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HOMOGENIZATION OF NONLINEAR VISCOELASTIC THREE-PHASE PARTICULATE COMPOSITES

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Summary: *We aim at modeling the mechanical behavior of a nonlinear viscoelastic heterogeneous material having an "inclusions-matrix" type microstructure. The different phases of this material may experience differential shrinkages or swellings (like thermal expansion) and we want to model the internal stresses induced by this loading. With homogenization, these internal stresses are correctly estimated when the behavior of the constituents is linear elastic or even aging linear viscoelastic. A modified secant formulation has been recently introduced to deal with nonlinear viscoelastic behaviour. The general formulation can handle stress-free strain. For a two-phase particulate composite, closed-form expressions of the time evolutions of the effective behaviour as well as phase-averaged fields have also been reported. We propose here to extend these closed-form expressions to three-phase particulate composites. Results of this model are compared to full-fields computations of representative volume elements. The effect of the third phase (inclusions) is particularly studied.*

1. INTRODUCTION

This work is devoted to the modeling of composites made of a matrix reinforced by particles. When the phases have an elasto-viscoplastic behavior, [1] proposed a modified secant approach. This approach conjugates the advantages of the internal variable representation of linear viscoelasticity [2] with the linearization procedure developed initially by [3] and incorporating during the linearization process the time evolution of the fields fluctuations. It yields estimates which are are consistent with the one given for non aging linear viscoelastic composites (correspondence principle) while coinciding with the upper bound [4] in the purely viscoplastic regime. For two-phases isotropic particulate composites, the time evolution of the mechanical answer of the composite (overall and phase-averaged responses) has been shown to be given by a set of first-order differential equations whose coefficients are explicitly given as a function of the microstructural parameters [1].

Here the effect of a second family of inclusions on the mechanical answer is investigated. These additional inclusions will also display an elasto-viscoplastic isotropic behaviour but their mechanical properties may differ strongly from the two other phases.

This extension to a three-phase model is presented in section 2. The model predictions are then compared to full-field computations by considering a composite material made of a ductile matrix partially reinforced by rigid inclusions in section 3.

2. A modified secant model for three-phase isotropic particulate composites

Each phase of the three-phase composite obey an isotropic behavior: $\mu_e^{(r)}$ and $\kappa_e^{(r)}$ denote the shear and bulk elastic moduli of each phase (r). The viscous strain rate $\dot{\boldsymbol{e}}_v$ is given by :

$$\dot{\boldsymbol{e}}_v(\boldsymbol{x},\, t) = \frac{\partial U_v^{(r)}}{\partial \boldsymbol{s}} \tag{1}$$

where the dissipation potential $U_v^{(r)}$ of a given phase (r) is a convex function of the equivalent stress σ_{eq} defined as usually by $\sigma_{eq} = \sqrt{3\,(\boldsymbol{s} : \boldsymbol{s})/2}$ with $\boldsymbol{s} = \boldsymbol{\sigma} - \sigma_m\, \boldsymbol{\delta}$ the deviatoric stress, $\boldsymbol{\delta}$ the identity second-order tensor and $\sigma_m = \mathrm{tr}(\boldsymbol{\sigma})/3$ the hydrostatic part of the stress.

In addition, the shape and the spatial distribution of the phases are also isotropic. In that particular situation, if we adopt the modified secant linearization procedure, we show in the following subsections that the overall elasto-viscoplastic behavior of the composite as well as the time evolution of the phase-averaged strain and stress can be easily derived from the general theory [1].

Hereafter, the strain and stress fields are nil for any time (t) lower or equal to the initial loading time $t = 0$ while the loading time functions consisting of the overall stress (or the overall strain) are continuous functions of time (no time discontinuities).

2.1 Three-phase isotropic composites, case of a non aging linear viscoelastic behavior

The Laplace-Carson transform (the correspondence principle [5]) can be used to solve this problem[1]. This method leads to a fictitious elastic problem in the Laplace-Carson domain, the symbolic elastic compliance of each phase (r) being isotropic.
Therefore, $m_d^{(r)*}(p) = \dfrac{1}{2\,\mu^{(r)*}(p)} = \dfrac{1}{2\,\mu_e^{(r)}} + \dfrac{1}{p}\,\dfrac{1}{2\,\mu_v^{(r)}}$ and $m_m^{(r)*}(p) = \dfrac{1}{3\,\kappa_e^{(r)}}$ denote respectively the Laplace-Carson transforms of the shear and bulk components of the creep function of a given phase (r). The usual homogenization methods can then be applied to estimate the Laplace-Carson transform of the effective creep or relaxation functions while the time-responses are deduced by the inversion of the Laplace-Carson transform.

