles, the first one being the pole of $\mu^{(1)*}$ and the second one being p_c associated with the class of microstructure considered here. Therefore the composite has two distinct relaxation times (deduced from the poles by means of the relation $\tau = -1/p$), one of them being the relaxation time $\tau^{(1)}$ of the matrix and the second one, $\overline{\tau}$, reflecting the microstructure.

A second example of instructive microstructure is a polycrystalline microstructure with randomly oriented grains. In dimension $d = 2$, the microstructural function $\widetilde{\mu}_L^*(z)$ reads as

$$\widetilde{\mu}_L^*(z) = \sqrt{z}, \tag{29}$$

which is exact for the checkerboard. The microstructure introduces a singularity for all z along the real negative axis. According to (25) which can be inverted as,

$$p = -\frac{1}{\tau^{(1)}\tau^{(2)}}\frac{\tau^{(1)}z - \tau^{(2)}\mu_{\mathsf{e}}^{(2)}/\mu_{\mathsf{e}}^{(1)}}{z - \mu_{\mathsf{e}}^{(2)}/\mu_{\mathsf{e}}^{(1)}}. \tag{30}$$

The cut along the negative real axis in the z-plane converts into a cut $]-1/\tau^{(1)}, -1/\tau^{(2)}[$ in

the p-plane and all p's in this interval are singular points for $\widetilde{\mu}_L^*(p)$. Therefore the relaxation spectrum $\overline{\boldsymbol{L}}$ is a continuous spectrum with support in the interval $]\tau^{(1)}, \tau^{(2)}[$. This spectrum is given in closed form in Masson et al. (2012). In dimension $d = 3$, Beurthey and Zaoui (2000) also obtained a similar continuous spectrum, to which two Dirac masses corresponding to the two individual phases must be added.

A third interesting example of microstructure is a distribution of square inclusions arranged periodically along a square lattice. When the volume fraction of the inclusion is $c^{(2)} = 0.25$ the microstructural function $\widetilde{\mu}_L^*(z)$ reads as (Obnosov, 1999)

$$\widetilde{\mu}_L^*(z) = \sqrt{\frac{1+3z}{3+z}}. \tag{31}$$

Hence the microstructure introduces a singularity for all z in the interval $]-3, -1/3[$. According to (30), $\widetilde{\mu}_L^*(p)$ is singular in the p-plane along a cut $]\check{p}_1, \check{p}_2[$ which is now *strictly* contained in the interval $]-1/\tau^{(1)}, -1/\tau^{(2)}[$. Therefore the effective modulus has two types of singularities, an isolated pole at $p = -1/\tau^{(1)}$ (singularity due to the matrix), and a cut in the interval $]\check{p}_1, \check{p}_2[$ due to the microstructure. Consequently the relaxation spectrum $\overline{\boldsymbol{L}}$ has two contributions, one being a Dirac mass at the relaxation time $\tau^{(1)}$ and the second being a continuous function with support in $]-1/\check{p}_2, -1/\check{p}_1[$.

In the above examples, the (microstructural) singularities of $\widetilde{\boldsymbol{L}}^*(z)$, as a function of the contrast z, have been investigated through the explicit forms (27), (29) or (31) of the effective modulus in the complex plane and we have observed that the singularities can take the form of isolated poles or cuts in the complex p-plane. Two natural questions arise as to know whether these are the only types of singularities that one can expect and whether one could infer the type of singularity that can be expected for a particular microstructure by a direct inspection of the features of microstructure, without any explicit relation for the effective moduli. The answer to the first question is that all singularities are located on the negative real axis (see Appendix C). They can be either isolated poles, or cuts or accumulation points of isolated poles (not present in the above examples) or combination of these three possibilities (Milton, 2002). The cuts are not necessarily connected and can consist of several intervals (examples of such situations are given for instance in Beurthey and Zaoui, 2000).

A complete answer to the second question, relation between the microstructure and the type of singularity, is not available in elasticity to the best of the authors' knowledge. However, according to Milton (2002), the broad picture is well understood in conductivity and can be schematically summarized as follows.

- For particulate composites, when the inclusions are well separated, the singularities[4] are contained in a finite interval $[-A, -B]$ where A and B are strictly positive scalars depending on the distance of separation between the inclusions. This result has been extended to plane elasticity by Bruno and Leo (1993).

- According to Milton (2002), Bergman has shown that when the microstructure is periodic

[4]The singularities discussed here are that due to the microstructure, or in the above notations the singularities with respect to the variable z.

and the interface between the phases is a smooth surface with radii of curvature bounded from below, the singularities are all isolated poles, except for a possible accumulation point.

- When the interface between the phases has sharp corners, Milton (2002) (following Hetherington and Thorpe) suggests that the singular set contains cuts and that it is the angles at the corners that determine the position of the branch cuts. This is consistent with the observation made for the checkerboard and the Obnosov microstructure. These angles also determine the singularities of the fields near the sharp corners, hence there is a relation between the cuts and the singularity of the local fields.

We shall not comment further the relations between the nature of the singularities and the microstructure, since we are mainly interested here in situations where there is a finite number of isolated poles. As recalled above, there are many microstructures of interest for which this result is *exact*. For other microstructures, it is a common practice (Schapery, 1967), referred to as the collocation method, to find an *approximation* of the actual relaxation spectrum in the form (21) of a Prony series. For a prescribed number of relaxation times $\overline{\tau}_i$, their location and the corresponding weights $\overline{\boldsymbol{L}}_i$ can be found by means of an optimization procedure to minimize the distance between the exact complex moduli and their approximation through Prony series (Rekik and Brenner, 2011).

3.4. Typical situations where the number of internal variables is finite.

The number of poles of $\widetilde{\boldsymbol{L}}^*(p)$ is finite as soon as it can be expressed as a rational function of p. This is typically the case in two-phase particulate composites[5] where the effective moduli read as

$$\widetilde{\boldsymbol{L}}^*(p) = \boldsymbol{L}^{(1)*}(p) + c^{(2)} \left(\boldsymbol{L}^{(2)*}(p) - \boldsymbol{L}^{(1)*}(p) \right) : \boldsymbol{A}^{(2)*}(p).$$

$\widetilde{\boldsymbol{L}}^*(p)$ possibly becomes singular when either $\boldsymbol{L}^{(1)*}(p)$, or $\boldsymbol{L}^{(2)*}(p)$, or $\boldsymbol{A}^{(2)*}(p)$ becomes singular (note that by the average condition $\boldsymbol{A}^{(1)*}(p)$ and $\boldsymbol{A}^{(2)*}(p)$ are singular simultaneously). The effective linear elastic properties of particulate composites are accurately estimated by Hashin-Shtrikman estimates, initially derived for isotropic phases with an isotropic distribution and generalized by Willis (1977) and Ponte Castañeda and Willis (1995) to anisotropic phases with ellipsoidal symmetry and ellipsoidal distribution. In such composites the strain localization tensor can be expressed as

$$\boldsymbol{A}^{(2)*}(p) = \left[\boldsymbol{I} + c^{(1)} \boldsymbol{P}^{(1)*}(p) : \left(\boldsymbol{L}^{(2)*}(p) - \boldsymbol{L}^{(1)*}(p) \right) \right]^{-1},$$

where the polarization tensor $\boldsymbol{P}^{(1)*}$ depends on the shape of the inclusions, on their arrangement and the properties of phase (1). When the shape and the distribution of the particles have the

[5] A particulate composite is a composite where one of the phases is connected and plays the role of the matrix, the other phases being dispersed in the matrix. The particles are ellipsoidal but it is not required that they have a more specific shape or a specific orientation and the composite may be isotropic or anisotropic.

same ellipsoidal symmetry, this tensor reads as

$$\boldsymbol{P}^{(1)*} = \frac{1}{4\pi\,|\boldsymbol{Z}|}\int_{|\boldsymbol{n}|=1} \boldsymbol{\Gamma}^{(1)*}(\boldsymbol{n})\left|\boldsymbol{Z}^{-1}\boldsymbol{n}\right|^{-3}\,dS(\boldsymbol{\xi}), \tag{32}$$

where $\boldsymbol{\Gamma}^{(1)*}$ is a fourth-order tensor with components $\Gamma^{(1)*}_{ijkh}(\boldsymbol{n}) = N^{(1)*}_{ki}(\boldsymbol{n})n_j n_h|_{(ij),(kh)}$, with $\boldsymbol{N}^{(1)*}$ the inverse of the acoustic tensor $\boldsymbol{K}^{(1)*}$ with components $K^{(1)*}_{ik} = L^{(1)*}(p)_{ijkh}n_j n_h$ and $\boldsymbol{Z}$ is the matrix defining the ellipsoidal shape of the inclusions through ${}^{\mathsf{T}}\boldsymbol{x}.\boldsymbol{Z}.\boldsymbol{x} \leq \lambda$ where λ depends on the size of the inclusion. When the shapes of the inclusions and their distribution have different ellipsoidal symmetries, the polarization tensor $\boldsymbol{P}^{(1)*}$ can be expressed by means of two different polarization tensors for the shape and the distribution labelled with subscripts i and d respectively (Ponte Castañeda and Willis, 1995).

Finding the poles of $\widetilde{\boldsymbol{L}}^{*}(p)$ requires to inspect the three possible sources of singularity due to either $\boldsymbol{L}^{(1)*}(p)$, $\boldsymbol{L}^{(2)*}(p)$ or $\boldsymbol{A}^{(2)*}(p)$. The first two options correspond to poles in the phases, whereas the third option correspond to poles created by the mixing procedure. Before dealing with a specific example in detail, let us first give an idea of the approach and of the results which can be expected in general. For this purpose, an alternative writing of effective moduli is useful,

$$\widetilde{\boldsymbol{L}}^{*} = \boldsymbol{L}^{(1)*} + c^{(2)}\left[\left(\boldsymbol{L}^{(2)*} - \boldsymbol{L}^{(1)*}\right)^{-1} + c^{(1)}\boldsymbol{P}^{(1)*}\right]^{-1}, \tag{33}$$

i) When $\boldsymbol{L}^{(2)*}(p)$ becomes singular for a specific value of p along a certain direction $\boldsymbol{e}$ in the space of second-order symmetric tensors, assuming that $\boldsymbol{L}^{(1)*}$ is non-singular for that value of p and using (33) , $\widetilde{\boldsymbol{L}}^{*} \sim \boldsymbol{L}^{(1)*} + c^{(2)}\left[(\boldsymbol{L}^{(2)*})^{-1} + c^{(1)}\boldsymbol{P}^{(1)*}\right]^{-1}$ in the direction of $\boldsymbol{e}$ and one has to check whether $\boldsymbol{P}^{(1)*}$ is invertible in that direction. If it is the case, then $\widetilde{\boldsymbol{L}}^{*} \sim \boldsymbol{L}^{(1)*}(p) + c^{(2)}/c^{(1)}(\boldsymbol{P}^{(1)*})^{-1}$ is not singular and poles of $\boldsymbol{L}^{(2)*}$ are not poles of $\widetilde{\boldsymbol{L}}^{*}$. On the other hand, if $\boldsymbol{P}^{(1)*}$ is not invertible in the direction $\boldsymbol{e}$, then $\widetilde{\boldsymbol{L}}^{*} \sim c^{(2)}\boldsymbol{L}^{(2)*}$ is singular. The former case is encountered for isotropically distributed spheroidal particles (see below), whereas the latter case is encountered in laminates (see Appendix B) or long-fiber composites.

ii) When $\boldsymbol{L}^{(1)*}(p)$ becomes singular for a specific value of p along a certain direction $\boldsymbol{e}$ in the space of second-order symmetric tensors then, in general, $\boldsymbol{P}^{(1)*}$ vanishes in that direction (according to (32)), and the product $\boldsymbol{L}^{(1)*} : \boldsymbol{P}^{(1)*}$ may have a finite limit. Assuming that $\boldsymbol{L}^{(2)*}$ is non-singular for that value of p, one has to check whether $\boldsymbol{L}^{(1)*} + c^{(2)}\left[(\boldsymbol{L}^{(2)*} - \boldsymbol{L}^{(1)*})^{-1} + c^{(1)}\boldsymbol{P}^{(1)*}\right]^{-1}$ is singular or not in that direction. In the examples below of isotropic two-phase composites, the effective shear modulus becomes singular when the shear modulus of the matrix becomes singular, so the time relaxation in shear of the matrix is also a time relaxation for the composite, whereas the effective bulk modulus remains finite when the bulk modulus of the matrix becomes singular, so that the relaxation time of the bulk modulus of the matrix is *not* a relaxation time for the composite.

iii) $\boldsymbol{A}^{(2)*}$ becomes singular when p is one of the roots of the equation

$$\det\left(\boldsymbol{I}+c^{(1)}\boldsymbol{P}^{(1)*}(p):\left(\boldsymbol{L}^{(2)*}(p)-\boldsymbol{L}^{(1)*}(p)\right)\right)=0.$$

These roots are in general different from the poles of the phases, they involve the relaxation times of both phases and are the poles created by homogenization.

