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Effective properties of linear viscoelastic heterogeneous media: Internal variables formulation and extension to ageing behaviours

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ABSTRACT

The Laplace–Carson transform classically used for homogenization of linear viscoelastic heterogeneous media yields integral formulations of effective behaviours. These are far less convenient than internal variables formulations with respect to computational aspects as well as to theoretical extensions to closely related problems such as ageing viscoelasticity. Noticing that the collocation method is usually adopted to invert the Laplace–Carson transforms, we first remark that this approximation is equivalent to an internal variables formulation which is exact in some specific situations. This result is illustrated for a two-phase composite with phases obeying a compressible maxwellian behaviour. Next, an incremental formulation allows to extend at each time step the previous general framework to ageing viscoelasticity. Finally, with the help of a creep test of a porous viscoelastic matrix reinforced with elastic inclusions, it is shown that the method yields accurate predictions (comparing to reference results provided by periodic cell finite element computations).

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1. Introduction

To derive the effective behaviour of non-ageing linear viscoelastic heterogeneous media, the Laplace–Carson transform is classically used (the so called correspondence principle, Mandel, 1966). This functional transform allows to define a symbolic linear elastic composite in the Laplace domain. Linear homogenization models are then applied to this fictitious elastic body to derive its effective properties. The viscoelastic effective properties (for instance the effective relaxation function) are then deduced by the inversion of the Laplace–Carson transform.

This methodology has been successfully applied in various situations: first work (see for instance, Christensen, 1969) has extended the composite sphere assemblage model (Hashin, 1962) to linear viscoelasticity. Afterwards, extension has been achieved for various approaches from linear elasticity to linear viscoelasticity. For instance, the self-consistent (Kröner, 1958) and the Mori and Tanaka (1973) models have been extended by Laws and McLaughlin (1978) and Wang and Weng (1992) (see also, Brinson and Lin, 1998), respectively. However, apart from some particular cases (see for instance, Rougier et al., 1993), the inversion of the Laplace–Carson transform is usually performed numerically (see the collocation method, Schapery, 1962). In addition, this method leads most of the time to integral equations over the whole loading path even if the different phases of the heterogeneous composite exhibit limited memory effects. This last point makes difficult direct extensions to more general situations manifested by a change of viscoelastic properties as a function of time: thermomechanical loading, ageing viscoelasticity.

To overcome these limitations, a new theory based on internal variables has been recently proposed by Lahellec and Suquet (2007). This method is able to address a large class of microstructures. It directly operates in the time domain by reducing the resolution of the discretized evolution equations to the minimization of an incremental energy function. This theory leads to an implicit system of non-linear equations at each time step. The present work aims also at deriving an internal variables formulation but in a different manner. Here, we remain consistent with the classical methodology (the Laplace–Carson transform and the correspondence principle) but we propose to take advantage of the usual Prony–Dirichlet series expansion of the effective properties (the collocation method) to reduce the general integral equations to an equivalent internal variables formulation. In contrast with the theory of Lahellec and Suquet (2007), the interest of the method proposed hereafter is twofold: first, it leads to an implicit system of linear equations which is significantly easier to solve at each time step. Second, the formulation turns out to be exact within the case of very useful inclusion–matrix types of composites.

The paper is organised as follows. In Section 2, the collocation method and the equivalency between a Prony–Dirichlet series

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expansion and an internal variables formulation are first recalled. Then, particular cases are studied to yield exact results for two-phase random macroscopically isotropic composites. This internal variables formulation being equivalent to the classical one, these last results are not fundamentally new. However, this formulation allows straightforward extensions to more general problem like ageing viscoelasticity. This last point is illustrated in Sections 3 and 4: the equivalent internal variables formulation is used to directly model the densification of a porous viscoelastic matrix reinforced by elastic inclusions.

2. Internal variables formulation of non-ageing linear viscoelastic heterogeneous media

2.1. A preliminary remark: from the collocation method to an internal variables formulation

Here, we consider a non-ageing linear viscoelastic material which is subjected to a given strain path defined by the function of time $\epsilon(\tau)$ $(0 \leqslant \tau \leqslant t)$. Its stress response is denoted by the function of time $\sigma(t)$ and reads (see, Mandel, 1966):

$$\sigma(t) = \int_0^t E(t-u)\mathrm{d}\epsilon(u) + E(t)\epsilon(0), \tag{1}$$

where the function of time $E(t)$ corresponds to the relaxation function of the material: $E(t-t')$ denotes the stress at time t caused by a unit constant strain applied at time $t' \leqslant t$. $\epsilon(t)$ and $\sigma(t)$ are assumed to be scalar in this section to shorten notations.

2.1.1. Expansion of the relaxation function as Prony series

The relaxation function $E(t)$ is expanded as Prony series as follows:

$$E(t) = \sum_{i=1}^{N_p} E_{\tau_i} \mathrm{e}^{-\frac{t}{\tau_i}}, \tag{2}$$

where τ_i denotes N_p relaxation times. If E^e is the elastic (instantaneous) modulus of the considered material (sometimes referred to as E_0), it is worth mentioning that the N_p scalar coefficients E_{τ_i} of the previous expansion satisfy: $\sum_{i=1}^{N_p} E_{\tau_i} = E^e$.

In some particular cases (see Section 2.2.3), this expansion will turn out to be exact with a finite (and rather small) number N_p of terms. In general, this expansion will represent an approximation of the relaxation function (the collocation method), namely:

$$E(t) \approx E_a(t) = \sum_{i=1}^{N_p^a} E_{\tau_i} \mathrm{e}^{-\frac{t}{\tau_i}},$$

where the N_p^a relaxation times τ_i are chosen arbitrarily. If the error due to this approximation is measured by

$$\int_0^{+\infty} (E(t) - E_a(t))^2 dt,$$

the optimal values of the coefficients E_{τ_i} are given by (see, Schapery, 1962):

$$1 \leqslant i \leqslant N_p^a \quad E^*\left(\frac{1}{\tau_i}\right) = E_a^*\left(\frac{1}{\tau_i}\right),$$

where $f^*(p) = p\int_0^{+\infty} f(t)\mathrm{e}^{-pt}\mathrm{d}t$ denotes the Laplace–Carson transform of any function of time $f(t)$ with respect to the parameter p.