2.1.1 Effective behavior

The overall behavior of the considered composites being isotropic, it depends also on only two scalar functions of time, the bulk and shear creep functions whose Laplace-Carson transform is expressed as the algebraic inverse of the bulk and shear components of the relaxation

[1]The Laplace-Carson transform of any function of time $f(t)$ with respect to the parameter p is denoted by $f^*(p)$ and is defined by : $\boldsymbol{f}^*\,(p) = p\,\int_0^{+\infty} \boldsymbol{f}\,(u)\, e^{-pu}\, du$

functions, namely $\left(\widetilde{m}_d^*(p) = \dfrac{1}{2\,\widetilde{\mu}^*(p)}, \widetilde{m}_m^*(p) = \dfrac{1}{3\,\widetilde{\kappa}^*(p)}\right)$. In the following, $(\widetilde{\mu}_e,\, \widetilde{\kappa}_e)$ and $\widetilde{\mu}_v$ respectively denote the effective moduli of the composites in the purely elastic $(p \to \infty)$ and purely viscous $(p \approx 0)$ regimes.

As demonstrated in [1], the Laplace-Carson transforms of the effective shear $\widetilde{m}_d^*(p)$ and bulk $\widetilde{m}_m^*(p)$ creep functions read (at least approximately) as Dirichlet series expansion :

$$\widetilde{m}_d^*(p) = \frac{1}{2\,\widetilde{\mu}_e} + \frac{1}{p}\,\frac{1}{2\,\widetilde{\mu}_v} + \sum_{i=1}^{i=N_c^d} \widetilde{m}_{d(i)}\,\frac{1}{p + \frac{1}{\widetilde{\tau}_{d(i)}}} \quad \text{and} \quad \widetilde{m}_m^*(p) = \frac{1}{3\,\widetilde{\kappa}_e} + \sum_{i=1}^{i=N_c^m} \widetilde{m}_{m(i)}\,\frac{1}{p + \frac{1}{\widetilde{\tau}_{m(i)}}}. \tag{2}$$

As a consequence, the macroscopic strain - stress relation can be expressed (at least approximately) as the following internal variables formulation:

$$\dot{\overline{\boldsymbol{e}}}(t) = \frac{1}{2\,\widetilde{\mu}_e}\dot{\overline{\boldsymbol{s}}}(t) + \frac{1}{2\,\widetilde{\mu}_v}\overline{\boldsymbol{s}}(t) + \sum_{i=1}^{i=N_c^d} \dot{\widetilde{\boldsymbol{\alpha}}}_{d(i)}(t) \quad \text{and} \quad \dot{\overline{\varepsilon}}_m(t) = \frac{1}{3\,\widetilde{\kappa}_e}\,\dot{\overline{\sigma}}_m(t) + \sum_{i=1}^{i=N_c^m} \dot{\widetilde{\alpha}}_{m(i)}(t) \tag{3}$$

where the N_c^d and N_c^m internal variables related to the shear and bulk overall behavior obey :

$$\begin{cases} i = 1..N_c^d : \quad \dot{\widetilde{\boldsymbol{\alpha}}}_{d(i)}(t) + \dfrac{1}{\widetilde{\tau}_{d(i)}}\,\widetilde{\boldsymbol{\alpha}}_{d(i)}(t) = \widetilde{m}_{d(i)}\,\overline{\boldsymbol{s}}(t), \qquad \widetilde{\boldsymbol{\alpha}}_{d(i)}(0) = 0 \\ i = 1..N_c^m : \quad \dot{\widetilde{\alpha}}_{m(i)}(t) + \dfrac{1}{\widetilde{\tau}_{m(i)}}\,\widetilde{\alpha}_{m(i)}(t) = \widetilde{m}_{m(i)}\,\overline{\sigma}_m(t), \quad \widetilde{\alpha}_{m(i)}(0) = 0. \end{cases} \tag{4}$$

Relations (3) and (4) define entirely the effective behavior of the considered three-phase composite. Of course, the effective elastic and viscous properties as well as the number of internal variables will depend on the chosen homogenization model.