3.5. Two-phase particulate composites with compressible Maxwellian phases

To get a better insight into these three options, consider the simple example of isotropic particulate materials made from a Maxwellian matrix containing Maxwellian spherical inclusions distributed isotropically[6],

$$\boldsymbol{L}^{(r)*}=3k^{(r)*}\boldsymbol{J}+2\mu^{(r)*}\boldsymbol{K},\ k^{(r)*}=k_{\mathsf{e}}^{(r)}\frac{p}{p+1/\tau_{\mathsf{m}}^{(r)}},\ \mu^{(r)*}=\mu_{\mathsf{e}}^{(r)}\frac{p}{p+1/\tau_{\mathsf{d}}^{(r)}}$$

Then, as is classical,

$$\boldsymbol{P}^{(1)*}=\frac{1}{3k_P^*}\boldsymbol{J}+\frac{1}{2\mu_P^*}\boldsymbol{K},\ k_P^*=k^{(1)^*}+\frac{4}{3}\mu^{(1)^*},\ \mu_P^*=\frac{5\mu^{(1)*}}{2}\frac{k^{(1)^*}+\frac{4}{3}\mu^{(1)^*}}{k^{(1)^*}+2\mu^{(1)^*}},\tag{34}$$

and

$$\widetilde{k}^*=k^{(1)*}+c^{(2)}\frac{k^{(2)*}-k^{(1)*}}{1+c^{(1)}\dfrac{k^{(2)*}-k^{(1)*}}{k_P^*}},\ \widetilde{\mu}^*=\mu^{(1)*}+c^{(2)}\frac{\mu^{(2)*}-\mu^{(1)*}}{1+c^{(1)}\dfrac{\mu^{(2)*}-\mu^{(1)*}}{\mu_P^*}}.\tag{35}$$

Let us concentrate first on the hydrostatic component of the tensors and examine the above three options to detect the singularities of $\widetilde{k}^*$.

i) When $k^{(2)*}$ becomes singular (*i.e.* when p approaches $-1/\tau_m^{(2)}$), $\widetilde{k}^*$ behaves as

$$\widetilde{k}^*\sim k^{(1)*}+\frac{c^{(2)}}{c^{(1)}}\left(k^{(1)^*}+\frac{4}{3}\mu^{(1)^*}\right),$$

which is non singular as $k^{(2)*}$ tends to ∞ (except when $k^{(1)*}$ is singular with $k^{(2)*}$). Therefore there is, in general, no pole of $\widetilde{k}^*$ originating from phase 2.

ii) When p approaches $-1/\tau_m^{(1)}$, $k^{(1)*}$ tends to ∞, then $\widetilde{k}^*$ behaves as

$$\widetilde{k}^*\sim\frac{1}{c^{(2)}}\left(k^{(2)*}+\frac{4}{3}c^{(1)}\mu^{(1)^*}\right),$$

which is non singular (except when $\tau_{\mathsf{m}}^{(1)}=\tau_{\mathsf{m}}^{(2)}$ or $\tau_{\mathsf{m}}^{(1)}=\tau_{\mathsf{d}}^{(1)}$ which should be considered separately). There is no singularity of $\widetilde{k}^*(p)$ originating from phase 1.

[6]The dependence on p of all starred quantities is omitted for simplicity.

iii) Finally we have to investigate the values of p for which $\boldsymbol{A}^{(2)*}$ becomes singular, *i.e.* when $1+c^{(1)}\frac{k^{(2)*}-k^{(1)*}}{k^{(1)*}+\frac{4}{3}\mu^{(1)*}}=0$. This happens when p is the root of the quadratic equation

$$\left.\begin{aligned}&p^2\left(\frac{4}{3}\mu_{\mathsf{e}}^{(1)}+c^{(2)}k_{\mathsf{e}}^{(1)}+c^{(1)}k_{\mathsf{e}}^{(2)}\right)+p\left[\frac{4}{3}\mu_{\mathsf{e}}^{(1)}\left(\frac{1}{\tau_{\mathsf{m}}^{(1)}}+\frac{1}{\tau_{\mathsf{m}}^{(2)}}\right)+\frac{1}{\tau_{\mathsf{d}}^{(1)}}\left(c^{(2)}k_{\mathsf{e}}^{(1)}+c^{(1)}k_{\mathsf{e}}^{(2)}\right)\right.\\&\left.+c^{(2)}\frac{k_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{m}}^{(2)}}+c^{(1)}\frac{k_{\mathsf{e}}^{(2)}}{\tau_{\mathsf{m}}^{(1)}}\right]+\frac{4}{3}\frac{\mu_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{m}}^{(1)}\tau_{\mathsf{m}}^{(2)}}+\frac{1}{\tau_{\mathsf{d}}^{(1)}}\left(c^{(2)}\frac{k_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{m}}^{(2)}}+c^{(1)}\frac{k_{\mathsf{e}}^{(2)}}{\tau_{\mathsf{m}}^{(1)}}\right)=0.\end{aligned}\right\}\tag{36}$$

Therefore there are, in general, two poles, or equivalently two relaxation times $\overline{\tau}_{1,\mathsf{m}}$ and $\overline{\tau}_{2,\mathsf{m}}$, for the complex bulk modulus of the composite, all of them originating from the homogenization procedure. There are two internal variables $\boldsymbol{\beta}_{i,\mathsf{m}}$, both being purely spherical, therefore two internal *scalar variables*.

The residues may be obtained by using the two limits $\widetilde{k}_{\mathsf{v}}$ (purely viscous bulk modulus) and $\widetilde{k}_{\mathsf{e}}$ (purely elastic bulk modulus) of $\widetilde{k}(p)$ as p goes to 0 and $+\infty$, namely

$$\overline{k}_1+\overline{k}_2=\widetilde{k}_{\mathsf{e}},\quad \overline{\tau}_{1,\mathsf{m}}\overline{k}_1+\overline{\tau}_{2,\mathsf{m}}\overline{k}_2=\widetilde{k}_{\mathsf{v}},\tag{37}$$

i.e.

$$\overline{k}_1=\frac{\widetilde{k}_{\mathsf{v}}-\overline{\tau}_{2,\mathsf{m}}\widetilde{k}_{\mathsf{e}}}{\overline{\tau}_{1,\mathsf{m}}-\overline{\tau}_{2,\mathsf{m}}},\quad \overline{k}_2=\frac{\widetilde{k}_{\mathsf{v}}-\overline{\tau}_{1,\mathsf{m}}\widetilde{k}_{\mathsf{e}}}{\overline{\tau}_{2,\mathsf{m}}-\overline{\tau}_{1,\mathsf{m}}}.\tag{38}$$

The same procedure can be followed to find the poles, or relaxation times, as well as the residues in shear. Alike the bulk modulus, there is no pole in the effective shear modulus associated with the inclusion phase. But unlike what was found for the bulk modulus, the pole of the matrix $-1/\tau_{\mathsf{d}}^{(1)}$ is also a pole for the composite. And there are two other poles which are the roots of the following quadratic equation

$$\left.\begin{aligned}&p^2\left[\mu_{\mathsf{e}}^{(1)}\left(k_{\mathsf{e}}^{(1)}+\frac{4}{3}\mu_{\mathsf{e}}^{(1)}\right)+\frac{2}{5}c^{(1)}\left(\mu_{\mathsf{e}}^{(2)}-\mu_{\mathsf{e}}^{(1)}\right)\left(k_{\mathsf{e}}^{(1)}+2\mu_{\mathsf{e}}^{(1)}\right)\right]\\&+p\left[\mu_{\mathsf{e}}^{(1)}\left(\frac{k_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{d}}^{(1)}}+\frac{4}{3}\frac{\mu_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{m}}^{(1)}}\right)+\frac{\mu_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{d}}^{(2)}}\left(k_{\mathsf{e}}^{(1)}+\frac{4}{3}\mu_{\mathsf{e}}^{(1)}\right)+\frac{2}{5}c^{(1)}\left(\mu_{\mathsf{e}}^{(2)}-\mu_{\mathsf{e}}^{(1)}\right)\left(\frac{k_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{d}}^{(1)}}+\frac{2\mu_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{m}}^{(1)}}\right)\right.\\&\left.+\frac{2}{5}c^{(1)}\left(\frac{\mu_{\mathsf{e}}^{(2)}}{\tau_{\mathsf{d}}^{(1)}}-\frac{\mu_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{d}}^{(2)}}\right)\left(k_{\mathsf{e}}^{(1)}+2\mu_{\mathsf{e}}^{(1)}\right)\right]\\&+\frac{\mu_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{d}}^{(2)}}\left(\frac{k_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{d}}^{(1)}}+\frac{4}{3}\frac{\mu_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{m}}^{(1)}}\right)+\frac{2}{5}c^{(1)}\left(\frac{\mu_{\mathsf{e}}^{(2)}}{\tau_{\mathsf{d}}^{(1)}}-\frac{\mu_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{d}}^{(2)}}\right)\left(\frac{k_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{d}}^{(1)}}+\frac{2\mu_{\mathsf{e}}^{(1)}}{\tau_{\mathsf{m}}^{(1)}}\right)=0.\end{aligned}\right\}\tag{39}$$

In conclusion the composite has three relaxation times in shear, on the one hand $\overline{\tau}_{1,\mathsf{d}}=\tau_{\mathsf{d}}^{(1)}$ being the relaxation time in shear of the matrix and on the other hand $\overline{\tau}_{2,\mathsf{d}}$, $\overline{\tau}_{3,\mathsf{d}}$ being associated with the roots of (39) and originating from the homogenization procedure. There are 3 tensorial (purely deviatoric) internal variables $\boldsymbol{\beta}_{i,\mathsf{d}}$, associated with each relaxation time. As for the

corresponding three residues, two equations similar to (37) are available to compute them,

$$\overline{\mu}_1 + \overline{\mu}_2 + \overline{\mu}_3 = \widetilde{\mu}_{\mathsf{e}}, \quad \overline{\tau}_{1,\mathsf{d}}\overline{\mu}_1 + \overline{\tau}_{2,\mathsf{d}}\overline{\mu}_2 + \overline{\tau}_{3,\mathsf{d}}\overline{\mu}_3 = \widetilde{\mu}_{\mathsf{v}}. \tag{40}$$

To obtain a third equation we recall that $p = -1/\tau_{\mathsf{d}}^{(1)}$ is a pole for both the matrix and the composite (denoted as $\overline{\tau}_{1,\mathsf{d}}$ for the composite). When p approaches $-1/\tau_{\mathsf{d}}^{(1)}$, μ_P^* in (34) is equivalent to $(5/3)\mu^{(1)*}(p)$ and therefore

$$\widetilde{\mu}^*(p) \sim \frac{2c^{(1)}}{5-3c^{(1)}}\mu^{(1)*} \quad \text{and} \quad \overline{\mu}_1 = \frac{2c^{(1)}}{5-3c^{(1)}}\mu_{\mathsf{e}}^{(1)}. \tag{41}$$

Finally, using (40)

$$\overline{\mu}_2 = \frac{\widetilde{\mu}_{\mathsf{v}} + (\overline{\tau}_{3,\mathsf{d}} - \overline{\tau}_{1,\mathsf{d}})\overline{\mu}_1 - \overline{\tau}_{3,\mathsf{d}}\widetilde{\mu}_{\mathsf{e}}}{\overline{\tau}_{2,\mathsf{d}} - \overline{\tau}_{3,\mathsf{d}}}, \quad \overline{\mu}_3 = \frac{\widetilde{\mu}_{\mathsf{v}} + (\overline{\tau}_{2,\mathsf{d}} - \overline{\tau}_{1,\mathsf{d}})\overline{\mu}_1 - \overline{\tau}_{2,\mathsf{d}}\widetilde{\mu}_{\mathsf{e}}}{\overline{\tau}_{3,\mathsf{d}} - \overline{\tau}_{2,\mathsf{d}}}. \tag{42}$$

In summary, the composite has five different relaxation times, one of them being a relaxation time for the matrix (in shear), the four other ones being created by the microstructure through homogenization. This is to be compared with the four relaxation times of the two phases, all together. So it is true that homogenization introduces additional memory effects, but, at least for two-phase isotropic composites, not as many as one could figure out from the general theory. Similarly there are 5 tensorial internal variables, two of them being spherical and three of them being purely deviatoric, so 17 scalar unknowns in total. This is to be compared with the 12 internal variables in the phases all together (6 per phase).

Remark 2. The physical interpretation of the internal $\boldsymbol{\beta}_i(t)$, and their relation to the local field of internal variables (viscous strain) $\boldsymbol{\varepsilon}_{\mathsf{v}}(\boldsymbol{x},t)$ is not completely known to the authors in the general case. In two particular cases considered in Appendix A and Appendix B, it can be shown that the $\boldsymbol{\beta}_i$ are related to the average per phase of the field $\boldsymbol{\varepsilon}_{\mathsf{v}}(\boldsymbol{x},t)$. However, other examples (relation (3.27) in Idiart et al., 2020) show that the effective internal variables $\boldsymbol{\beta}_i$ may also depend on the second moment per phase of the field of viscous strain.

4. Effective potentials and field statistics

Throughout this section it is assumed that the effective relations of the composite can be put in the form (23), or in other words that a finite number of relaxation times and internal variables are sufficient to reproduce (exactly or approximately) the response of the composite. It applies in particular to particulate composites.

4.1. Associated rheological model

The constitutive relations (23) are the three-dimensional analog of a rheological (one-dimensional) generalized Maxwell model, where N Maxwell one-dimensional models are assembled in parallel. An example of the rheological model corresponding to the composite analyzed in section 3.5 is given in Figure 1.

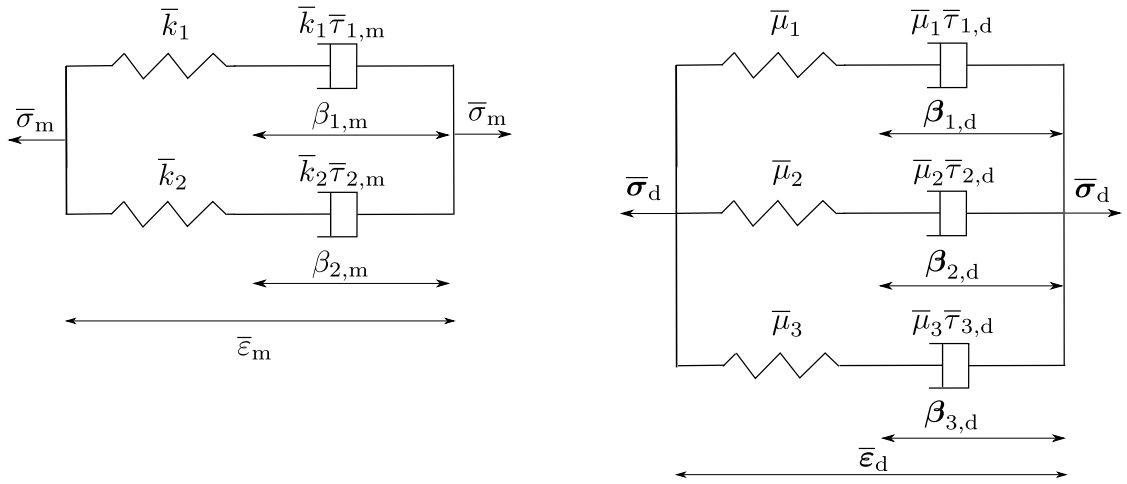

Figure 1: Diagram of the generalized Maxwell rheological model with five branches corresponding to the effective response of a particulate composite made of Maxwellian compressible phases (section 3.5). Left: two branches for the hydrostatic response, right: three branches for the deviatoric response.