2.1.2. An equivalent internal variables formulation

As the number of terms in the Prony series expansion (2) is finite, the integral strain–stress relation (1) could be expressed as a constitutive relation with a finite number of internal variables (Mandel, 1966). These N_p internal variables, denoted as $\alpha_{\tau_i}(t)$ hereafter $(1 \leqslant i \leqslant N_p)$, are solutions of the N_p first order linear differential equations (3) with the characteristic times τ_i ranging from τ_1 to τ_{N_p}:

$$1 \leqslant i \leqslant N_p, \quad \dot{\alpha}(t) + \frac{1}{\tau_i}\alpha(t) = \frac{1}{\tau_i}\frac{E_{\tau_i}}{E^e}\epsilon(t) \quad \text{with } \alpha(0) = 0. \tag{3}$$

Since:

$$1 \leqslant i \leqslant N_p, \quad \alpha_{\tau_i}(t) = \frac{E_{\tau_i}}{\tau_i E^e}\mathrm{e}^{-\frac{t}{\tau_i}}\int_0^t \mathrm{e}^{\frac{u}{\tau_i}}\epsilon(u)du$$

and:

$$1 \leqslant i \leqslant N_p, \quad \int_0^t \mathrm{e}^{-\frac{(t-u)}{\tau_i}}\mathrm{d}\epsilon(u) = \epsilon(t) - \frac{1}{\tau_i}\mathrm{e}^{-\frac{t}{\tau_i}}\int_0^t \mathrm{e}^{\frac{u}{\tau_i}}\epsilon(u)\mathrm{d}u - \epsilon(0)\mathrm{e}^{-\frac{t}{\tau_i}},$$

the constitutive law (1) reads:

$$\sigma(t) = E^e \quad \epsilon(t) - \sum_{i=1}^{N_p}\alpha_{\tau_i}(t)\Bigg). \tag{4}$$

As a result, the expansion (2) allows to reduce the integral expression (1) of the viscoelastic constitutive law to the constitutive relation (4) with the N_p internal variables α_{τ_i}. These internal variables are viscous strains. Their sum gives the viscous strain of the material. Finally, it is worth emphasizing that the evolution with time of the N_p internal variables during the relaxation test $\epsilon(t) = \epsilon^0,\ t > 0^+$ reads:

$$i = 1\ldots N_p, \quad t > 0^+, \quad \alpha_{\tau_i}(t) = \frac{E_{\tau_i}}{E^e}\epsilon^0(1 - \mathrm{e}^{-\frac{t}{\tau_i}}). \tag{5}$$

As a result, the ratios $\frac{E_{\tau_i}}{E^e}$ represent the relative magnitude of each internal variable (α_{τ_i}). These ratios satisfy $0 \leqslant \frac{E_{\tau_i}}{E^e} \leqslant 1$.

2.2. Application to two-phase composites

In this section, we consider a viscoelastic micro-structurally random composite material. This composite is made of two perfectly bonded phases which obey an isotropic compressible maxwellian behaviour:

$$\dot{\varepsilon}_m = \frac{1}{3k^e}\dot{\sigma}_m + \frac{1}{3k_i^v}\sigma_m, \quad \dot{\boldsymbol{e}} = \frac{1}{2\mu^e}\dot{\boldsymbol{s}} + \frac{1}{2\mu_i^v}\boldsymbol{s},$$
$$\text{for each phase } i = 1, 2, \tag{6}$$

where $\boldsymbol{e}$, $\boldsymbol{s}$ are the strain and stress deviators, ε_m and σ_m the respective spherical-tensor parts. Bulk and shear moduli are, respectively, denoted by k and μ. Upper scripts e and v denote, respectively, the homogeneous (for the sake of simplicity) elastic and the heterogeneous viscous behaviour for each phase (i) whose volume fraction is denoted by c_i.

2.2.1. Overall behaviour

A representative volume element (RVE) of the considered composite is subjected to a known macroscopic history of strain $\overline{\boldsymbol{\varepsilon}}(t)$ from time $t = 0$. $\overline{\boldsymbol{\sigma}}(t)$ denotes hereafter the overall stress response of this RVE. If we now consider an isotropic material, the relaxation function depends on the two scalar functions $(\tilde{\mu}(t), \tilde{k}(t))$, namely:

$$\overline{\boldsymbol{\sigma}}(t) = 3\int_0^t \tilde{k}(t-u)\mathrm{d}\overline{\varepsilon}_m(u)\boldsymbol{\delta} + 2\int_0^t \tilde{\mu}(t-u)\mathrm{d}\overline{\boldsymbol{e}}(u), \tag{7}$$

where $\boldsymbol{\delta}$ is the second-order identity tensor and $\overline{\varepsilon}_m = \frac{1}{3}\boldsymbol{\delta} : \overline{\boldsymbol{\varepsilon}}$. Following Section 2.1.1, these two relaxation functions expand as follows:

$$\tilde{\mu}(t) = \sum_{i=1}^{N_p^d}\mu_{\overline{\tau}_i^d}\mathrm{e}^{-\frac{t}{\overline{\tau}_i^d}}, \quad \tilde{k}(t) = \sum_{i=1}^{N_p^m}k_{\overline{\tau}_i^m}\mathrm{e}^{-\frac{t}{\overline{\tau}_i^m}}, \tag{8}$$

with the two following constraints:

$$\sum_{i=1}^{N_p^d} \mu_{\overline{\tau}_i^d} = \mu^e, \quad \sum_{i=1}^{N_p^m} k_{\overline{\tau}_i^m} = k^e. \tag{9}$$

These expansions are proved to be exact for the microstructures considered in Section 2.2.3. In general, they represent approximations of the relaxation functions as explained in Section 2.1. Substituting $(\epsilon(t), E(t), \sigma(t))$ by $(\overline{\boldsymbol{e}}(t), \tilde{\mu}(t), \overline{\boldsymbol{s}}(t))$ and afterwards by $(\overline{\varepsilon}_m(t), \tilde{k}(t), \overline{\sigma}_m(t))$, we apply the results derived in Section 2.1 to yield:

$$\overline{\boldsymbol{s}}(t) = 2\mu^e \left(\overline{\boldsymbol{e}}(t) - \sum_{i=1}^{N_p^d} \boldsymbol{\alpha}_{\overline{\tau}_i^d}^d(t) \right),$$
$$\overline{\sigma}_m(t) = 3k^e \left(\overline{\varepsilon}_m(t) - \sum_{i=1}^{N_p^m} \alpha_{\overline{\tau}_i^m}^m(t) \right), \tag{10}$$

where the internal variables $\alpha_{\overline{\tau}_i^m}^m(t)$ and $\boldsymbol{\alpha}_{\overline{\tau}_i^d}^d(t)$ are, respectively, scalar and (second-order) tensorial functions of time which satisfy the following evolution laws:

$$\dot{\alpha}(t) + \frac{1}{\overline{\tau}_i^m}\alpha(t) = \frac{1}{\overline{\tau}_i^m}\frac{k_{\overline{\tau}_i^m}}{k^e}\overline{\varepsilon}_m(t) \quad 1 \leqslant i \leqslant N_p^m,$$
$$\dot{\boldsymbol{\alpha}}(t) + \frac{1}{\overline{\tau}_i^d}\boldsymbol{\alpha}(t) = \frac{1}{\overline{\tau}_i^d}\frac{\mu_{\overline{\tau}_i^d}}{\mu^e}\overline{\boldsymbol{e}}(t) \quad 1 \leqslant i \leqslant N_p^d. \tag{11}$$

In that isotropic case, the viscoelastic effective response reduces to an incremental formulation with $N_p^m + 5N_p^d$ scalar internal variables at the utmost.