2.1.2 Time evolutions of the phase-averaged stresses

In [1], we took advantage of particular relations related to two-phases composites to derive direct expressions of the Laplace-Carson transform of the stress localization tensors. Here, the general expression (14)-left in [1] must be used but can still be considerably simplified given that the composites under considerations are isotropic. Indeed, the Laplace-Carson transform of the averages per phase of the stress localization tensor displays two independent components, namely its shear and bulk components as given by (at least approximately) :

$$\widetilde{b}_m^{(r)*}(p) = \widetilde{b}_{m(e)}^{(r)} + \sum_{i=1}^{i=N_c^m} \widetilde{b}_{m(i)}^{(r)}\,\frac{\frac{1}{\widetilde{\tau}_{m(i)}}}{p + \frac{1}{\widetilde{\tau}_{m(i)}}} \quad \text{and} \quad \widetilde{b}_d^{(r)*}(p) = \widetilde{b}_{d(e)}^{(r)} + \sum_{i=1}^{i=N_c^d} \widetilde{b}_{d(i)}^{(r)}\,\frac{\frac{1}{\widetilde{\tau}_{d(i)}}}{p + \frac{1}{\widetilde{\tau}_{d(i)}}} \tag{5}$$

In these last relations, the collocation times appearing in the two Prony series are the ones of the shear and bulk creep functions in (4). This choice is indeed possible if these last relations are approximations but is also relevant for the particular Hashin-Shtrikman estimates considered below.

Foremost, notice that the relation (13) in [1] can be considerably simplified in that isotropic case. Therefore, the deviatoric phase-averaged stresses for a given phase (r) reads :

$$\overline{\boldsymbol{s}}^{(r)}(t) = \widetilde{b}_{d(e)}^{(r)}\, \overline{\boldsymbol{s}}(t) + \sum_{i=1}^{i=N_c^d} \widetilde{\boldsymbol{\beta}}_{d(i)}^{(r)}(t) \tag{6}$$

$$\text{with} \quad i = 1..N_c^d : \dot{\widetilde{\boldsymbol{\beta}}}_{d(i)}^{(r)}(t) + \frac{1}{\widetilde{\tau}_{d(i)}}\, \widetilde{\boldsymbol{\beta}}_{d(i)}^{(r)}(t) = \frac{\widetilde{b}_{d(i)}^{(r)}}{\widetilde{\tau}_{d(i)}}\, \overline{\boldsymbol{s}}(t),\ \widetilde{\boldsymbol{\beta}}_{d(i)}^{(r)}(0) = 0$$

the time evolution of the deviatoric part of the macroscopic stress $\overline{\boldsymbol{s}}(t)$ being prescribed. As explained in [1], if the macroscopic strain is prescribed, the macroscopic stress will be derived from the macroscopic constitutive law - relations (3) and (4).
Concerning the hydrostatic component of the phase-averaged stresses, the time evolution reads :

$$\overline{\sigma}_m^{(r)}(t) = \widetilde{b}_{m(e)}^{(r)}\, \overline{\sigma}_m(t) + \sum_{i=1}^{i=N_c^m} \widetilde{\beta}_{m(i)}^{(r)}(t) \tag{7}$$

$$\text{with} \quad i = 1..N_c^m : \dot{\widetilde{\beta}}_{m(i)}^{(r)}(t) + \frac{1}{\widetilde{\tau}_{m(i)}}\, \widetilde{\beta}_{m(i)}^{(r)}(t) = \frac{\widetilde{b}_{m(i)}^{(r)}}{\widetilde{\tau}_{m(i)}}\, \overline{\sigma}_m(t),\ \widetilde{\beta}_{m(i)}^{(r)}(0) = 0$$

Relations (6) and (7) define explicitly the time evolution of the phase-averaged stresses for a three-phase linear viscoelastic composite, whatever the choice of the homogenization theory.

2.1.3 Hashin-Shtrikman estimates

If the Hashin-Shtrikman model is used, as already remarked in [2], the Laplace-Carson transforms of the shear and bulk components of the effective properties – described in Appendix A – are rational function of the variable p. As a result, the Laplace-Carson of these effective properties as well as the ones of the localization tensors express exactly as a Dirichlet series expansion. In other words, the relations (3) and (4) define exactly the effective behavior of the composite. For the considered 3-phases composite, the number of internal variables related to the time evolution of the shear and bulk creep functions equals $N_c^d = 4$ and $N_c^m = 2$, respectively. The corresponding algebraic developments extend the ones given in [1] related to two-phases isotropic composites as well as the ones obtained by [6] and related to homogeneous elastic 3-phases viscoelastic composites.