The free-energy of this generalized Maxwell model is obtained by summing up the elastic energy of all springs in the model and reads

$$\overline{w}(\overline{\boldsymbol{\varepsilon}}, \boldsymbol{\beta}) = \sum_{i=1}^{N} \frac{1}{2}(\overline{\boldsymbol{\varepsilon}} - \boldsymbol{\beta}_i) : \overline{\boldsymbol{L}}_i : (\overline{\boldsymbol{\varepsilon}} - \boldsymbol{\beta}_i). \tag{43}$$

It is therefore tempting to consider that the composite material has the same free-energy as its associated rheological model. However, integrating[7] the relation (23) with respect to $\overline{\boldsymbol{\varepsilon}}$ leaves a degree of freedom, a function $g(\boldsymbol{\beta})$, in the general form of the actual effective free-energy of the composite which reads as

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}, \boldsymbol{\beta}) = \sum_{i=1}^{N} \frac{1}{2}(\overline{\boldsymbol{\varepsilon}} - \boldsymbol{\beta}_i) : \overline{\boldsymbol{L}}_i : (\overline{\boldsymbol{\varepsilon}} - \boldsymbol{\beta}_i) + g(\boldsymbol{\beta}). \tag{44}$$

We would like to have $g(\boldsymbol{\beta}) = 0$. This can be proved rigorously in a number of particular cases including two-phase incompressible composites (Appendix A), laminates (Appendix B) and the compressible composite sphere under hydrostatic loading (not included here). But we do not have a rigorous proof applying to all possible situations (microstructures), so this result is formulated here as a conjecture,

(C) it is assumed in the sequel that the effective free-energy of the composite is that of the associated rheological model, or in other words that $g(\boldsymbol{\beta}) = 0$ in (44).

Alternatively, a similar assumption could have been formulated on the dissipation function

[7]This is legitimate since it follows from (21) that the N fourth-order tensors $\overline{\boldsymbol{L}}_i$ have minor and major symmetries and are positive semi-definite.

$\widetilde{d}$ of the composite. The dissipation function for the associated rheological model reads as

$$\overline{d} = \sum_{i=1}^{N} \overline{\boldsymbol{\sigma}}_i : \dot{\boldsymbol{\beta}}_i = \sum_{i=1}^{N} \overline{\tau}_i \dot{\boldsymbol{\beta}}_i : \overline{\boldsymbol{L}}_i : \dot{\boldsymbol{\beta}}_i = \sum_{i=1}^{N} \frac{1}{\overline{\tau}_i} \overline{\boldsymbol{\sigma}}_i : \overline{\boldsymbol{M}}_i : \overline{\boldsymbol{\sigma}}_i, \tag{45}$$

where $\overline{\boldsymbol{\sigma}}_i$ denotes the stress in the i-th branch of the model (see (23)), $\overline{\boldsymbol{M}}_i$ is the inverse of $\overline{\boldsymbol{L}}_i$ (or its partial inverse when $\overline{\boldsymbol{L}}_i$ is only invertible on a subspace of symmetric second-order tensors). A conjecture similar to (C) could therefore be

(C') the effective dissipation function $\widetilde{d}$ of the composite coincides with $\overline{d}$ the dissipation function of the associated rheological model (45).

The two conjectures (C) and (C') are indeed equivalent. Assuming that (C) holds true and using relation (9) the effective dissipation reads as

$$\widetilde{d} = \overline{\boldsymbol{\sigma}} : \dot{\overline{\boldsymbol{\varepsilon}}} - \dot{\widetilde{w}} = \sum_{i=1}^{N} \overline{\boldsymbol{\sigma}}_i : \dot{\overline{\boldsymbol{\varepsilon}}} - (\overline{\boldsymbol{\varepsilon}} - \boldsymbol{\beta}_i) : \overline{\boldsymbol{L}}_i : (\dot{\overline{\boldsymbol{\varepsilon}}} - \dot{\boldsymbol{\beta}}_i) = \sum_{i=1}^{N} \overline{\boldsymbol{\sigma}}_i : \dot{\boldsymbol{\beta}}_i, \tag{46}$$

which coincides with (45). Therefore (C) implies (C') and vice-versa.

4.2. Statistics of the fields

The average per phase of the strain and stress fields in viscoelastic composites can be directly derived by the correspondence principle. For example, the Laplace-Carson transform of the time evolution of the phase-averaged strain field are given by the localization relations in this phase :

$$\boldsymbol{\varepsilon}^{(r)*}(p) \overset{\text{def}}{=} \langle \boldsymbol{\varepsilon}^*(p) \rangle^{(r)} = \boldsymbol{A}^{(r)*}(p) \, : \, \overline{\boldsymbol{\varepsilon}}^*(p) \tag{47}$$

When the effective fourth-order tensor of complex moduli can be expressed as a Prony series, the fourth-order tensors $\boldsymbol{A}^{(r)*}(p)$ are likely to be represented by a finite sum of Dirac masses at discrete relaxation times. In two-phase composites, this result is a direct consequence of Levin's relations,

$$\boldsymbol{A}^{(2)*} = \frac{1}{c^{(2)}} \left(\widetilde{\boldsymbol{L}}^* - \boldsymbol{L}^{(1)*} \right) : \left(\boldsymbol{L}^{(2)*} - \boldsymbol{L}^{(1)*} \right). \tag{48}$$

The average per phase of the strain field is obtained after transforming back the relation (47) to the time domain. The exact expressions of the average per phase of the stress field, $\boldsymbol{\sigma}^{(r)}(t) \overset{\text{def}}{=} \langle \boldsymbol{\sigma}(t) \rangle^{(r)}$, $r = 1 \dots R$, can be obtained in the same way.

Unfortunately, the correspondence principle does not permit to determine the Laplace-Carson transform of the second-order moments of the strain or stress fields[8]. So far the only exact result (to the best of the author's knowledge) is the second moment of the deviatoric stress-field in a two-phase particulate composite with incompressible phases (Badulescu et al., 2015).

[8]Indeed, the Laplace transform of a product of functions is not the product of the Laplace transforms of the individual functions, unless the quasi-elastic approximation is used to invert the Laplace-Carson transform. This approximation is used in Brenner and Masson (2005) but, unfortunately, the relation (6) and a statement in section 2.3 of this reference suggest that this equality is general, which is clearly inaccurate.

Fortunately, the effective free-energy and the effective dissipation introduced in section 2 give two relations[9] involving the second moments per phase of the stress field as follows :

$$\widetilde{w} = \langle w(t)\rangle = (1/2)\sum_{r=1}^{R} c^{(r)} \left\langle \boldsymbol{\sigma} : \boldsymbol{M}_{\mathsf{e}}^{(r)} : \boldsymbol{\sigma} \right\rangle^{(r)},\ \widetilde{d} = \langle d(t)\rangle = \sum_{r=1}^{R} c^{(r)} \left\langle \boldsymbol{\sigma} : \boldsymbol{M}_{\mathsf{v}}^{(r)} : \boldsymbol{\sigma} \right\rangle^{(r)}. \quad (49)$$

Therefore, the expressions (44) and (45), together with conjecture (C), give two relations between the second moments of the stress field.

4.3. Stress fluctuations in isotropic two-phase particulate compressible composites

Consider two-phase isotropic particulate composites. Both phases are viscoelastic and Maxwellian, with compressible elasticity (characterized by a bulk and a shear elastic modulus) and incompressible viscosity (characterized by a shear viscous modulus). The Hashin-Shtrikman estimates are relevant for particulate composites and, as seen previously, the corresponding relaxation function can be expressed as a Prony series, with two relaxation times for the hydrostatic relaxation function and three relaxation times for the shear relaxation function.

The two last relations (49) read as :

$$\widetilde{w}(t) = \frac{1}{2}\sum_{r=1}^{2} c^{(r)} \left(\frac{1}{3\mu_{\mathsf{e}}^{(r)}} \left\langle \sigma_{\mathsf{eq}}^2(t)\right\rangle^{(r)} + \frac{1}{k_{\mathsf{e}}^{(r)}} \left\langle \sigma_{\mathsf{m}}^2(t)\right\rangle^{(r)} \right),\ \widetilde{d}(t) = \sum_{r=1}^{2} \frac{c^{(r)}}{3\mu_{\mathsf{v}}^{(r)}} \left\langle \sigma_{\mathsf{eq}}^2(t)\right\rangle^{(r)}, \quad (50)$$

where $\sigma_{\mathsf{m}} = (1/3)\mathrm{tr}(\boldsymbol{\sigma})$, $\boldsymbol{\sigma}_{\mathsf{d}} = \boldsymbol{\sigma} - \sigma_{\mathsf{m}}\boldsymbol{i}$ and $\sigma_{\mathsf{eq}} = \left(\frac{3}{2}\boldsymbol{\sigma}_{\mathsf{d}} : \boldsymbol{\sigma}_{\mathsf{d}}\right)^{1/2}$. The only non-vanishing fluctuations of the stress field are in phase 1 (matrix) with

$$C_{\mathsf{d}}^{(1)}(t) = \frac{2}{3}\left(\left\langle \sigma_{\mathsf{eq}}^2(t)\right\rangle^{(1)} - (\overline{\sigma}_{\mathsf{eq}}^{(1)}(t))^2 \right), \quad C_{\mathsf{m}}^{(1)}(t) = \left\langle \sigma_{\mathsf{m}}^2(t)\right\rangle^{(1)} - (\overline{\sigma}_{\mathsf{m}}^{(1)}(t))^2.$$

It follows from (50), that

$$C_{\mathsf{d}}^{(1)}(t) = \frac{2\,\mu_{\mathsf{v}}^{(1)}}{c^{(1)}} \left(\widetilde{d}(t) - \frac{c^{(2)}}{3\,\mu_{\mathsf{v}}^{(2)}} (\overline{\sigma}_{\mathsf{eq}}^{(2)}(t))^2 \right) - \frac{2}{3}(\overline{\sigma}_{\mathsf{eq}}^{(1)}(t))^2, \quad (51)$$

and

$$C_{\mathsf{m}}^{(1)}(t) = \frac{k_{\mathsf{e}}^{(1)}}{c^{(1)}} \left[2\,\widetilde{w}(t) - \frac{c^{(1)}}{3\,\mu_{\mathsf{e}}^{(1)}} \left(\frac{3}{2} C_{\mathsf{d}}^{(1)}(t) + (\overline{\sigma}_{\mathsf{eq}}^{(1)}(t))^2 \right) - c^{(2)} \left(\frac{1}{3\,\mu_{\mathsf{e}}^{(2)}} (\overline{\sigma}_{\mathsf{eq}}^{(2)}(t))^2 + \frac{1}{k_{\mathsf{e}}^{(2)}} (\overline{\sigma}_{\mathsf{m}}^{(2)}(t))^2 \right) \right] \quad (52)$$

Therefore the stress fluctuations in the matrix along a path of imposed overall strain $\overline{\boldsymbol{\varepsilon}}(t)$ can be obtained as follows: the $\overline{\boldsymbol{L}}_i$ and $\overline{\tau}_i$ being known, the $\boldsymbol{\beta}_i(t)$ are determined by integration of the differential equations (23), then the effective free-energy and the effective dissipation function are evaluated using (44) and (46), the phase-average of the stresses are computed by

[9] The Hill-Mandel relation used in Badulescu et al. (2015) does not provide a third independent relation by virtue of (9).

the correspondence principle and finally the stress fluctuations in the matrix are obtained by means of (51) and (52).

4.4. Comparisons to full-field simulations for dispersions of elastic or viscous inclusions in a viscoelastic matrix

This exact result for the stress fluctuations is compared with full-field simulations. The RVE is a cube containing spherical particles with the same radius distributed isotropically using the random sequential addition (or adsorption, RSA) algorithm (Widom, 1966): the particles are progressively added into the RVE. To prevent overlapping of the inclusions, a new particle is added only if it does not intersect any of the already existing particles. If an inclusion intersects the boundary of the RVE, it is duplicated on the opposite face to ensure periodicity of the microstructure. Five different RVEs have been generated using this algorithm with 10% volume fraction of inclusions (containing over 120 particles). They are shown in Figure 2. The

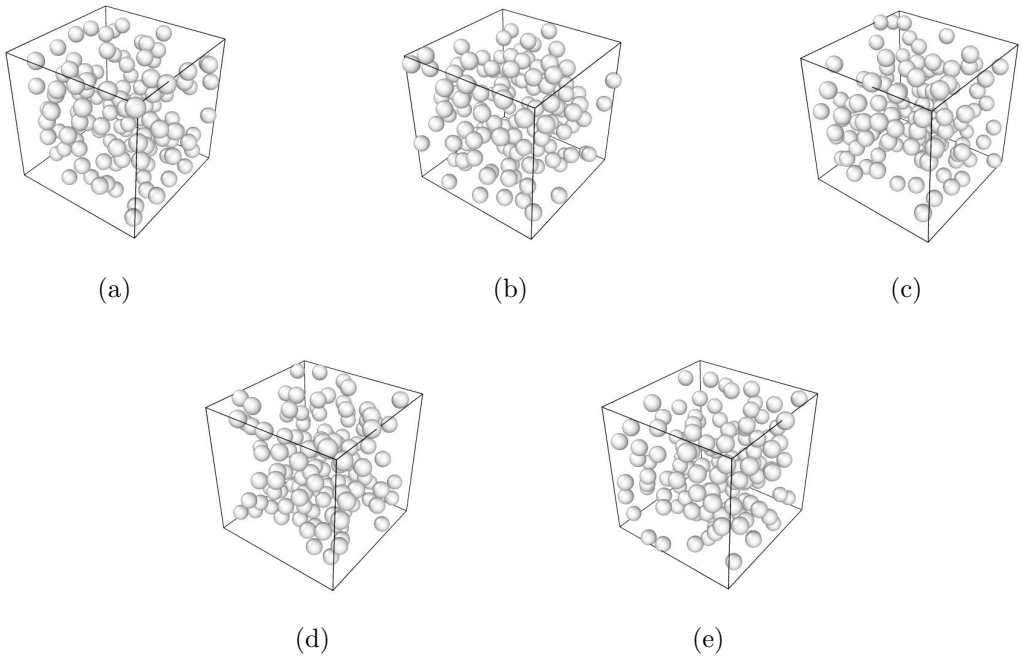

Figure 2: The different volume elements considered in this study ($c^{(2)} = 10\%$).

computational method used for this analysis is based on Fast Fourier Transforms, originally proposed by Moulinec and Suquet (1994) and Moulinec and Suquet (1998). The fixed point algorithm of Moulinec and Suquet (1998) is used, an implicit (backward) Euler method is used for the integration in time of the local constitutive relations. The material data (see table 1) are identical to those in Badulescu et al. (2015) and Cotelo et al. (2020), except for the volume fraction of inclusions which is 10% here.