2.2.2. Evolution laws of the averaged stresses per phase

Here, we take advantage of the number of phases (two phases) to express the stress average over the phase (1) as an explicit expression of the overall stress and strain, and their derivatives (x_i denotes the average over the phase (i) of the field x in the sequel).

First, the application of the Laplace–Carson transform to the average over phase (i = 1,2) of the constitutive relations (6) yields:

$$\sigma_{im}^*(p) = 3k_i^*(p)\varepsilon_{im}^*(p), \quad \boldsymbol{s}_i^*(p) = 2\mu_i^*\boldsymbol{e}_i^*(p), \tag{12}$$

p being the Laplace–Carson parameter and $(\mu_i^*(p), k_i^*(p))$ corresponding to the symbolic linear elastic moduli of any phase (i):

$$k_i^*(p) = \frac{1}{\frac{1}{k^e} + \frac{1}{pk_i^v}}, \quad \mu_i^*(p) = \frac{1}{\frac{1}{\mu^e} + \frac{1}{p\mu_i^v}}. \tag{13}$$

For a given real value of p, we now consider the heterogeneous composite whose microstructure is the same as the previous one but with phases obeying the linear elastic behaviour defined by the relations (12). Within this fictitious heterogeneous composite, the two following relations hold:

$$c_1\boldsymbol{s}_1^*(p) + c_2\boldsymbol{s}_2^*(p) = \overline{\boldsymbol{s}}^*(p), \quad c_1\boldsymbol{e}_1^*(p) + c_2\boldsymbol{e}_2^*(p) = \overline{\boldsymbol{e}}^*(p). \tag{14}$$

Substituting the deviatoric part of the constitutive relations (12) in the latest relation yields:

$$\boldsymbol{s}_2^*(p) = \frac{1}{c_2}\left(2\mu_2^*(p)\overline{\boldsymbol{e}}^*(p) - c_1\frac{\mu_2^*(p)}{\mu_1^*(p)}\boldsymbol{s}_1^*(p)\right). \tag{15}$$

Reporting this relation in the left-hand side of (14) gives:

$$\boldsymbol{s}_1^*(p) = \frac{2}{c_1}\left(\frac{1}{\mu_1^*(p)} - \frac{1}{\mu_2^*(p)}\right)^{-1}\left(\overline{\boldsymbol{e}}^*(p) - \frac{1}{2\mu_2^*(p)}\overline{\boldsymbol{s}}^*(p)\right), \tag{16}$$

while a similar relation holds for the spherical part of the average stress in phase (1), namely:

$$\sigma_{1m}^*(p) = \frac{3}{c_1}\left(\frac{1}{k_1^*(p)} - \frac{1}{k_2^*(p)}\right)^{-1}\left(\overline{\varepsilon}_m^*(p) - \frac{1}{3k_2^*(p)}\overline{\sigma}_m^*(p)\right). \tag{17}$$

As $\overline{\sigma}_m(0) = 3k^e\overline{\varepsilon}_m(0)$ and $\overline{\boldsymbol{s}}(0) = 2\mu^e\overline{\boldsymbol{e}}(0)$, the average stress in phase (1) at a time t is given by the inverse Laplace–Carson transform of the two previous relations:

$$\sigma_{1m}(t) = \frac{3}{c_1}\left(\frac{1}{k_1^v} - \frac{1}{k_2^v}\right)^{-1}\left(\dot{\overline{\varepsilon}}_m(t) - \frac{1}{3}\left(\frac{1}{k^e}\dot{\overline{\sigma}}_m(t) + \frac{1}{k_2^v}\overline{\sigma}_m(t)\right)\right),$$
$$\boldsymbol{s}_1(t) = \frac{2}{c_1}\left(\frac{1}{\mu_1^v} - \frac{1}{\mu_2^v}\right)^{-1}\left(\dot{\overline{\boldsymbol{e}}}(t) - \frac{1}{2}\left(\frac{1}{\mu^e}\dot{\overline{\boldsymbol{s}}}(t) + \frac{1}{\mu_2^v}\overline{\boldsymbol{s}}(t)\right)\right). \tag{18}$$

In addition, as the composite is elastically homogeneous, it comes: $\sigma_{1m}(0) = \overline{\sigma}_m(0)$ and $\boldsymbol{s}_1(0) = \overline{\boldsymbol{s}}(0)$. Obviously, similar expressions can be derived for the stress average over the phase (2) while the averaged strain in each phase can be easily inferred by using the constitutive equations (6). The approach we have presented above leads to shorten theoretical expressions in which no additional internal variables are needed to compute stress (or strain) averages in each phase.

2.2.3. Exact results for some particular microstructures

The following particular microstructures are considered hereafter:

- a "series" microstructure which would correspond to Reuss-type estimates,
- a "parallel" microstructure which would correspond to Voigt-type estimates,
- a matrix–inclusion microstructure with a low volume fraction of inclusions (say, up to 0.2): the overall behaviour will be estimated through the classical Mori and Tanaka (1973) scheme.

This last estimate identifies in the present case to the lower Hashin–Shtrikman bounds. We will denote hereafter by the subscripts $\gamma = R, V$ the reference to the Reuss and Voigt schemes, respectively. Using Eqs. (13), the Laplace–Carson transforms of the estimates of the shear and bulk parts $(\tilde{\mu}_\gamma(t), \tilde{k}_\gamma(t))$ of the overall relaxation function moduli are:

$$\tilde{\mu}_R^* = \left\langle \frac{1}{\mu^*} \right\rangle^{-1}, \quad \tilde{k}_R^* = \left\langle \frac{1}{k^*} \right\rangle^{-1}, \quad \tilde{\mu}_V^* = \langle \mu^* \rangle, \quad \tilde{k}_V^* = \langle k^* \rangle,$$

while the Mori–Tanaka scheme yields ($i = 1$ denotes the matrix):

$$\tilde{\mu}_{MT}^* = \mu_1^* + c_2\left(\frac{\mu_2^* - \mu_1^*}{1 + c_1\frac{\mu_2^* - \mu_1^*}{\hat{\mu}^* + \mu_1^*}}\right), \quad \tilde{k}_{MT}^* = k_1^* + c_2\left(\frac{k_2^* - k_1^*}{1 + c_1\frac{k_2^* - k_1^*}{\hat{k}^* + k_1^*}}\right),$$

with $\hat{k}^* = \frac{4}{3}\mu_1^*, \quad \hat{\mu}^* = \frac{\mu_1^*}{6}\frac{9k_1^* + 8\mu_1^*}{k_1^* + 2\mu_1^*}$.