The Laplace-Carson transforms of the effective shear and bulk creep functions can be written as a function of the shear and bulk components of the Laplace-Carson transform of the averages per phase of the stress localization tensor:

$$\widetilde{m}_m^*(p) = \frac{1}{3}\left(\frac{1}{\kappa_e^{(1)}} + \sum_{r=2}^{r=3} c^{(r)}\, \widetilde{b}_m^{(r)*}(p)\left(\frac{1}{\kappa_e^{(r)}} - \frac{1}{\kappa_e^{(1)}}\right)\right) \tag{8}$$

$$\widetilde{m}_d^*(p) = \frac{1}{2}\left(\frac{1}{\mu^{(1)*}(p)} + \sum_{r=2}^{r=3} c^{(r)}\,\widetilde{b}_d^{(r)*}(p)\left(\frac{1}{\mu^{(r)*}(p)} - \frac{1}{\mu^{(1)*}(p)}\right)\right) \tag{9}$$

where $c^{(r)}$ denotes the volume fraction of each phase (r), $\sum_{r=1}^{r=3} c^{(r)} = 1$.

The elastic part of the effective shear and bulk and the viscous part of the effective shear can then be written :

$$\widetilde{\mu}_e = \left(\frac{1}{\mu_e^{(1)}} + \sum_{r=2}^{r=3} c^{(r)}\,\widetilde{b}_{d(e)}^{(r)}\left(\frac{1}{\mu_e^{(r)}} - \frac{1}{\mu_e^{(1)}}\right)\right)^{-1},\ \widetilde{\kappa}_e = \left(\frac{1}{\kappa_e^{(1)}} + \sum_{r=2}^{r=3} c^{(r)}\,\widetilde{b}_{m(e)}^{(r)}\left(\frac{1}{\kappa_e^{(r)}} - \frac{1}{\kappa_e^{(1)}}\right)\right)^{-1}$$

$$\widetilde{\mu}_v = \left(\frac{1}{\mu_v^{(1)}} + \sum_{r=2}^{r=3} c^{(r)}\left(\widetilde{b}_{d(e)}^{(r)} + \sum_{i=1}^{i=4}\widetilde{b}_{d(i)}^{(r)}\right)\left(\frac{1}{\mu_v^{(r)}} - \frac{1}{\mu_v^{(1)}}\right)\right)^{-1}.$$

As we have a relation between stress localization tensor in the different phases, namely :

$$\sum_{r=1}^{r=3} c^{(r)}\,\widetilde{b}_m^{(r)*}(p) = 1 \quad \text{and} \quad \sum_{r=1}^{r=3} c^{(r)}\,\widetilde{b}_d^{(r)*}(p) = 1,$$

we only need to calculate the stress localization tensor in the two inclusion phases. Also, because the two inclusion phases are interchangeable, we can easily deduce the expression of $\widetilde{b}_m^{(3)*}$ from $\widetilde{b}_m^{(2)*}$ and $\widetilde{b}_d^{(3)*}$ from $\widetilde{b}_d^{(2)*}$ by replacing phase (2) with (3) and phase (3) with (2).

In what follows, the 31 scalar coefficients:

$$\left(\widetilde{\tau}_{m(i)}\right)_{i=1,2},\ \left(\widetilde{\tau}_{d(i)}\right)_{i=1..4},\ \left(\widetilde{b}_{m(e)}^{(r)}, \left(\widetilde{b}_{m(i)}^{(r)}\right)_{i=1,2}\right)_{r=2,3},\ \left(\widetilde{b}_{d(e)}^{(r)}, \left(\widetilde{b}_{d(i)}^{(r)}\right)_{i=1..4}\right)_{r=2,3},$$
$$\left(\widetilde{m}_{m(i)}\right)_{i=1,2},\ \left(\widetilde{m}_{d(i)}\right)_{i=1..4},\ \widetilde{\mu}_e, \widetilde{\kappa}_e, \widetilde{\mu}_v$$

are given as a function of the phase volume fractions and their elastic and viscous properties.