The isotropy and stationarity of the volume element are assessed by computing the effective properties in the Laplace-Carson space for a very small value of the real variable p which corresponds to a highly contrasted composite (almost purely viscous behaviour). In this extreme situation, the influence of the grid refinement has first been studied as follows: when the number

	$\mu_{\mathsf{e}}^{(1)}$	$\mu_{\mathsf{e}}^{(2)}$	$\mu_v^{(1)}$	$\mu_v^{(2)}$	$\kappa_e^{(1)}$	$\kappa_e^{(2)}$
Figure 3	3GPa	6GPa	30 GPa s	∞	10 GPa	20 GPa
Figure 4	2GPa	20GPa	20 GPa s	2 GPa s	10 GPa	20 GPa

Table 1: Material Data used in the simulations.

of voxels is increased from 256^3 to 512^3, the relative deviation on the effective shear modulus does not exceed 0.8% (this deviation is of the order of 1.4% when the number of voxels is decreased from 256^3 to 128^3). Then, for a grid refinement larger than 256^3, the simulated overall shear modulus of this pseudo-elastic composite varies as follows :

- Deviation from isotropy : when the volume element is submitted to shear tests in six different possible directions (3 axisymmetric shear loading, 3 pure shear loading) the deviation does not exceed 0.7% ;
- Influence of the sample : the deviation between the five different realizations of this volume element is less than 0.9%.

In conclusion, a resolution of 256^3 voxels appears to be a good compromise between computational cost and accuracy and is used in the results presented below.

4.4.1. The "well-ordered" case

We consider first an example where the material data of the phases are "well-ordered", or in other words the inclusions are stiffer than the matrix in terms of both their elastic properties and viscous properties (the inclusions being purely elastic).

Following Badulescu et al. (2015) and Cotelo et al. (2020), a first test consisting of a prescribed macroscopic strain path in the form of an axisymmetric shear loading is considered :

$$\dot{\overline{\boldsymbol{\varepsilon}}}(t) = \dot{\bar{\varepsilon}}_0 \left(\boldsymbol{e}_1 \otimes \boldsymbol{e}_1 - \frac{1}{2}\, \boldsymbol{e}_2 \otimes \boldsymbol{e}_2 - \frac{1}{2}\, \boldsymbol{e}_3 \otimes \boldsymbol{e}_3 \right)$$

where $\dot{\bar{\varepsilon}}_0$ oscillates between positive and negative values:

$$\text{when} \quad 0 \leq t \leq T \text{ and } 3\,T \leq t \leq 4\,T : \dot{\bar{\varepsilon}}_0 = 5\,10^{-2}\,s^{-1}, \quad \text{and when} \quad T < t < 3\,T : \dot{\bar{\varepsilon}}_0 = -5\,10^{-2}\,s^{-1},$$

with $T = 30\,s$. Hashin-Shtrikman estimates provided by the correspondence principle yield the time evolutions of the macroscopic stress and the phase-average of the stress (and strain) fields. These estimates are denoted CP HS in the next figures. As the Laplace-Carson inverse is exact in this situation, the results in the time domain are exact within the approximation of the Hashin-Shtrikman estimate.

The expressions (51) and (52) provide two relations for the fluctuations of the stress invariants in the matrix (these fluctuations vanish in the inclusions). In the present example, the viscous strains in the phase are purely deviatoric so that the effective constitutive relations are those of a generalized Maxwell model with four branches: one branch for hydrostatic stresses[10]

[10]The equation (36) defining the relaxation times on the hydrostatic behaviour has only one non zero solution when the two relaxation times of the phase ($\tau_{\mathsf{m}}^{(1)}$, $\tau_{\mathsf{m}}^{(2)}$) tend to infinity.

and three branches for deviatoric stresses.

As expected (at least for the considered volume fraction), the macroscopic and phase averages of the stress are well predicted by the Hashin-Shtrikman estimates provided by the correspondence principle (see Figure 3 left). In addition, the predictions of the fluctuations in the matrix using relations (51) and (52) are very similar to the ones predicted by full-field simulations as shown in Figure 3 right.

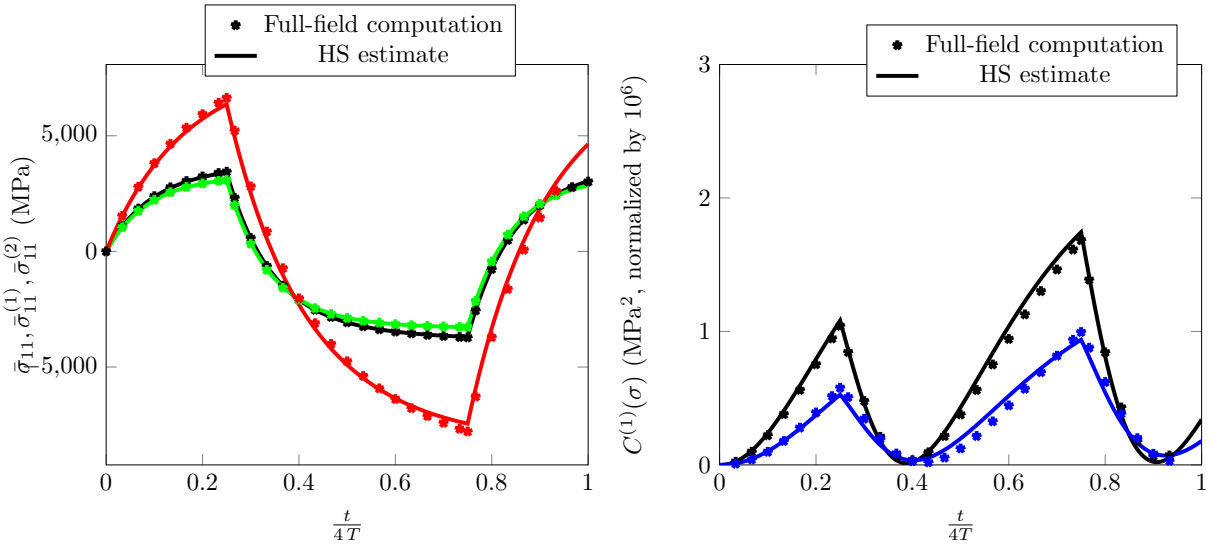

Figure 3: Compressible particulate composites with spherical inclusions. Material data and loading as in table 1, $c^{(2)} = 10\%$)/ Left: macroscopic (black) and average stress in the inclusions (red) and the matrix (green). Right: stress fluctuations in the matrix (black: deviatoric stress (51), blue: hydrostatic stress (52)).

4.4.2. The "non-well-ordered" case

Following Cotelo et al. (2020), we now consider the case where the inclusions, which now exhibit a viscous behaviour, are stiffer than the matrix in terms of elastic properties and softer in terms of viscous properties. The corresponding material data are reported in table 1 (see the line corresponding to figures 4 and 5).

A macroscopic strain in the form of a pure shear loading is prescribed :

$$\overline{\boldsymbol{\varepsilon}}(t) = \frac{\overline{\gamma}(t)}{2}\Big(\boldsymbol{e}_1 \otimes \boldsymbol{e}_2 + \boldsymbol{e}_2 \otimes \boldsymbol{e}_1\Big), \quad \text{with } \dot{\overline{\gamma}} = \sqrt{3}\,10^{-3}\,s^{-1}.$$

Two volume fractions of inclusions have been considered $c^{(2)} = 0.1$ and $c^{(2)} = 0.2$ respectively and the results are reported in figures 4 and 5.

Again, the CP HS predictions for the time evolution of the macroscopic stress, the phase-average of the stress (and strain) fields as well as their fluctuations are in good agreement with the full-field simulations. Moreover, the non monotonic evolution with time of the fluctuations observed by Cotelo et al. (2020) are confirmed here for these two particular volume fractions, both by the exact expressions of the fluctuations and by the full-field simulations.

The agreement is slightly better at low volume fraction (10%) than at higher volume fraction (20%).The reason is that the distribution of inclusions in the FFT simulations is monodisperse

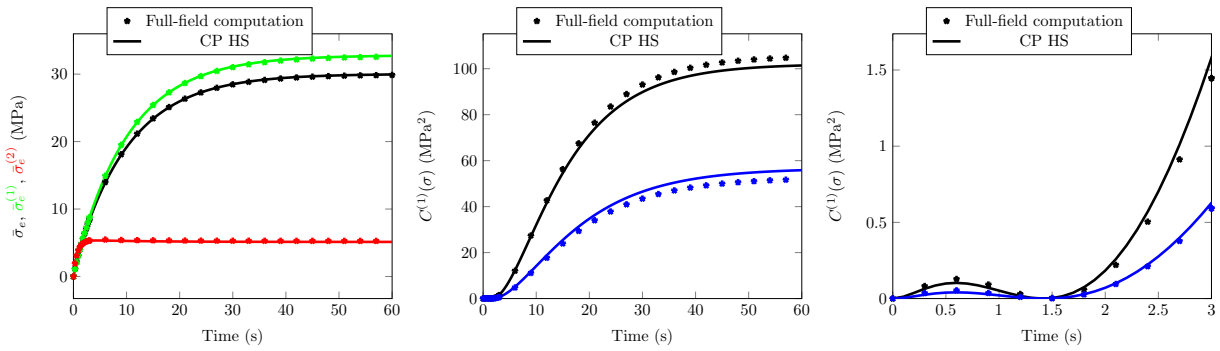

Figure 4: Compressible particulate composites with spherical inclusions. Material data and loading as in table 1, $c^{(2)} = 10\%$)/ Left: macroscopic (black) and average stress in the inclusions (red) and the matrix (green). Middle: stress fluctuations in the matrix over the whole range of time (black: deviatoric stress (51), blue: hydrostatic stress (52)). Right: look-up of the stress fluctuations at small times.

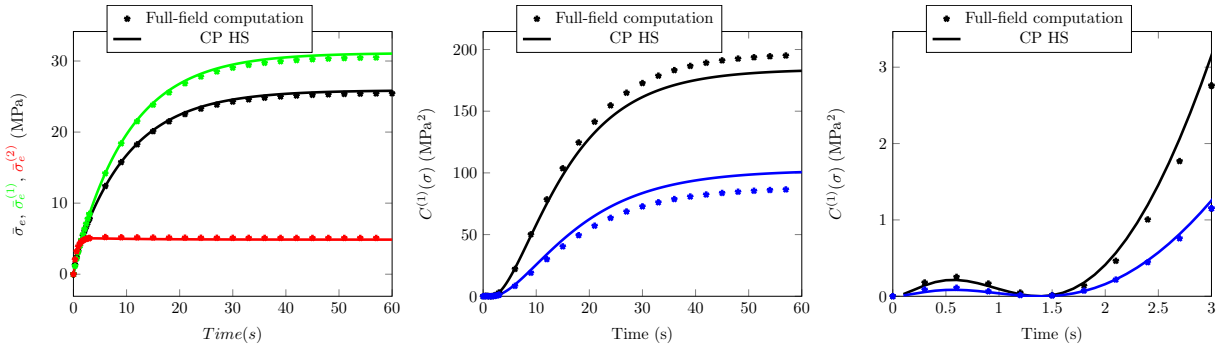

Figure 5: Compressible particulate composites with spherical inclusions. Material data and loading as in table 1, $c^{(2)} = 20\%$)/ Left: macroscopic (black) and average stress in the inclusions (red) and the matrix (green). Middle: stress fluctuations in the matrix over the whole range of time (black: deviatoric stress (51), blue: hydrostatic stress (52)). Right: look-up of the stress fluctuations at small times.

(all inclusions have the same size). It is therefore expected that the FFT results deviate from the predictions of the Hashin-Shtrikman relations as the volume fraction increases, the interaction between neighbouring inclusions being taken into account differently in both approaches. A closer inspection of the local fields in the FFT simulations reveals that there are fluctuations in the strain field within the inclusion phase, in contrast with a building block of the Hashin-Shtrikman theory.

The effect of the volume fraction of inclusions can be seen by comparing the two plots at different volume fractions. The asymptotic overall stress (black curve on the most left plot of each figure) is indeed lower for the composite at 20% volume fraction than at 10%. This counter-intuitive observation results from the material data, the viscosity in the inclusions being lower than that of the matrix (therefore the effective viscosity of the composite is lower at a volume fraction of 20% than at 10%). A closer inspection of the plots at small strains (in the elastic regime), shows that the curves are inverted in that regime, *i.e.* that the composite at 20% volume fraction of inclusions is indeed stiffer (elastically) than the composite at 10%.