These various expressions of the Laplace–Carson transforms of the shear and bulk parts of the overall relaxation function are rational functions of the variable p. These rational functions admit exact expansions which read as (γ=R, V, MT):

$$\tilde{\mu}_\gamma^*(p) = \sum_{i=1}^{N_{(\gamma)p}^d} \mu_{\overline{\tau}_{(\gamma)i}^d}\frac{p}{p + \frac{1}{\overline{\tau}_{(\gamma)i}^d}}, \quad \tilde{k}_\gamma^*(p) = \sum_{i=1}^{N_{(\gamma)p}^m} k_{\overline{\tau}_{(\gamma)i}^m}\frac{p}{p + \frac{1}{\overline{\tau}_{(\gamma)i}^m}}, \tag{19}$$

with the constraints (9). These expressions are the Laplace–Carson transforms of former expansions (8). As a result, these three schemes lead to an exact expansion (8). Expressions of the effective relaxation times $(\overline{\tau}_i^d, \overline{\tau}_i^m)$, as well as their numbers (N_p^d, N_p^m) and their magnitudes $(\mu_{\overline{\tau}_i^d}, k_{\overline{\tau}_i^m})$ defined by relation (5) are reported in Table 1 for each internal variable. On the one hand, for Voigt and Reuss estimates, these results are directly expressed as a function of the phases volume fractions, the elastic moduli and the relaxation times in each phase ($i = 1, 2$, $\tau_i^m = \frac{k_i^v}{k^e}$, $\tau_i^d = \frac{\mu_i^v}{\mu^e}$). On the other hand, for the Mori–Tanaka estimate, the results are expressed as a function of the four additional constants:

$$\frac{1}{\hat{\tau}_a^m} = \left\langle \frac{1}{\tau^m} \right\rangle + \frac{1}{4+3\xi}\left(\frac{3\xi}{\tau_1^d} + 4\left(\frac{c_2}{\tau_1^m} + \frac{c_1}{\tau_2^m}\right)\right),$$

$$\frac{1}{\hat{\tau}_b^m} = \sqrt{\frac{1}{4+3\xi}\left(\frac{3\xi}{\tau_1^d}\left\langle \frac{1}{\tau^m} \right\rangle + \frac{4}{\tau_1^m \tau_2^m}\right)},$$

$$\frac{1}{\hat{\tau}_a^d} = \frac{\left(\frac{15}{\tau_1^d} + \frac{9}{\tau_2^d}\right)\xi + (6\xi+12)\left\langle \frac{1}{\tau^d} \right\rangle + \frac{8}{\tau_2^d} + \frac{20}{\tau_1^m}}{15\xi+20},$$

$$\frac{1}{\hat{\tau}_b^d} = \sqrt{\frac{\frac{(9+6c_2)\xi}{\tau_1^d \tau_2^d} + \frac{8+12c_2}{\tau_2^d \tau_1^m} + \frac{6\xi c_1}{\tau_1^{d\,2}} + \frac{12c_1}{\tau_1^d \tau_1^m}}{15\xi+20}},$$

($\xi = \frac{k^e}{\mu^e}$) as well as the polynomial functions:

$$p^d(x) = \frac{5c_2\mu^e}{(2+3c_2)\tau_2^d(15\xi+20)} \quad \frac{(20+15\xi)\tau_2^d}{(\hat{\tau}_b^d)^2} + x\left((6+9c_2)\xi + 8 + 12c_2 + (9\xi+12)c_1\frac{\tau_2^d}{\tau_1^d}\right)$$

$$p^m(x) = k^e\left(\left\langle \frac{1}{\tau^m} \right\rangle x + \frac{1}{4+3\xi}\left(\frac{4}{\tau_1^m \tau_2^m} + 3\xi\left\langle \frac{1}{\tau^m} \right\rangle \frac{1}{\tau_1^d}\right)\right).$$

The number of internal variables obviously depends on the microstructure: Reuss estimate predicts a maxwellian overall behaviour (one single relaxation time) while the shear part of the relaxation function, as predicted by the Mori–Tanaka model, displays three different relaxation times (including the one of the matrix ($\alpha_{\tau_1^d}$)). In addition, the relative magnitudes $\left(\frac{\mu_{\tau_i^d}}{\mu^e}, \frac{k_{\tau_i^m}}{k^e}\right)$ as predicted by Voigt and Reuss models only depend on the phases volume fraction while these magnitudes also depend on the contrast between phases according to the Mori–Tanaka model. For a particular contrast, we have reported in Fig. 1 the evolutions of the magnitudes and the relaxation times of the three shear internal variables, with respect to the volume fraction, as predicted by the Mori–Tanaka model.

For a low volume fraction of inclusions, the overall behaviour of the composite tends towards the one of the matrix. As a result, in this limiting case, the magnitudes of the internal variables ($\alpha_{\tau_2}, \alpha_{\tau_3}$) tend towards zero while α_{τ_1} tends towards the viscous strain of the matrix. Conversely, the overall behaviour tends towards the one of the inclusions for a high volume fraction of inclusions. The internal variables ($\alpha_{\tau_1}, \alpha_{\tau_2}$) tend to vanish while α_{τ_3} tends to the viscous strain of the inclusions (notice that the corresponding relaxation time tends to the one of the inclusions (Fig. 1b)). For moderate volume fraction of inclusions ($c_2 \approx 0.2$ for instance), the internal variable $\alpha_{\tau_1^d}$ displays the highest relative magnitude (≈0.6) but the two remaining internal variables can not be neglected.

2.2.4. General microstructures: approximated results

Results obtained hereabove are consistent for some two-phases composites of inclusion–matrix type. Results are exact because the relaxation function has, in each case, a discrete spectrum. Actually, for more general microstructures in which no phase plays the specific role of matrix (polycrystals,...), the relaxation function should have a continuous spectrum (see, Rougier et al., 1993). Moreover, even for such composites of inclusion–matrix type (but with higher volume fraction of inclusions), Rougier et al. (1993) show that a generalized self-consistent scheme (see, Christensen, 1979) also leads to a relaxation function with continuous spectrum. Hence, one may use Prony series development of the relaxation function to obtain an approximated solution:

$$\tilde{\mu}(t) \approx \tilde{\mu}_d(t) = \sum_{i=1}^{N_p^d} \mu_{\tau_i^d} e^{-\frac{t}{\tau_i^d}}, \quad \tilde{k}(t) \approx \tilde{k}_m(t) = \sum_{i=1}^{N_p^m} k_{\tau_i^m} e^{-\frac{t}{\tau_i^m}}.$$

The so-called collocation method does not require the exact relaxation function but only some values of its Laplace transform:

$$1 \leqslant i \leqslant N_p^d \quad \mu^*\left(\frac{1}{\tau_i}\right) = \mu_d^*\left(\frac{1}{\tau_i}\right), \qquad 1 \leqslant i \leqslant N_p^m \quad k^*\left(\frac{1}{\tau_i}\right) = k_m^*\left(\frac{1}{\tau_i}\right).$$

The accuracy of the result depends on the number of values, that is the number of the terms (N_p^d and N_p^m) within the Prony series expansions. For polycrystalline microstructures (see, Turner et al., 1994), 20 terms in the Prony series are enough to yield satisfying results. More recently, Rekik and Brenner (2007) improved the collocation method to decrease the number of terms within the Prony series expansion. These authors obtained with far less than 20 terms very accurate results when considering the case of a continuous spectrum (two-phases isotropic incompressible self-consistent model) for which the homogenized relaxation function is known.

3. Extension to ageing linear viscoelasticity

Hereafter, the results derived in the previous section are applied to predict the overall response of an ageing viscoelastic heterogeneous medium. From a general point of view, at least one of the phase of this heterogeneous medium displays a viscoelastic ageing behaviour (see for instance, Mandel, 1974):

$$\sigma(t) = \int_0^t E(t, t-u)\mathrm{d}\epsilon(u), \tag{20}$$

with (ϵ, σ) the shorten notations of strain and stress introduced in Section 2.1. In this new expression, the relaxation function $E(t, t')$ depends separately on t and t' rather than only on the time lag $(t - t')$. This general framework enlarges constitutive behaviours

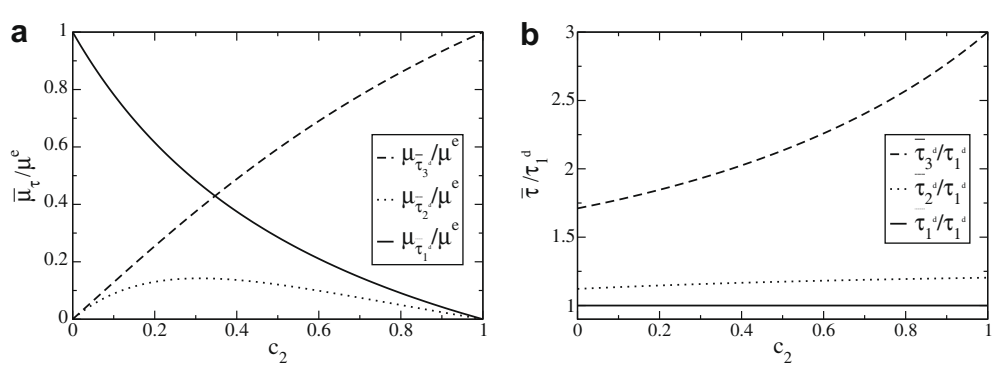

Fig. 1. Shear internal variables (Mori–Tanaka model): evolutions of the magnitudes (a) and the relaxation times (b) as a function of the volume fraction of inclusions $\left(\frac{\tau_2^d}{\tau_1^d} = 3,\ \frac{\tau_2^m}{\tau_1^m} = 1,\ \frac{\tau_1^m}{\tau_1^d} = 2 \text{ and } \xi \approx 2.61\right)$.

for which the time appears explicitly (for instance, the ageing of concrete by chemical processes) as well as constitutive behaviours for which the time dependency is the result of the loading. This last case is also of practical importance when considering, for instance, thermomechanical loading: the parameters of the constitutive law (e.g. the shear viscous modulus) depend generally on temperature. Similarly, the loading may also induce physical changes throughout one of the phase of the considered heterogeneous medium (damage, densification processes,...). Consequently some parameters of the constitutive law will also depend on time. We study this last situation in the sequel for the particular case of a matrix–inclusion microstructure with a low volume fraction of inclusions.

3.1. An ageing viscoelastic problem

We consider hereafter an ageing porous viscoelastic matrix reinforced with elastic inclusions. In order to remain within the general framework presented above, we assume low initial volume fraction of pores (up to 10%). Porosity evolution effects on the elastic behaviour of the matrix will then be neglected. Hence, elastic properties of the composite are homogeneous and given below.

The inclusions obey an elastic behaviour (Young modulus = 183.4 GPa, Poisson ratio = 0.31) while the matrix obeys a viscoelastic behaviour. At a much lower scale, the matrix displays pores with a volume fraction denoted by f_p (see Fig. 2).

After homogenization at the lower scale (see Fig. 2), the inelastic strain rate second-order tensor in the matrix is given by:

$$\dot{\boldsymbol{\varepsilon}}^v = \frac{\mathrm{d}\psi(\boldsymbol{\sigma}, f_p)}{\mathrm{d}\boldsymbol{\sigma}},$$

with $\psi(\boldsymbol{\sigma}, f_p)$ the dissipative potential. The evolution equation for the volume fraction of voids reads (volume balance):

$$\dot{f}_p = 3(1 - f_p)\dot{\varepsilon}_m. \tag{21}$$

Hereafter, the dissipative potential of the porous matrix is approximated by Michel and Suquet (1992):

$$\psi(\boldsymbol{\sigma}, f_p) = \frac{1}{2}\sigma_0\dot{\varepsilon}^0 \quad A(f_p)\left(\frac{3\sigma_m}{2\sigma_0}\right)^2 + B(f_p)\left(\frac{\sigma_{eq}}{\sigma_0}\right)^2\Bigg),$$

with:

$$A(f_p) = \frac{f_p}{1 - f_p} \quad \text{and} \quad B(f_p) = \frac{1 + \frac{2}{3}f_p}{1 - f_p}.$$

As a result, the inelastic strain in the matrix reads:

$$\boldsymbol{\varepsilon}^v = \frac{3\dot{\varepsilon}^0 B(f_p)}{2\sigma_0}\boldsymbol{s} + \frac{3\dot{\varepsilon}^0 A(f_p)}{4\sigma_0}\sigma_m\boldsymbol{\delta}.$$

Alternatively, the viscous bulk and shear moduli of the matrix depend on the porosity at any point (x) of the matrix and read:

$$k_1^v(x) = \frac{4\zeta_1}{3A(f_p(x))}, \quad \mu_1^v(x) = \frac{\zeta_1}{B(f_p(x))} \quad \text{with } \zeta_1 = \frac{\sigma_0}{3\dot{\varepsilon}^0}. \tag{22}$$

Since the internal variable f_p depends on time (see the evolution law (21)), this last expression clearly shows that the bulk and shear viscous moduli will implicitly depend on time for a given loading. Hence, the associated bulk and shear relaxation functions display a time dependency similar to the one defined in relation (20). In other words, the viscoelastic behaviour of this matrix–inclusion heterogeneous medium is of the ageing kind.