5. Conclusions

New theoretical relations in linear viscoelasticity have been derived by combining two different points of view. On the one hand, the general thermodynamic framework makes it possible to define the energy stored and the energy dissipated in linearly viscoelastic composites (and more generally in dissipative composites), as the average of the local free-energy and dissipation in the phases. On the other hand, thanks to the correspondence principle, the macroscopic strain-stress relation can be formulated as ordinary differential equations for a set of effective internal variables. The number of these internal variables depends on the microstructure and this dependence has been illustrated. However a finite and small number of internal variables is rigorously sufficient in several cases of interest, including in particular (but not limited to) particulate composites where an inclusion phase is dispersed in another phase playing the role of a matrix. In simple cases, these internal variables can be directly related to the statistics of the field of viscous strain within the phases.

Interpreting the macroscopic response (derived by means of the correspondence principle) as a rheological generalized Maxwell model allows us to express the macroscopic free and dissipated energy of the composite by means of quantities (fourth-order tensors and relaxation times) which appear in the overall constitutive relations, without explicit knowledge of the local fields. This interpretation is proved to be exact in several cases of interest (incompressible isotropic particulate composites or laminates) and the authors believe that it is a general result. Coupled with the Hashin-Shtrikman estimates, these thermodynamic functions provide additional information on the statistics of the field. For instance, beyond the average per-phase of the stress field directly available from the correspondence principle, two additional relations on the second moments per phase of this field are derived. Comparison with full-field simulations on particulate composites show the relevance of these relations.

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Appendix A. Isotropic two-phase particulate and incompressible composites

The composites considered in this appendix are two-phase composites composed of Maxwellian, isotropic and incompressible phases. Phase 1 is the matrix in which spheroidal particles of phase 2 are randomly dispersed. This class of composites have been studied in Suquet (2012) and Badulescu et al. (2015) [11] (among others) where it is established that the effective constitutive relations correspond to a generalized Maxwell model with two relaxation times[12], which read (with the corresponding residues) as[13]

$$\left.\begin{array}{l} \overline{\tau}_1 = \tau^{(1)}, \overline{\mu}_1 = \mu_{\mathsf{e}}^{(1)} \dfrac{c^{(1)}(1-\beta)}{1-\beta c^{(1)}}, \quad \beta = \dfrac{2}{2+d}, \\ \overline{\tau}_2 = \tau^{(1)}\tau^{(2)} \dfrac{A}{B}, \quad \overline{\mu}_2 = \dfrac{c^{(2)}}{1-\beta c^{(1)}} \dfrac{\mu_{\mathsf{e}}^{(1)}\mu_{\mathsf{e}}^{(2)}}{A}, \\ \text{where } A = \mu_{\mathsf{e}}^{(1)}(1-\beta c^{(1)}) + \mu_{\mathsf{e}}^{(2)}\beta c^{(1)}, \quad B = \tau^{(1)}\mu_{\mathsf{e}}^{(1)}(1-\beta c^{(1)}) + \tau^{(2)}\mu_{\mathsf{e}}^{(2)}\beta c^{(1)}, \end{array}\right\} \tag{A.1}$$

There are therefore 2 internal variables $\boldsymbol{\beta}_1$ and $\boldsymbol{\beta}_2$ which are deviatoric second-order tensors. The effective constitutive relations (for the stress deviator only, since the composite is incompressible) are,

$$\overline{\boldsymbol{\sigma}}_{\mathsf{d}} = \sum_{i=1}^{2} \overline{\boldsymbol{\sigma}}_i, \quad \overline{\boldsymbol{\sigma}}_i = 2\overline{\mu}_i(\overline{\boldsymbol{\varepsilon}} - \boldsymbol{\beta}_i), \quad \overline{\tau}_i\dot{\boldsymbol{\beta}}_i + \boldsymbol{\beta}_i = \overline{\boldsymbol{\varepsilon}},\ i = 1, 2,$$

which can be re-arranged into

$$\dot{\overline{\boldsymbol{\sigma}}}_i + \frac{1}{\overline{\tau}_i}\overline{\boldsymbol{\sigma}}_i = 2\overline{\mu}_i\dot{\overline{\boldsymbol{\varepsilon}}}, \quad \overline{\boldsymbol{\sigma}}_i = 2\overline{\tau}_i\dot{\boldsymbol{\beta}}_i,\ i = 1, 2. \tag{A.2}$$

Appendix A.1. Physical interpretation of the internal variables $\boldsymbol{\beta}_1$ and $\boldsymbol{\beta}_2$.

The Laplace-Carson transform of the viscous strain field is related to the Laplace-Carson transform of the strain field by the following relation :

$$\boldsymbol{\varepsilon}_{\mathsf{v}}^*(p, \boldsymbol{x}) = \frac{1}{p}\,\boldsymbol{M}_{\mathsf{v}}^{(r)} : \boldsymbol{\sigma}^*(p, \boldsymbol{x}) = \frac{1}{p}\,\boldsymbol{M}_{\mathsf{v}}^{(r)}\,\boldsymbol{L}^{(r)*}(p) : \boldsymbol{\varepsilon}^*(p, \boldsymbol{x}),\ \boldsymbol{x} \in V^{(r)}$$

and averaging this relation over phase r yields :

$$\langle \boldsymbol{\varepsilon}_{\mathsf{v}}^* \rangle^{(r)}(p) = \frac{1}{p}\,\boldsymbol{M}_{\mathsf{v}}^{(r)} : \boldsymbol{L}^{(r)*}(p) : \boldsymbol{A}^{(r)*}(p) : \overline{\boldsymbol{\varepsilon}}^*(p) \tag{A.3}$$

[11] Note that in these earlier works the phase numbering is reversed with respect to the present notations.

[12] Since the phases are incompressible the relaxation times in shear $\tau_{\mathsf{d}}^{(r)}$ are simply denoted $\tau^{(r)}$ in this appendix.

[13] In these relations, β should not be confused with one of the internal variables, $\beta = 1/2$ in dimension 2 and $\beta = 2/5$ in dimension 3.

In the present situation

$$\boldsymbol{A}^{(2)*}(p) = \frac{\mu^{(1)*}(p)}{\mu^{(1)*}(p)\,(1-\beta c^{(1)}) + \mu^{(2)*}(p)\,\beta\,c^{(1)}}\boldsymbol{K},$$

$$\boldsymbol{L}^{(2)*}(p) : \boldsymbol{A}^{(2)*}(p) = \frac{2\mu^{(1)*}(p)\,\mu^{(2)*}(p)}{\mu^{(1)*}(p)\,(1-\beta c^{(1)}) + \mu^{(2)*}(p)\,\beta\,c^{(1)}}\boldsymbol{K} = \frac{2\mu_{\mathsf{e}}^{(1)}\,\mu_{\mathsf{e}}^{(2)}}{A}\frac{p}{p+1/\overline{\tau}_2}\boldsymbol{K},$$

and with $\boldsymbol{M}_{\mathsf{v}}^{(2)} = 1/\left(2\tau^{(2)}\mu_{\mathsf{e}}^{(2)}\right)\boldsymbol{K}$,

$$\langle\boldsymbol{\varepsilon}_{\mathsf{v}}^{*}\rangle^{(2)}(p) = \frac{\mu_{\mathsf{e}}^{(1)}}{\tau^{(2)}A}\left(\frac{p}{p+1/\overline{\tau}_2}\right)\overline{\boldsymbol{\varepsilon}}^{*}(p) = \frac{\overline{\tau}_2}{\tau^{(2)}}\frac{\mu_{\mathsf{e}}^{(1)}}{A}\boldsymbol{\beta}_2^{*}(p).$$

In other words

$$\boldsymbol{\beta}_2(t) = \left(1-\beta c^{(1)} + \beta c^{(1)}\frac{\mu_{\mathsf{e}}^{(2)}\tau^{(2)}}{\mu_{\mathsf{e}}^{(1)}\tau^{(1)}}\right)\langle\boldsymbol{\varepsilon}_{\mathsf{v}}\rangle^{(2)}(t). \tag{A.4}$$

Similarly

$$c^{(1)}\boldsymbol{L}^{(1)*}(p) : \boldsymbol{A}^{(1)*}(p) = \widetilde{\boldsymbol{L}}(p) - c^{(2)}\boldsymbol{L}^{(2)*}(p) : \boldsymbol{A}^{(2)*}(p) = \left(2\overline{\mu}_1\frac{p}{p+1/\overline{\tau}_1} + 2\overline{\mu}_2\beta c^{(1)}\frac{p}{p+1/\overline{\tau}_2}\right)\boldsymbol{K}$$

and

$$\langle\boldsymbol{\varepsilon}_{\mathsf{v}}^{*}\rangle^{(1))}(p) = \frac{1}{c^{(1)}}\frac{\overline{\mu}_1\overline{\tau}_1}{\mu_{\mathsf{e}}^{(1)}\tau^{(1)}}\boldsymbol{\beta}_1^{*}(p) + \beta\frac{\overline{\tau}_2\overline{\mu}_2}{\mu_{\mathsf{e}}^{(1)}\tau^{(1)}}\boldsymbol{\beta}_2^{*}(p),$$

or in other words

$$\boldsymbol{\beta}_1(t) = \frac{1-\beta c^{(1)}}{1-\beta}\langle\boldsymbol{\varepsilon}_{\mathsf{v}}\rangle^{(1)}(t) - \frac{\beta c^{(2)}}{1-\beta}\frac{\mu_{\mathsf{e}}^{(2)}\tau^{(2)}}{\mu_{\mathsf{e}}^{(1)}\tau^{(1)}}\langle\boldsymbol{\varepsilon}_{\mathsf{v}}\rangle^{(2)}(t). \tag{A.5}$$

Relations (A.4) and (A.5) display how the internal variables $\boldsymbol{\beta}_i$ are related to the average of the local viscous strain field in the phases in this specific example.

Appendix A.2. Proof of conjecture (C') for this class of composites

The local constitutive relations in phase (r) read as

$$\dot{\boldsymbol{\varepsilon}}(\boldsymbol{x},t) = \frac{1}{2\mu_{\mathsf{e}}^{(r)}}\left(\dot{\boldsymbol{\sigma}}_{\mathsf{d}}(\boldsymbol{x},t) + \frac{1}{\tau_{\mathsf{d}}^{(r)}}\boldsymbol{\sigma}_{\mathsf{d}}(\boldsymbol{x},t)\right). \tag{A.6}$$

Multiplying (A.6) by $\boldsymbol{\sigma}_{\mathsf{d}}$, averaging over V and applying the Hill-Mandel lemma (8), a differential equation is obtained relating the average dissipation $d^{(r)}$ in the different phases

$$\sum_{r=1}^{2} c^{(r)}\left(\frac{1}{2}\tau^{(r)}\dot{d}^{(r)} + d^{(r)}\right) = \overline{\boldsymbol{\sigma}}_{\mathsf{d}} : \dot{\overline{\boldsymbol{\varepsilon}}}, \tag{A.7}$$

where (thanks to the incompressibility of the phases)

$$d^{(r)} = \langle \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}}_{\mathsf{v}} \rangle^{(r)} = \langle \boldsymbol{\sigma}_{\mathsf{d}} : \dot{\boldsymbol{\varepsilon}}_{\mathsf{v}} \rangle^{(r)} = \frac{1}{2\mu_{\mathsf{e}}^{(r)}\tau^{(r)}} \langle \boldsymbol{\sigma}_{\mathsf{d}} : \boldsymbol{\sigma}_{\mathsf{d}} \rangle^{(r)} . \tag{A.8}$$

and after a few manipulations, the effective dissipation $\widetilde{d} = \sum_{r=1}^{2} c^{(r)} d^{(r)}$ of the composite is found to solve the differential equation

$$\frac{1}{2}\tau^{(1)}\dot{\widetilde{d}} + \widetilde{d} = \overline{\boldsymbol{\sigma}}_{\mathsf{d}} : \dot{\overline{\boldsymbol{\varepsilon}}} - c^{(2)}(\tau^{(2)} - \tau^{(1)})\dot{d}^{(2)}. \tag{A.9}$$

Symmetrically, multiplying the first equation in (A.2) by $\overline{\boldsymbol{\sigma}}_i$ yields two differential equations for the dissipation in each branch of the rheological model

$$\overline{d}_i = \overline{\boldsymbol{\sigma}}_i : \dot{\boldsymbol{\beta}}_i = \frac{1}{2\,\overline{\tau}_i\,\overline{\mu}_i}\overline{\boldsymbol{\sigma}}_i : \overline{\boldsymbol{\sigma}}_i, \quad \frac{1}{2}\overline{\tau}_i\,\dot{\overline{d}}_i + \overline{d}_i = \overline{\boldsymbol{\sigma}}_i : \dot{\overline{\boldsymbol{\varepsilon}}}, \quad i = 1, 2.$$

Adding these two differential equations and recalling that $\overline{\tau}_1 = \tau^{(1)}$ gives a differential equation for the total dissipation in the rheological model $\overline{d} = \overline{d}_1 + \overline{d}_2$,

$$\frac{1}{2}\tau^{(1)}\dot{\overline{d}} + \overline{d} = \overline{\boldsymbol{\sigma}}_{\mathsf{d}} : \dot{\boldsymbol{\varepsilon}} + (\overline{\tau}_2 - \tau^{(1)})\dot{\overline{d}}_2. \tag{A.10}$$

According to (A.2), the stress $\overline{\boldsymbol{\sigma}}_2$ in the second branch of the rheological model solves a first-order differential equation

$$\dot{\overline{\boldsymbol{\sigma}}}_2 + \frac{1}{\overline{\tau}_2}\overline{\boldsymbol{\sigma}}_2 = 2\overline{\mu}_2\dot{\overline{\boldsymbol{\varepsilon}}}. \tag{A.11}$$