3.2. The incremental model

Let us consider an overall stress loading path $\overline{\sigma}(u)$ on the time interval $[0, t - \delta t]$. We aim at the determination of the effective response $\overline{\varepsilon}(t)$. We must assume that the problem is solved until $u = t - \delta t$. At time t, the viscous properties of the matrix are functions of the (non-uniform) porosity field $f_p(t, x)$. However, to keep the formulation with a finite number of internal variables, the viscous properties at any point (x) of the matrix are approximated at time t by:

$$k_1^v(f_p(t,x)) \approx k_1^v(f_{1p}(t)) \quad \text{and} \quad \mu_1^v(f_p(t,x)) \approx \mu_1^v(f_{1p}(t)), \tag{23}$$

where $f_{1p}(t)$ denotes the average porosity over the matrix at time t ($f_{1p}(t) = \langle f_p(t,x)\rangle_1$).

The average strains over the two phases as well as the internal variables $(\boldsymbol{\alpha}^d_{\overline{\tau}^d_i}, \alpha^m_{\overline{\tau}^m_i})$ and the average porosity f_{1p} over the matrix are thus known on $[0, t - \delta t]$ and the unknowns are: $\boldsymbol{\varepsilon}_1(t)$, $\boldsymbol{\varepsilon}_2(t)$, $\boldsymbol{\alpha}^d_{\overline{\tau}^d_j}(t)$, $\alpha^m_{\overline{\tau}^m_i}(t)$, $f_{1p}(t)$ (we recall that i and j depend on the used homogenization scheme, see Section 2.2.3). At time t, the behaviour at any point (x) of the matrix is approximated by:

$$\begin{aligned} \dot{\varepsilon}_{1m}(t,x) &= \frac{1}{3k^e}\dot{\sigma}_m(t,x) + \frac{1}{3k_1^v(f_{1p}(t))}\sigma_m(t,x), \\ \dot{\boldsymbol{e}}(t,x) &= \frac{1}{2\mu^e}\dot{\boldsymbol{s}}(t,x) + \frac{1}{2\mu_1^v(f_{1p}(t))}\boldsymbol{s}(t,x), \end{aligned} \tag{24}$$

where the viscous and shear moduli now depend on the average porosity f_{1p} over the matrix.

We consider the heterogeneous composite whose microstructure is the same as the previous one but with the matrix obeying the non-ageing linear viscoelastic behaviour as defined by (24). To estimate the effective response at time t of this composite, the Mori–Tanaka scheme is adopted since the volume fraction of inclusions remains low. Using the results derived in Section 2.2.1, the overall response of this non-ageing viscoelastic composite is given by:

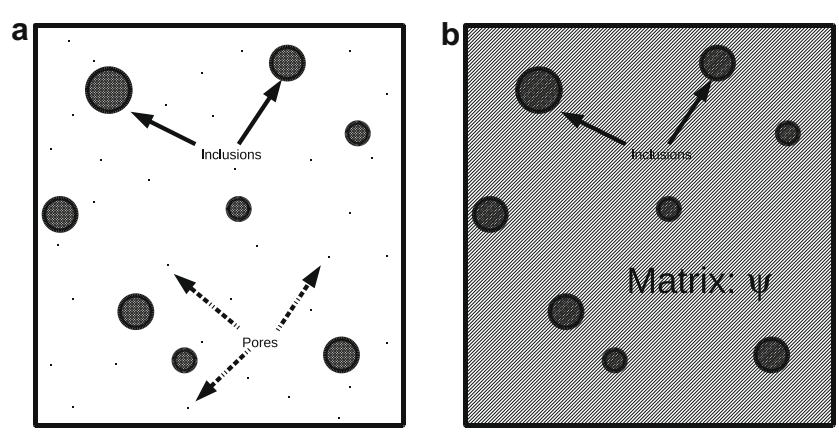

Fig. 2. Schematic representations of the composite: (a) at the lowest scale, the pores are represented by dots, (b) after homogenization of the lowest scale (ψ is the effective potential of the porous matrix). In both pictures the elastic inclusions are represented by hatched disks.

Table 1
Exact values of the effective relaxation times as predicted by Voigt, Reuss and Mori–Tanaka homogenization schemes. The magnitudes of each internal variable are also reported.

	Reuss	Voigt	Mori–Tanaka
Shear			
Relaxation times	$N_p^d = 1$	$N_p^d = 2$	$N_p^d = 3$
		$\overline{\tau}_1^d = \tau_1^d$	$\overline{\tau}_1^d = \tau_1^d$
	$\overline{\tau}^d = \langle \frac{1}{\tau^d} \rangle^{-1}$	$\overline{\tau}_2^d = \tau_2^d$	$\overline{\tau}_2^d = 2\hat{\tau}_a^d \left(1 - \sqrt{1 - 4(\hat{\tau}_a^d/\hat{\tau}_b^d)^2}\right)^{-1}$
			$\overline{\tau}_3^d = 2\hat{\tau}_a^d \left(1 + \sqrt{1 - 4(\hat{\tau}_a^d/\hat{\tau}_b^d)^2}\right)^{-1}$
Magnitudes		$\mu_{\overline{\tau}_1^d} = c_1 \mu^e$	$\mu_{\overline{\tau}_1^d} = \frac{2c_1}{(2+3c_2)}\mu^e$, $\mu_{\overline{\tau}_2^d} = \frac{\overline{\tau}_2^d}{\frac{1}{\hat{\tau}_a^d} - \frac{1}{\overline{\tau}_2^d}} p^d \left(-\frac{1}{\overline{\tau}_2^d}\right)$
	$\mu_{\overline{\tau}^d} = \mu^e$	$\mu_{\overline{\tau}_2^d} = c_2 \mu^e$	$\mu_{\overline{\tau}_3^d} = \mu^e - \mu_{\overline{\tau}_1^d} - \mu_{\overline{\tau}_2^d}$
Bulk			
Relaxation times	$N_p^m = 1$	$N_p^m = 2$	$N_p^m = 2$
	$\overline{\tau}^m = \langle \frac{1}{\tau^m} \rangle^{-1}$	$\overline{\tau}_1^m = \tau_1^m$	$\overline{\tau}_1^m = 2\hat{\tau}_a^m \left(1 - \sqrt{1 - 4(\hat{\tau}_a^m/\hat{\tau}_b^m)^2}\right)^{-1}$
		$\overline{\tau}_2^m = \tau_2^m$	$\overline{\tau}_2^m = 2\hat{\tau}_a^m \left(1 + \sqrt{1 - 4(\hat{\tau}_a^m/\hat{\tau}_b^m)^2}\right)^{-1}$
Magnitudes		$k_{\overline{\tau}_1^m} = c_1 k^e$	$k_{\overline{\tau}_1^m} = \frac{(\overline{\tau}_1^m)^2 \overline{\tau}_2^m}{\overline{\tau}_1^m - \overline{\tau}_2^m} p^m \left(-\frac{1}{\overline{\tau}_1^m}\right)$
	$k_{\overline{\tau}^m} = k^e$	$k_{\overline{\tau}_2^m} = c_2 k^e$	$k_{\overline{\tau}_2^m} = k^e - k_{\overline{\tau}_1^m}$