Badulescu et al. (2015) established that the first moment of the stress deviator $\boldsymbol{\sigma}_{\mathsf{d}}{}^{(2)}$ in phase 2 solves a quite similar first-order differential equation, except for its right hand side,

$$\dot{\boldsymbol{\sigma}}_{\mathsf{d}}^{(2)} + \frac{1}{\overline{\tau}_2}\boldsymbol{\sigma}_{\mathsf{d}}^{(2)} = 2\overline{\mu}_2\frac{1 - \beta c^{(1)}}{c^{(2)}}\dot{\overline{\boldsymbol{\varepsilon}}}. \tag{A.12}$$

Since it is assumed that the initial conditions for $\overline{\boldsymbol{\sigma}}_2$ and $\boldsymbol{\sigma}_{\mathsf{d}}^{(2)}$ are the same ($\mathbf{0}$), the solutions of (A.11) and (A.12) are proportional,

$$\overline{\boldsymbol{\sigma}}_2 = \frac{c^{(2)}}{1 - \beta c^{(1)}}\boldsymbol{\sigma}_{\mathsf{d}}^{(2)}. \tag{A.13}$$

By virtue of (A.13), the dissipation $\overline{d}_2$ in the second branch of the rheological model can be related to the dissipation $d^{(2)}$ in the inclusion phase,

$$\overline{d}_2 = \frac{1}{2\,\overline{\mu}_2\,\overline{\tau}_2}\overline{\boldsymbol{\sigma}}_2 : \overline{\boldsymbol{\sigma}}_2 = \frac{1}{2\,\overline{\mu}_2\,\overline{\tau}_2}\left(\frac{c^{(2)}}{1 - \beta c^{(1)}}\right)^2 \boldsymbol{\sigma}_{\mathsf{d}}^{(2)} : \boldsymbol{\sigma}_{\mathsf{d}}^{(2)} = \frac{\mu_{\mathsf{e}}^{(2)}\tau^{(2)}}{\overline{\mu}_2\,\overline{\tau}_2}\left(\frac{c^{(2)}}{1 - \beta c^{(1)}}\right)^2 d^{(2)}.$$

A straightforward calculation using (A.1) shows that

$$(\overline{\tau}_2 - \tau^{(1)})\overline{d}_2 = c^{(2)}(\tau^{(2)} - \tau^{(1)})\, d^{(2)}.$$

Therefore the two differential equations (A.9) and (A.10) coincide, and their solutions $\widetilde{d}$ and $\overline{d}$ having the same initial conditions (zero) are equal. This proves that the effective dissipation of the composite is equal to the dissipation in the associated rheological model $\widetilde{d} = \overline{d}$.

Appendix A.3. Retrieving the differential equation of Badulescu et al. (2015) for the second moment of the stress in the matrix.

Badulescu et al. (2015) derived in their appendix A.1 (between their equations (A.13) and (A.14)) a differential equation for the second moment of the stress field in the matrix. This differential equation can be retrieved from equation (A.7) by arranging it as

$$2\mu_{\mathsf{e}}^{(1)}\left(\tau^{(1)}\dot{d}^{(1)} + 2d^{(1)}\right) = \frac{4\mu_{\mathsf{e}}^{(1)}}{c^{(1)}}\overline{\boldsymbol{\sigma}}_{\mathsf{d}} : \dot{\overline{\boldsymbol{\varepsilon}}} - 2\mu_{\mathsf{e}}^{(1)}\frac{c^{(2)}}{c^{(1)}}\left(\tau^{(2)}\dot{d}^{(2)} + 2d^{(2)}\right),$$

which coincides with the differential equation in Badulescu et al. (2015), after recalling that the dissipation in phase r and the second moment of the stress in that phase are related by (A.8).

Appendix B. Laminates

The materials considered here are laminates with layers normal to a unit vector $\boldsymbol{n}$. To avoid overwhelming technical details, attention is limited here to incompressible phases. However all results (in particular the validity of conjecture (C')) extend to compressible phases, at the expense of rather involved expressions.

Appendix B.1. Linear elasticity

The effective elasticity tensor of a laminate can be expressed as

$$\left(\widetilde{\boldsymbol{L}} - \boldsymbol{L}^{(1)}\right)^{-1} = \frac{1}{c^{(2)}}\left(\boldsymbol{L}^{(2)} - \boldsymbol{L}^{(1)}\right)^{-1} + \frac{c^{(1)}}{c^{(2)}}\boldsymbol{\Gamma}^{(1)}(\boldsymbol{n}), \tag{B.1}$$

with

$$\boldsymbol{\Gamma}^{(1)}(\boldsymbol{n}) = \boldsymbol{n}\otimes\boldsymbol{N}^{(1)}(\boldsymbol{n})\otimes\boldsymbol{n}|_s, \quad \boldsymbol{N}^{(1)} = \left(\boldsymbol{K}^{(1)}\right)^{-1}, \quad \boldsymbol{K}^{(1)} = \boldsymbol{n}.\boldsymbol{L}^{(1)}.\boldsymbol{n},$$

where the symbol $|_s$ denotes symmetrization (major and minor symmetries). For an isotropic material with Lamé constant λ and μ, then

$$\boldsymbol{\Gamma}^{(1)}(\boldsymbol{n}) = \frac{1}{\mu}\boldsymbol{n}\otimes\boldsymbol{i}\otimes\boldsymbol{n}|_s - \frac{\lambda+\mu}{\mu(\lambda+2\mu)}\boldsymbol{n}\otimes\boldsymbol{n}\otimes\boldsymbol{n}\otimes\boldsymbol{n}.$$

The local fields in a laminate are uniform within each phase. The local strain and stress field read as

$$\boldsymbol{\varepsilon}^{(2)} = \boldsymbol{A}^{(2)} : \overline{\boldsymbol{\varepsilon}} \text{ with } \boldsymbol{A}^{(2)} = \left[\boldsymbol{I} + c^{(1)}\boldsymbol{\Gamma}^{(1)} : \left(\boldsymbol{L}^{(2)} - \boldsymbol{L}^{(1)}\right)\right]^{-1},\ \boldsymbol{\varepsilon}^{(1)} = \frac{1}{c^{(1)}}\left(\overline{\boldsymbol{\varepsilon}} - c^{(2)}\boldsymbol{\varepsilon}^{(2)}\right). \tag{B.2}$$

Since the fields are uniform within each phase, higher order moments of the fields are directly obtained from their first moment.

When the phases are incompressible, then the laminate itself is incompressible and the stiffness tensor reads as

$$\boldsymbol{L}^{(r)} = +\infty \boldsymbol{J} + 2\mu^{(r)}\boldsymbol{K}, \quad \boldsymbol{\Gamma}^{(1)}(\boldsymbol{n}) = \frac{1}{2\mu^{(1)}}\boldsymbol{K}_L,$$

and (B.1) becomes

$$\widetilde{\boldsymbol{L}} = +\infty \boldsymbol{J} + 2\left\langle \mu \right\rangle (\boldsymbol{K}_T + \boldsymbol{K}_E) + 2\left\langle \frac{1}{\mu} \right\rangle^{-1} \boldsymbol{K}_L, \tag{B.3}$$

where the orthogonal projectors $\boldsymbol{K}_T$, $\boldsymbol{K}_E$, $\boldsymbol{K}_L$ are introduced in Appendix B.5. Similarly the localization relations (B.2) read, in phase 2, as

$$\left.\begin{aligned} \boldsymbol{\varepsilon}^{(2)} &= \left(\boldsymbol{K}_T + \boldsymbol{K}_E + \mu^{(2)} \left\langle \frac{1}{\mu} \right\rangle \boldsymbol{K}_L \right)^{-1} \overline{\boldsymbol{\varepsilon}}, \\ \boldsymbol{\sigma}_{\mathsf{d}}^{(2)} &= \left[2\mu^{(2)} (\boldsymbol{K}_T + \boldsymbol{K}_E) + 2\left\langle \frac{1}{\mu} \right\rangle^{-1} \boldsymbol{K}_L \right] : \overline{\boldsymbol{\varepsilon}}. \end{aligned}\right\} \tag{B.4}$$

Remark 3. It is worth noticing that the components of $\boldsymbol{A}^{(2)}$ on $\boldsymbol{K}_E$ and $\boldsymbol{K}_T$ are just 1 and this is consistent with the fact that, as result of strain compatibility, the in-plane components of the strain are the same in all phases.

Appendix B.2. Linear viscoelasticity, incompressible phases

In this section each layer r of the laminate is Maxwellian and incompressible (elastic modulus $\mu_{\mathsf{e}}^{(r)}$, relaxation time $\tau^{(r)}$), and therefore the only material constant of the phases is its complex shear modulus

$$\mu^{(r)*}(p) = \mu_{\mathsf{e}}^{(r)} \frac{p}{p + 1/\tau^{(r)}}.$$

The relation (B.3) applies to the complex moduli of the laminate with

$$\left.\begin{aligned} \left\langle \mu^*(p) \right\rangle &= c^{(1)}\mu_{\mathsf{e}}^{(1)} \frac{p}{p + 1/\tau^{(1)}} + c^{(2)}\mu_{\mathsf{e}}^{(2)} \frac{p}{p + 1/\tau^{(2)}}, \\ \left\langle \frac{1}{\mu^*(p)} \right\rangle &= \left\langle \frac{1}{\mu_{\mathsf{e}}} \right\rangle + \frac{1}{p} \left\langle \frac{1}{\mu_{\mathsf{e}}\tau} \right\rangle = \left\langle \frac{1}{\mu_{\mathsf{e}}} \right\rangle \frac{p + 1/\overline{\tau}_L}{p}, \end{aligned}\right\} \tag{B.5}$$

where the longitudinal relaxation time $\overline{\tau}_L$ is defined through the relation

$$\left\langle \frac{1}{\mu_{\mathsf{e}}} \right\rangle \frac{1}{\overline{\tau}_L} = \left\langle \frac{1}{\mu_{\mathsf{e}}\tau} \right\rangle.$$

Substituting (B.5) in (B.3) leads to an expression of the complex moduli of the laminate (for deviatoric strains) in the form (22) with $N = 3$ and:

$$\overline{\boldsymbol{L}}_i = 2\overline{\mu}_i (\boldsymbol{K}_T + \boldsymbol{K}_E), \ \overline{\mu}_i = c^{(i)}\mu_{\mathsf{e}}^{(i)}, \ \overline{\tau}_i = \tau^{(i)} \text{ for } i = 1, 2 \ . \tag{B.6}$$

and

$$\overline{\boldsymbol{L}}_3 = 2\overline{\mu}_3 \boldsymbol{K}_L,\ \overline{\mu}_3 = \left\langle \frac{1}{\mu_{\mathsf{e}}} \right\rangle^{-1},\ \overline{\tau}_3 = \overline{\tau}_L. \tag{B.7}$$

Following section 3.2, three internal variables are introduced, and the overall stress reads in Laplace space,

$$\overline{\boldsymbol{\sigma}}^*(p) = \sum_{i=1}^{3} \overline{\boldsymbol{L}}_i : (\overline{\boldsymbol{\varepsilon}}^*(p) - \boldsymbol{\beta}_i^*(p)) \tag{B.8}$$

with

$$i = 1, 2: \quad \boldsymbol{\beta}_i^*(p) = \frac{1/\overline{\tau}_i}{p + 1/\overline{\tau}_i} (\boldsymbol{K}_T + \boldsymbol{K}_E) : \overline{\boldsymbol{\varepsilon}}^*(p), \quad \boldsymbol{\beta}_3^*(p) = \frac{1/\overline{\tau}_3}{p + 1/\overline{\tau}_3} \boldsymbol{K}_L : \overline{\boldsymbol{\varepsilon}}^*(p), \tag{B.9}$$

or equivalently in time space:

$$\overline{\boldsymbol{\sigma}}(t) = \sum_{i=1}^{3} \overline{\boldsymbol{L}}_i : (\overline{\boldsymbol{\varepsilon}}(t) - \boldsymbol{\beta}_i(t)) \tag{B.10}$$

with

$$i = 1, 2: \quad \overline{\tau}_i \dot{\boldsymbol{\beta}}_i + \boldsymbol{\beta}_i = (\boldsymbol{K}_T + \boldsymbol{K}_E) : \overline{\boldsymbol{\varepsilon}}(t),\ \overline{\tau}_3 \dot{\boldsymbol{\beta}}_3 + \boldsymbol{\beta}_3 = \boldsymbol{K}_L : \overline{\boldsymbol{\varepsilon}}(t). \tag{B.11}$$

Appendix B.3. Number of internal variables and their physical interpretation.

In the general theory (Rice, 1971; Suquet, 1987), the formulation of the effective constitutive relations requires the whole field of viscous strains as internal variables. In the present case, this would mean 2 tensorial variables, the viscous strain $\boldsymbol{\varepsilon}_{\mathsf{v}}$ in each phase, since the viscous strain is uniform in each phase. In other words 10 scalar internal variables are, *a priori*, needed for a two-phase laminate thanks to incompressibility of the viscous strain (5 independent variables per phase).

On the other hand, the constitutive relations (B.10), (B.11) are that of a generalized Maxwell model with 3 branches and therefore 3 relaxation times, namely the relaxation times $\tau^{(1)}$ and $\tau^{(2)}$ of the individual phases and the relaxation time $\overline{\tau}_L$ of longitudinal shear. At first it seems that 3 tensorial internal variables $\boldsymbol{\beta}_i$ are needed in the constitutive relations (B.10). Since these variables are traceless, this would mean 15 scalar unknowns for these internal variables. However, according to the dimension of the spaces $\mathcal{D}_T$, $\mathcal{D}_L$ and $\mathcal{D}_E$ (see (B.17)), the number of independent unknowns in each of these variables is indeed smaller. $\boldsymbol{\beta}_1$ and $\boldsymbol{\beta}_2$ depend on 3 independent unknowns each and $\boldsymbol{\beta}_3$ has 2 independent unknowns, so the total of independent unknowns is actually 8.