$$\overline{\boldsymbol{e}}(t) = \frac{1}{2\mu^e}\overline{\boldsymbol{s}}(t) + \sum_{i=1}^{3} \boldsymbol{\alpha}^d_{\overline{\tau}_i^d}(t), \quad \overline{\varepsilon}_m(t) = \frac{1}{3k^e}\overline{\sigma}_m(t) + \sum_{i=1}^{2} \alpha^m_{\overline{\tau}_i^m}(t), \tag{25}$$

where the internal variables $\alpha^m_{\overline{\tau}_i^m}(t)$ $(1 \leqslant i \leqslant 2)$ and $\boldsymbol{\alpha}^d_{\overline{\tau}_i^d}(t)$ $(1 \leqslant i \leqslant 3)$ are, respectively, scalar and (second-order) tensorial functions of time. They, respectively, satisfy the evolution laws (11) while the five scalar coefficients $(\mu_{\overline{\tau}_1^d}, \mu_{\overline{\tau}_2^d}, \mu_{\overline{\tau}_3^d}, \mu_{\overline{\tau}_1^m}, \mu_{\overline{\tau}^m})$ are given in the Table 1 as a function of material constants. These coefficients depend on the bulk and shear viscous moduli of the matrix appearing in the relations (23). The results derived in Section 2.2.2 enable to calculate the average strain over the matrix at time t.

To close these equations, we have to derive the average porosity over the matrix at time t. Hence, the evolution equation of porosity at any point x of the matrix is given by:

$$\dot{f}_p(t,x) = 3(1 - f_p(t,x))\dot{\varepsilon}_m(t,x).$$

As a result, the evolution law of the average porosity over the matrix is given by:

$$\dot{f}_{1p}(t) = 3\langle (1 - f_p(t,x))\dot{\varepsilon}_m(t,x)\rangle_1.$$

The porosity being small, a first order expansion of the previous equation reads:

$$\dot{f}_{1p}(t) \approx 3(1 - f_{1p}(t))\dot{\varepsilon}_{1m}(t). \tag{26}$$

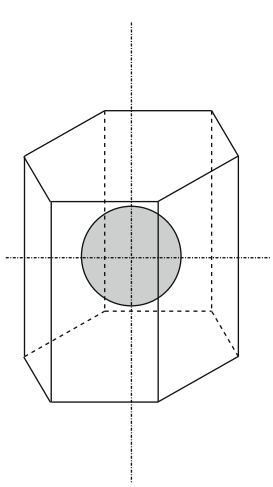

Fig. 3. The hexahedric periodic cell.

Finally, it is worth remarking that the averaged spherical strain in the matrix appearing in the right term of Eq. (26) implicitly depends on the average porosity over the matrix at time t since the bulk and shear viscous moduli of the matrix are (non-linear) functions of this unknown. Hence, Eq. (26) is a scalar but non-linear first order differential equation of the unknown $f_{1p}(t)$.

4. Results

The composite considered in Section 3 is subjected to a compressive creep test at $t = 0$. The overall stress is given by $\overline{\boldsymbol{\sigma}} = \overline{\sigma}^0 \boldsymbol{\delta}$ with $\overline{\sigma}^0 < 0$.

To assess the accuracy of the model presented above, its predictions are compared to exact response of a specific class of microstructures: a periodic distribution of inclusions. The full-field simulation of this periodic microstructure is performed by considering a three-dimensional unit cell as depicted in Fig. 3. Thanks to symmetry considerations, one eighth of this unit cell is meshed with 274 quadratic elements (see Fig. 4). The accuracy of next simulations has been evaluated by increasing the number of elements up to 1210: relative deviations have never exceeded 1%.

Fig. 5 shows the evolution of the overall strain (spherical part) as a function of normalized time $\frac{t}{\tau^d}$ for different values of the initial volume fraction of pores f_{1p} (parameter ζ_1 in relation (22) is easily deduced from the relaxation time of the matrix τ_1^d as given in Table 1 and the initial porosity $f_{1p}(0)$). As a limiting case, one can note

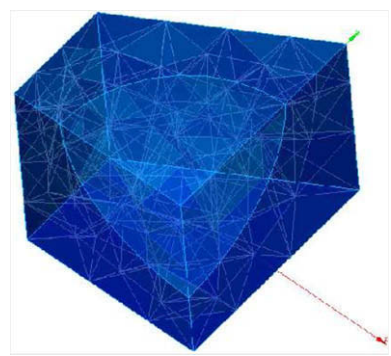

Fig. 4. Finite element discretization of one eighth of the periodic cell.

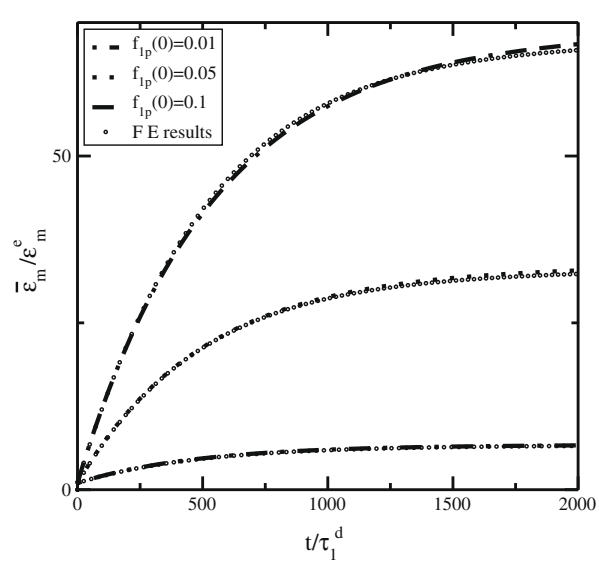

Fig. 5. Evolutions of the predicted overall strain as a function of time for different values of the initial porosity ($c_2 = 0.2$, $\xi \approx 2.3$, $\mu^e = 70$ GPa and $\overline{\sigma}^0 = -200$ MPa).