These internal variables can be given a physical interpretation. First, $\boldsymbol{\beta}_1$ and $\boldsymbol{\beta}_2$ are the components of the viscous strain along $\boldsymbol{K}_T + \boldsymbol{K}_E$ in each individual phase. Then, it follows from equilibrium that the out-of-plane components of the stress tensor are the same in all phases (see Remark 3). Therefore the longitudinal shear viscous strains in each phase are linked by

$$2\mu_{\mathsf{e}}^{(1)} \tau^{(1)} \boldsymbol{K}_L \dot{\boldsymbol{\varepsilon}}_{\mathsf{v}}^{(1)} = 2\mu_{\mathsf{e}}^{(2)} \tau^{(2)} \boldsymbol{K}_L \dot{\boldsymbol{\varepsilon}}_{\mathsf{v}}^{(2)} = \boldsymbol{K}_L : \overline{\boldsymbol{\sigma}}_{\mathsf{d}} = \overline{\boldsymbol{\sigma}}_3.$$

Therefore only two, out of four, components of the longitudinal viscous strains are needed in

the list of internal variables (hence the reduction in number of internal variables from 10 down to 8). In addition, the internal variable $\boldsymbol{\beta}_3$ is the average of these longitudinal shear viscous strains

$$\boldsymbol{\beta}_3 = \langle \boldsymbol{K}_L : \boldsymbol{\varepsilon}_{\mathsf{v}} \rangle \, .$$

$\boldsymbol{\beta}_3$ is the overall longitudinal viscous strain.

Appendix B.4. Local and effective dissipation. Conjecture (C').

We now consider two possible expressions for the global dissipation, obtained either by averaging the local dissipation or from the effective constitutive relations seen as a rheological model. The local dissipation is uniform per phase and its average over phase r read as

$$d^{(r)} = \boldsymbol{\sigma}^{(r)} : \dot{\boldsymbol{\varepsilon}}_{\mathsf{v}}^{(r)} = \frac{1}{2\mu_{\mathsf{e}}^{(r)}\tau^{(r)}} \boldsymbol{\sigma}_{\mathsf{d}}^{(r)} : \boldsymbol{\sigma}_{\mathsf{d}}^{(r)} \text{ in phase } r, \ \langle d \rangle = \sum_{r=1}^{2} \frac{c^{(r)}}{2\mu_{\mathsf{e}}^{(r)}\tau^{(r)}} \boldsymbol{\sigma}_{\mathsf{d}}^{(r)} : \boldsymbol{\sigma}_{\mathsf{d}}^{(r)}, \tag{B.12}$$

where use has been made of the fact that the stresses $\boldsymbol{\sigma}_{\mathsf{d}}^{(r)}$ are uniform per phase. By virtue of (B.4), they read as

$$\boldsymbol{\sigma}_{\mathsf{d}}^{(2)*}(p) = \left[2\mu_{\mathsf{e}}^{(2)} \frac{p}{p + 1/\tau^{(2)}} \left(\boldsymbol{K}_T + \boldsymbol{K}_E \right) + 2 \left\langle \frac{1}{\mu_{\mathsf{e}}} \right\rangle^{-1} \frac{p}{p + 1/\overline{\tau}_L} \boldsymbol{K}_L \right] : \overline{\boldsymbol{\varepsilon}}^*(p),$$

hence, using the expression of the internal variables (B.9) and the expressions of $\overline{\mu}_i$ and $\overline{\tau}_i$ given in (B.6) and (B.7),

$$\boldsymbol{\sigma}_{\mathsf{d}}^{(2)}(t) = \frac{2\overline{\mu}_2}{c^{(2)}} \, \overline{\tau}_2 \, \dot{\boldsymbol{\beta}}_2(t) + 2\overline{\mu}_3 \, \overline{\tau}_3 \, \dot{\boldsymbol{\beta}}_3(t). \tag{B.13}$$

A relation similar to (B.13) holds for $\boldsymbol{\sigma}_{\mathsf{d}}^{(1)}(t)$, replacing the phase index (2) by (1). The average dissipation reads as

$$\langle d \rangle = 2\overline{\mu}_1\overline{\tau}_1 \dot{\boldsymbol{\beta}}_1(t) : \dot{\boldsymbol{\beta}}_1(t) + 2\overline{\mu}_2\overline{\tau}_2 \dot{\boldsymbol{\beta}}_2(t) : \dot{\boldsymbol{\beta}}_2(t) + 2\overline{\mu}_3\overline{\tau}_3 \dot{\boldsymbol{\beta}}_3(t) : \dot{\boldsymbol{\beta}}_3(t). \tag{B.14}$$

Note that $\boldsymbol{\beta}_i : \boldsymbol{\beta}_3 = 0, i = 1, 2$. Making use of the differential equations (A.4) for the internal variables, it is found that the dissipation (45) of the rheological model coincides with the average dissipation (B.14). Hence conjecture (C') is proved in that case.

Appendix B.5. Elements of algebra for transversely isotropic and incompressible materials

For incompressible materials, the part of the stiffness tensor acting on pure deviators depends only on three independent coefficients

$$\boldsymbol{L} = \delta \boldsymbol{K}_T + \delta' \boldsymbol{K}_L + \delta'' \boldsymbol{K}_E. \tag{B.15}$$

In addition, the five-dimensional space of symmetric second-order deviators $\mathcal{D}$ can be decomposed into purely transverse, purely longitudinal and mixed deviators using the orthogonal projectors $\boldsymbol{K}_T$, $\boldsymbol{K}_L$ and $\boldsymbol{K}_E$

$$\mathcal{D} = \mathcal{D}_T \bigoplus \mathcal{D}_L \bigoplus \mathcal{D}, \quad \mathcal{D}_T = \boldsymbol{K}_T(\mathcal{D}), \quad \mathcal{D}_L = \boldsymbol{K}_L(\mathcal{D}), \quad \mathcal{D}_E = \boldsymbol{K}_E(\mathcal{D}). \tag{B.16}$$

For clarity, consider that the direction normal to the layers is $\boldsymbol{n} = \boldsymbol{e}_3$. Then for any symmetric second-order tensor $\boldsymbol{a}$

$$\left.\begin{aligned} \boldsymbol{K}_T.\boldsymbol{a} &= \frac{a_{11}-a_{22}}{2}\left(\boldsymbol{e}_1\otimes\boldsymbol{e}_1 - \boldsymbol{e}_2\otimes\boldsymbol{e}_2\right) + a_{12}\left(\boldsymbol{e}_1\otimes\boldsymbol{e}_2 + \boldsymbol{e}_2\otimes\boldsymbol{e}_1\right), \\ \boldsymbol{K}_L.\boldsymbol{a} &= a_{13}\left(\boldsymbol{e}_1\otimes\boldsymbol{e}_3 + \boldsymbol{e}_3\otimes\boldsymbol{e}_1\right) + a_{23}\left(\boldsymbol{e}_2\otimes\boldsymbol{e}_3 + \boldsymbol{e}_3\otimes\boldsymbol{e}_2\right), \\ \boldsymbol{K}_E.\boldsymbol{a} &= \frac{a_{11}+a_{22}-2a_{33}}{6}\left(\boldsymbol{e}_1\otimes\boldsymbol{e}_1 + \boldsymbol{e}_2\otimes\boldsymbol{e}_2 - 2\boldsymbol{e}_3\otimes\boldsymbol{e}_3\right). \end{aligned}\right\} \tag{B.17}$$

It follows from (B.17) that $\mathcal{D}_T$, $\mathcal{D}_L$ and $\mathcal{D}_E$ are orthogonal vector spaces of dimension 2, 2 and 1 respectively.

Remark 4. The present study of laminates with incompressible phases can be extended in two directions. First, laminates with compressible phases can be addressed using a similar approach. However in that case the effective tensor $\widetilde{\boldsymbol{L}}$ cannot be expressed with orthogonal projectors, as was the case for incompressible phases by means of the projectors $\boldsymbol{K}_E$, $\boldsymbol{K}_T$ and $\boldsymbol{K}_L$. As a result, the final expressions are much more involved and could only be handled using a symbolic computation software. The details will not be discussed here, suffices to say that it has been checked that the conjecture (C') holds true for compressible phases as well.

Second, rank-two laminates obtained by double lamination have also been examined, again using a symbolic computation software. Here again the validity of conjecture (C') was checked. It should be noted that this rank-two laminate which is a two-phase composite may also be considered as a three-phase composite, two of the phases having the same material data, since the phase which is used in both steps of the lamination is present at two different scales. The fields are uniform in each of the three phases but differ in the phases with the same material data. Therefore the conjecture is also valid in a composite obtained from two successive laminations with field fluctuations in one of the phases. By induction (although we did not go beyond rank-2 laminates) the conjecture should be valid for multi-laminated microstructures and therefore for all microstructures attaining the Hashin-Shtrikman bounds, compressible or not.

Appendix C. Equivalence between two viscoelastic models

It is well known that different assemblages of springs and dashpots can lead to the same mechanical response under general loadings. In particular two equivalent representations of the effective viscoelastic properties for composites (15) and (18) have been given in section 3. Similarly, when reducing the number of internal variables to a finite number (section 3.2), two different Prony series (22) and (24) can be used in one formulation or the other and the aim of this section is to investigate the relation between these two series. The starting point is the constitutive relation for a generalized Maxwell model

$$\overline{\boldsymbol{\sigma}} = \sum_{i=1}^{N} \overline{\boldsymbol{L}}_i : (\overline{\boldsymbol{\varepsilon}} - \boldsymbol{\beta}_i), \quad \overline{\tau}_i\dot{\boldsymbol{\beta}}_i + \boldsymbol{\beta}_i = \overline{\boldsymbol{\varepsilon}}, \tag{C.1}$$

where for simplicity the relaxation times $\overline{\tau}_i$ are assumed to be all different and for definiteness they are ordered $\overline{\tau}_1 < \overline{\tau}_2 \leq ... \leq \overline{\tau}_N$. It may happen that $\overline{\tau}_N = +\infty$ in which case the generalized Maxwell model under consideration contains a purely elastic element with no internal

variable $\boldsymbol{\beta}_N$, a situation which corresponds physically to a material exhibiting a residual elasticity under creep. This is typically the case for particulate composites under hydrostatic stress (section Appendix C.4).

Appendix C.1. Poles and zeroes of the relaxation function

In many cases of interest, the material tensors relevant to the problem at hand can be decomposed on projectors on orthogonal spaces

$$\overline{\boldsymbol{L}}_i = \sum_{p=1} \ell_p \boldsymbol{\Pi}_p, \quad \boldsymbol{\Pi}_p : \boldsymbol{\Pi}_q = \delta_{pq} \boldsymbol{\Pi}_p, \quad \mathcal{E}_p = \boldsymbol{\Pi}_p \mathcal{E}, \quad \mathcal{E} = \oplus \mathcal{E}_p, \tag{C.2}$$

where $\mathcal{E}$ is the space of second-order symmetric tensors of interest for the problem at hand (it can be the full space of symmetric second-order tensors on $\mathbb{R}^d$, or only the subspace of purely deviatoric tensors etc...). A typical example is a composite composed of isotropic phases with overall isotropy. Then the elasticity and viscosity tensors of the phases $\boldsymbol{L}_{\mathsf{e}}^{(r)}$ and $\boldsymbol{L}_{\mathsf{v}}^{(r)}$, as well as the effective relaxation $\widetilde{\boldsymbol{L}}$ can be decomposed on the two projectors $\boldsymbol{J}$ and $\boldsymbol{K}$ (projecting on the spaces $\mathcal{E}_1$ and $\mathcal{E}_2$ of purely spherical and purely deviatoric symmetric second-order tensors respectively).

Thanks to the orthogonality between the subspaces $\mathcal{E}_p$, the inversion of (C.1) can be performed in each subspace at a time. Restricting attention to one subspace, the $\overline{L}_i$'s can be assumed to be scalar (and denoted as $\overline{\ell}_i$ from now on), whereas $\boldsymbol{\sigma}, \boldsymbol{\varepsilon}, \boldsymbol{\beta}$ are second-order tensors belonging to the subspace under consideration (either purely deviatoric or purely hydrostatic tensors when this is the relevant decomposition in the problem at hand).

Taking the Laplace-Carson transform of the differential equations for the internal variables yields

$$\boldsymbol{\beta}_i^*(p) = \frac{1}{\overline{\tau}_i p + 1}, \overline{\boldsymbol{\varepsilon}}^*(p), \quad \text{hence} \quad \overline{\boldsymbol{\sigma}}^*(p) = \widetilde{\ell}^*(p) \overline{\boldsymbol{\varepsilon}}^*(p), \tag{C.3}$$

with

$$\widetilde{\ell}^*(p) = \sum_{i=1}^{N} \overline{\ell}_i \frac{p}{p + \frac{1}{\overline{\tau}_i}} = \frac{p \sum_{i=1}^{N} \overline{\ell}_i \prod_{j=1, j\neq i}^{N} \left(p + \frac{1}{\overline{\tau}_j} \right)}{\prod_{j=1}^{N} \left(p + \frac{1}{\overline{\tau}_j} \right)}. \tag{C.4}$$

The following limits are easily derived and help to understand the dependence on p of $\widetilde{\ell}^*(p)$ on

the real axis,

$$\left.\begin{array}{rl}
\text{when } p \to -\infty & \lim \widetilde{\ell}^*(p) = \displaystyle\sum_{i=1}^{N} \overline{\ell}_i > 0, \\
\text{when } p \to (-1/\overline{\tau}_j)^- & \widetilde{\ell}^*(p) \sim \overline{\ell}_j \dfrac{p}{p + \frac{1}{\overline{\tau}_j}} \to +\infty, \quad j = 1, 2, ..., N \\
\text{when } p \to (-1/\overline{\tau}_j)^+ & \widetilde{\ell}^*(p) \sim \overline{\ell}_j \dfrac{p}{p + \frac{1}{\overline{\tau}_j}} \to -\infty, \quad j = 1, 2, ..., N \\
\text{when } p = 0 & \widetilde{\ell}^*(p) = 0, \\
\text{when } p \to +\infty & \lim \widetilde{\ell}^*(p) = \displaystyle\sum_{i=1}^{N} \overline{\ell}_i > 0,
\end{array}\right\} \tag{C.5}$$

where the superscripts $.^-$ and $.^+$ denote limits from below and above respectively. It follows from this limit that the numerator of the rational fraction of the left-hand-side of (C.4), which is a polynom of degree N, has $N-1$ roots located in the intervals $]-\frac{1}{\overline{\tau}_{j+1}}, -\frac{1}{\overline{\tau}_j}[$, $j = 1, .., N-1$ and one root at $p = 0$ and since this polynom has only N roots in $\mathbb{C}$ all these roots are located on the negative real axis (including 0). Moreover the zeroes of $\widetilde{\ell}^*(p)$ alternate with its poles.

As an example the variations of $\widetilde{k}^*(p)$ and $\widetilde{\mu}^*(p)$ on the real (negative) axis are shown in figure C.6 for an isotropic particulate two-phase composite, using relations (35). The material data are that of the example of Figure 3. The alternance between the poles and the zeroes is evidenced. As expected, the shear complex modulus displays 3 poles and 3 zeroes ($N = 3$). Since the viscous strain is purely deviatoric in this example, the bulk modulus has only a single pole and a single zero.

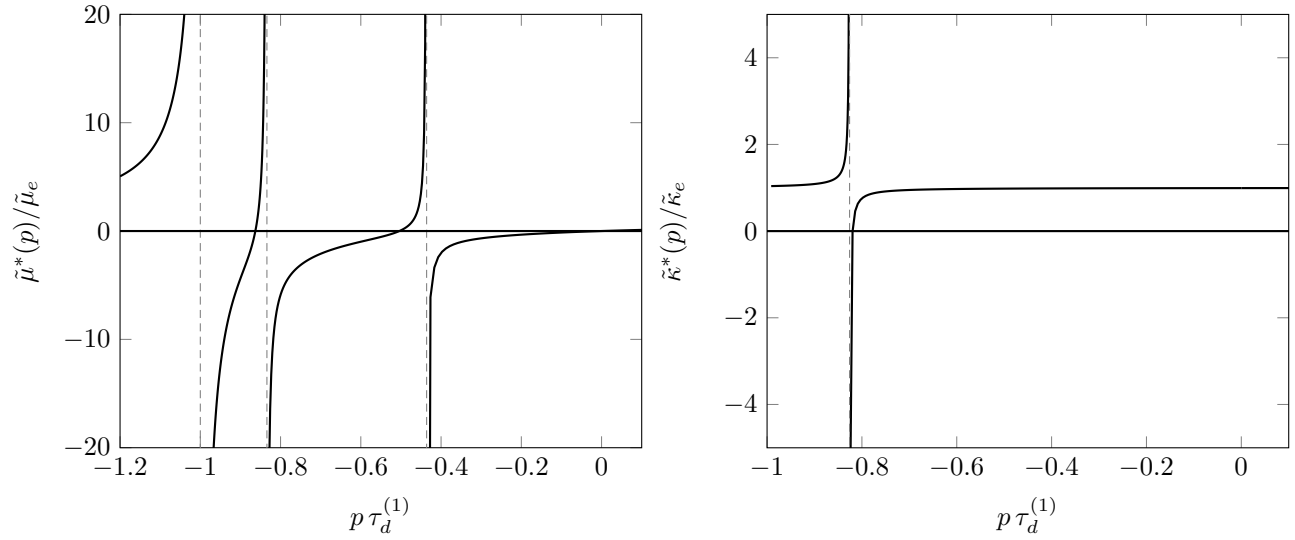

Figure C.6: Two-phase particulate composite with overall isotropy. Laplace Carson transforms of the shear modulus $\tilde{\mu}^*(p)$ (left) and bulk modulus $\tilde{\kappa}^*(p)$(right). Same material data as in Figure 3 with $c^{(2)} = 10\%$ (see table 1). These moduli are normalized by their values in the purely elastic regime ($p \to +\infty$) while $\tau_{\mathsf{d}}^{(1)}$ denotes the relaxation time in shear of the matrix.

Upon inversion (denoting by $\widetilde{m}^*$ the inverse of $\widetilde{\ell}^*$), one gets

$$\widetilde{m}^*(p) = \frac{1}{p} \frac{\displaystyle\prod_{j=1}^{N} \left(p + \frac{1}{\overline{\tau}_j}\right)}{\displaystyle\sum_{i=1}^{N} \overline{\ell}_i \prod_{j=1, j\neq i}^{N} \left(p + \frac{1}{\overline{\tau}_j}\right)}. \tag{C.6}$$

The poles of $\widetilde{m}^*(p)$ are the zeroes of $\widetilde{\ell}^*(p)$ and conversely the poles of $\widetilde{\ell}^*(p)$ are the zeroes of $\widetilde{m}^*(p)$. Therefore $\widetilde{m}^*(p)$ has N poles, one being $p = 0$ and the $N-1$ other ones being the zeroes of $\widetilde{\ell}^*(p)$ else than 0 and correspond to the $N-1$ retardation times entering (24). Being the zeroes of the denominator in (C.6) they are such that

$$\sum_{i=1}^{N} \overline{\ell}_i \prod_{j=1, j\neq i}^{N} \left(-\frac{1}{\hat{\tau}_k} + \frac{1}{\overline{\tau}_j}\right) = 0, \quad k = 1, ..., N-1, \tag{C.7}$$

or alternatively

$$\sum_{i=1}^{N} \overline{\ell}_i \frac{\overline{\tau}_i}{\hat{\tau}_k - \overline{\tau}_i} = 0, \quad k = 1, ..., N-1. \tag{C.8}$$

The two terms of highest and lowest degree (N and 0 respectively) in this polynom are respectively

$$\sum_{i=1}^{N} \overline{\ell}_i \quad \text{and} \quad \frac{\displaystyle\sum_{i=1}^{N} \overline{\ell}_i \overline{\tau}_i}{\displaystyle\prod_{j=1}^{N} \overline{\tau}_j},$$

and therefore the denominator of the second rational fraction in (C.6) reads as

$$\left(\sum_{i=1}^{N} \overline{\ell}_i\right) \prod_{k=1}^{N-1} \left(p + \frac{1}{\hat{\tau}_k}\right) \quad \text{with} \quad \frac{\displaystyle\sum_{i=1}^{N} \overline{\ell}_i \overline{\tau}_i}{\displaystyle\prod_{j=1}^{N} \overline{\tau}_j} = \frac{\displaystyle\sum_{i=1}^{N} \overline{\ell}_i}{\displaystyle\prod_{k=1}^{N-1} \hat{\tau}_k}. \tag{C.9}$$

So far no distinction has been made as of whether $\overline{\tau}_N$ is finite or not, since the above relations have a finite limit in both cases. We now make this distinction.

Appendix C.2. No residual elasticity: $\overline{\tau}_N < +\infty$.

Introducing the following notations

$$\tilde{\ell}_{\mathsf{e}} = \sum_{i=1}^{N} \overline{\ell}_i, \quad \tilde{\ell}_{\mathsf{v}} = \sum_{i=1}^{N} \overline{\ell}_i \overline{\tau}_i, \quad \tilde{\tau} = \frac{\tilde{\ell}_{\mathsf{v}}}{\tilde{\ell}_{\mathsf{e}}}, \quad \tilde{m}_{\mathsf{e}} = \tilde{\ell}_{\mathsf{e}}^{-1}, \quad \tilde{m}_{\mathsf{v}} = \tilde{\ell}_{\mathsf{v}}^{-1}, \tag{C.10}$$

it is noticed in particular that

$$\prod_{j=1}^{N} \overline{\tau}_j = \tilde{\tau} \prod_{k=1}^{N-1} \hat{\tau}_k. \tag{C.11}$$

The rational fraction (C.6) can now be decomposed as

$$\widetilde{m}^*(p) = \tilde{m}_e + \frac{1}{p}\tilde{m}_{\mathsf{v}} + \sum_{k=1}^{N-1} \frac{\hat{m}_k}{p + \frac{1}{\hat{\tau}_k}}, \tag{C.12}$$

where

$$\hat{m}_k = -\frac{\tilde{m}_{\mathsf{v}}}{\hat{\tau}_k^2} \frac{\displaystyle\prod_{j=1}^{N} (\hat{\tau}_k - \overline{\tau}_j)}{\displaystyle\prod_{\ell=1,\ell\neq k}^{N-1} (\hat{\tau}_k - \hat{\tau}_\ell)}. \tag{C.13}$$

Appendix C.3. Residual elasticity: $\overline{\tau}_N = +\infty$.

Taking the limit of (C.10) as τ_N tends to $+\infty$ yields

$$\ell_{\mathsf{v}} \sim \overline{\ell}_N \overline{\tau}_N, \quad \widetilde{\tau} \sim \frac{\overline{\ell}_N}{\widetilde{\ell}_{\mathsf{e}}} \overline{\tau}_N, \quad \prod_{j=1}^{N} \overline{\tau}_j \sim \overline{\tau}_N \prod_{j=1}^{N-1} \overline{\tau}_j, \quad \widetilde{m}_{\mathsf{v}} \sim \frac{1}{\overline{\ell}_N \overline{\tau}_N}, \tag{C.14}$$

and therefore the limits as τ_N tends to $+\infty$ of (C.12), (C.11) and (C.13) read respectively

$$\widetilde{m}^*(p) = \tilde{m}_e + \sum_{k=1}^{N-1} \frac{\hat{m}_k}{p + \frac{1}{\hat{\tau}_k}}, \ \prod_{k=1}^{N-1} \hat{\tau}_k = \frac{\widetilde{\ell}_{\mathsf{e}}}{\overline{\ell}_N} \prod_{j=1}^{N-1} \overline{\tau}_j, \ \hat{m}_k = \frac{\overline{m}_N}{\hat{\tau}_k^2} \frac{\displaystyle\prod_{j=1}^{N-1} (\hat{\tau}_k - \overline{\tau}_j)}{\displaystyle\prod_{\ell=1,\ell\neq k}^{N-1} (\hat{\tau}_k - \hat{\tau}_\ell)}. \tag{C.15}$$

Appendix C.4. Elastic particles in a viscoelastic matrix under hydrostatic stress

As a typical example, the relations (C.15) can be applied to composites made of elastic particles in a Maxwellian matrix, where the viscous strains are purely deviatoric. Let us consider only the hydrostatic response, therefore $\boldsymbol{\Pi} = \boldsymbol{J}$ in (C.2), and the corresponding residues $\overline{\ell}_i$ are denoted as $\overline{k}_i$. One can pass to the limit in (36) as $\tau_{\mathsf{m}}^{(1)}$ and $\tau_{\mathsf{m}}^{(2)}$ tend to $+\infty$. Then (36) has two roots, corresponding to two relaxation times,

$$\overline{\tau}_{1,\mathsf{m}} = \tau_{\mathsf{d}}^{(1)} \frac{k^{(1)} + \frac{4}{3}\mu_{\mathsf{e}}^{(1)} + c^{(1)}\Delta k}{k^{(1)} + c^{(1)}\Delta k}, \quad \overline{\tau}_{2,\mathsf{m}} = +\infty,$$

and the corresponding residues in the expansion in rational fractions of the effective complex bulk modulus are

$$\overline{k}_1 = \frac{4c^{(1)}c^{(2)}\mu_e^{(1)}(\Delta k)^2}{3(k^{(1)} + \frac{4}{3}\mu_{\mathsf{e}}^{(1)} + c^{(1)}\Delta k)(k^{(1)} + c^{(1)}\Delta k)},\ \overline{k}_2 = \frac{k^{(1)}k^{(2)}}{c^{(1)}k^{(2)} + c^{(2)}k^{(1)}} = \left\langle \frac{1}{k} \right\rangle^{-1},\ \Delta k = k^{(2)} - k^{(1)}, \tag{C.16}$$

with $\widetilde{k}_{\mathsf{e}} = \overline{k}_1 + \overline{k}_2$. The relation (C.15) between the relaxation and the retardation times becomes $\hat{\tau}_{1,\mathsf{m}} = \overline{\tau}_{1,\mathsf{m}}(\overline{k}_1 + \overline{k}_2)/\overline{k}_2$. After some straightforward algebra, and noting the relations

$$\langle k \rangle - \left\langle \frac{1}{k} \right\rangle^{-1} = \frac{c^{(1)}c^{(2)}(\Delta k)^2}{k^{(1)} + c^{(1)}\Delta k}, \quad \left\langle \frac{1}{k} \right\rangle^{-1} = \frac{k^{(1)}k^{(2)}}{k^{(1)} + c^{(1)}\Delta k},$$

it is found that

$$\hat{\tau}_{1,\mathsf{m}} = 1 + \frac{4}{3}\mu_{\mathsf{e}}^{(1)} \frac{\langle k \rangle}{k^{(1)}k^{(2)}},$$

which coincides with the retardation time found in Masson et al. (2020) (appendix A, denoted $\widetilde{\tau}_1^m$). Similarly the general relation (C.15) for $\hat{m}_k$ becomes in the present context

$$\hat{m}_1 = \frac{1}{3\overline{k}_2} \frac{\hat{\tau}_{1,\mathsf{m}} - \overline{\tau}_{1,\mathsf{m}}}{\hat{\tau}_1^2}.$$

After some straightforward algebra it is found that

$$\hat{m}_1 = \frac{4c^{(1)}c^{(2)}\mu_{\mathsf{e}}^{(1)}}{9\hat{\tau}_1^2} \left(\frac{\Delta k}{k^{(1)}k^{(2)}} \right)^2,$$

which again coincides with the expression given in Masson et al. (2020) (Appendix, relation (A.6)).