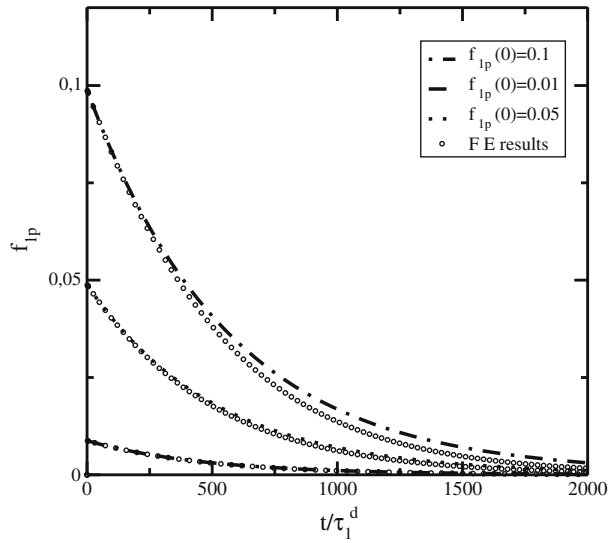

Fig. 6. Evolutions of the average porosity f_{1p} over the matrix as a function of time for different values of the initial porosity ($c_2 = 0.2$, $\xi \approx 2.3$, $\mu^e = 70$ GPa and $\overline{\sigma}^0 = -200$ MPa).

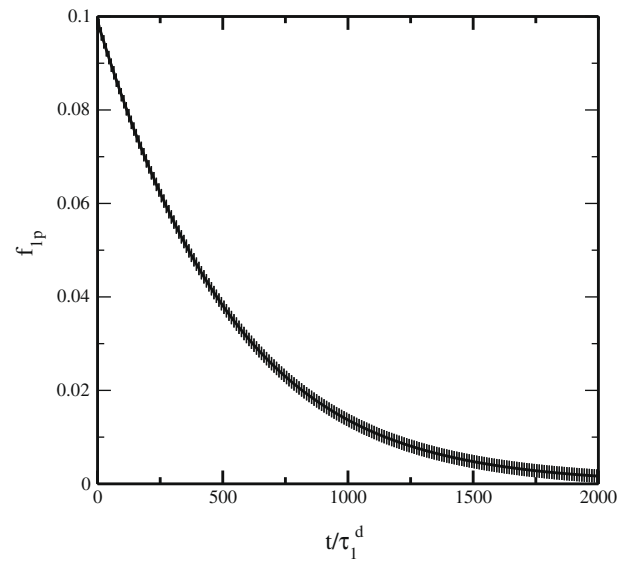

Fig. 7. Evolution of the average porosity (continuous line) and its spatial fluctuation (vertical bars) as a function of time for an initial porosity of 10%.

that for a dense matrix (i.e. $f_{1p}(0) = 0$), the viscous strain throughout the matrix is traceless. As a result, the overall response of the composite is purely elastic.[2]

Conversely, a non-zero initial volume fraction of pores induces creep phenomena inside the matrix. This inelastic strain is provided by the decrease of the average porosity f_{1p} as depicted in Fig. 6. As expected, when the volume fraction of porosity tends towards zero, the overall strain rate decreases: the creep spherical strain is bounded (see Fig. 5).

Finally, the reference results as predicted by full finite element simulations (and identified by circles in Figs. 5 and 6) are very close to the ones of the incremental model. This excellent agreement can be explained as follows: first, the porosity field is non-uniform (see Fig. 7) but its spatial fluctuations remain low. As a result, the use of an average porosity (approximations (23) and (26)) is consistent in this situation. In addition, the first order effect of the evolution of the porosity on the constitutive behaviour of the matrix is well represented: at each time step, the viscous compressibility of the matrix increases and the viscoelastic compressibility tends towards a purely elastic one. This last point illustrates the ability of the proposed incremental model to represent ageing effects.

5. Conclusions

In order to derive the overall behaviour of heterogeneous media, the internal variables formulation presented hereabove as well as the one proposed by Lahellec and Suquet (2007) offers an interesting alternative to integral methods. First, it should be useful when introducing viscoelastic overall constitutive laws in finite element codes. Based on internal variables, these formulations overcome some computational limits like the requirement of the storage of all the stress (or strain) history from the initial time to the present time at which the effective response of the composite is estimated. As explained above, the merit of the proposed internal variables formulation lies on the fact that it provides exact results for the particular two-phase composites considered ("series", "parallel" and matrix–inclusion microstructures at low fraction of inclusions). Exact expressions have been reported (see Table 1) when the two phases of the (homogeneous elastic) composite obey an isotropic maxwellian behaviour. Conversely, it is worth mentioning that the easiness of this method remains limited to a large range of constitutive laws as well as microstructures types. Even for the microstructures considered above, the study of more general constitutive behaviours (Maxwell chain model for instance) would require more intricate algebraic computations which still lead to exact expansions but with a higher number of terms. In addition, algebraic computations of the effective properties would fail (see for instance, Rougier et al., 1993) for more general situations (matrix–inclusion microstructures at higher volume fraction for instance). Nevertheless, our method remains valid. The use of appropriate and accurate collocation methods (see for instance, Rekik and Brenner, 2007) leads as Lahellec and Suquet (2007) to approximated results.

Last but not least, these internal variables formulations enable straightforwardly extension to ageing behaviours. This last point has been illustrated with the help of the proposed internal variables formulation: a theoretical framework has been developed to derive effective properties of ageing linear viscoelastic composite. Finally as an example, effective properties of a porous viscous matrix with elastic inclusions have consistently been derived with this internal variables formulation. Such results have been compared with success to full-field simulations of a periodic microstructure.

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[2] The elastic properties of the composite being homogeneous, the elastic stress field obeys at any point (x) of the composite: $\boldsymbol{\sigma}(x) = \overline{\boldsymbol{\sigma}} = \overline{\sigma}^0 \boldsymbol{\delta}$. Hence, the deviatoric part of the stress field vanishes and the viscous strain at any point (x) of the matrix reads: $\lim_{k^v \to \infty} \dot{\boldsymbol{\varepsilon}}^v(x) = 0$.

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