The hydrostatic part of the stress localization tensor in the inclusion phases yields (more details can be found in Appendix A of [7]) :

$$\widetilde{b}_m^{(2)*}(p) = \widetilde{b}_{m(e)}^{(2)}\,\frac{\prod_{i=1}^{i=2}\left(p + \frac{1}{\tau_{m(i)}^{(2)}}\right)}{\prod_{i=1}^{i=2}\left(p + \frac{1}{\widetilde{\tau}_{m(i)}}\right)} \tag{10}$$

Introducing $\overline{\kappa}_e$ the volume average of the elastic bulks and $\hat{\kappa}_e$ as :

$$\overline{\kappa}_e = \sum_{r=1}^{r=3} c^{(r)}\kappa_e^{(r)},\ \hat{\kappa}_e = \sqrt{\kappa_e^{(1)}\kappa_e^{(2)}(c^{(1)} + c^{(2)}) + \kappa_e^{(1)}\kappa_e^{(3)}(c^{(1)} + c^{(3)}) + \kappa_e^{(2)}\kappa_e^{(3)}(c^{(2)} + c^{(3)})},$$

the elastic part $\widetilde{b}^{(2)}_{m(e)}$ and the numerator roots are given as :

$$\widetilde{b}^{(2)}_{m(e)} = \frac{\kappa_e^{(2)}(4\,\mu_e^{(1)} + 3\,\kappa_e^{(1)})\,(4\,\mu_e^{(1)} + 3\,\kappa_e^{(3)})}{16\,\left(\mu_e^{(1)}\right)^2 \overline{\kappa}_e + 12\,\mu_e^{(1)}\,(\hat{\kappa}_e)^2 + 9\,\kappa_e^{(1)}\kappa_e^{(2)}\kappa_e^{(3)}}$$

$$\tau^{(2)}_{m(1)} = \tau^{(1)}\left(1 + \frac{4\,\mu_e^{(1)}}{3\,\kappa_e^{(1)}}\right),\ \tau^{(2)}_{m(2)} = \tau^{(1)}\left(1 + \frac{4\,\mu_e^{(1)}}{3\,\kappa_e^{(3)}}\right) \quad \text{with} \quad \tau^{(1)} = \frac{\mu_v^{(1)}}{\mu_e^{(1)}}$$

while the two relaxation times are deduced from the two real roots $\left(-{}^{1}\!/_{\widetilde{\tau}_{m(i)}}\right)_{i=1,2}$ of the quadratic polynomial :

$$\begin{aligned} \left(\mu_v^{(1)}\right)^2 \left(16\,\left(\mu_e^{(1)}\right)^2 \overline{\kappa}_e + 12\,\mu_e^{(1)}\,(\hat{\kappa}_e)^2 + 9\,\kappa_e^{(1)}\kappa_e^{(2)}\kappa_e^{(3)}\right)\,p^2 & \\ +\mu_v^{(1)}\mu_e^{(1)}\left(12\,\mu_e^{(1)}\,(\hat{\kappa}_e)^2 + 18\,\kappa_e^{(1)}\kappa_e^{(2)}\kappa_e^{(3)}\right)\,p & \\ +9\,\left(\mu_e^{(1)}\right)^2 \kappa_e^{(1)}\kappa_e^{(2)}\kappa_e^{(3)} = 0 & \end{aligned} \tag{11}$$

Decomposing the rational fraction $\widetilde{b}^{(2)*}_m(p)$ in simple poles as in (5) we can then deduce the coefficients $\left(\widetilde{b}^{(2)}_{m(i)}\right)_{i=1,2}$. Injecting $\widetilde{b}^{(2)*}_m$ and $\widetilde{b}^{(3)*}_m$ in (8), we obtain by identification with equation (2) the expression of the $\left(\widetilde{m}_{m(i)}\right)_{i=1,2}$:

$$i = 1\ldots 2 : \widetilde{m}_{m(i)} = \frac{1}{3\,\widetilde{\tau}_{m(i)}} \sum_{r=2}^{r=3} c^{(r)}\widetilde{b}^{(r)}_{m(i)}\left(\frac{1}{\kappa_e^{(r)}} - \frac{1}{\kappa_e^{(1)}}\right)$$

The deviatoric part of the stress localization tensor for the phase (2) can be written as (more details can be found in Appendix A of [7]) :

$$\widetilde{b}^{(2)*}_d(p) = \widetilde{b}^{(2)}_{d(e)} \frac{\prod_{i=1}^{i=4}\left(p + \frac{1}{\tau^{(2)}_{d(i)}}\right)}{\prod_{i=1}^{i=4}\left(p + \frac{1}{\widetilde{\tau}_{d(i)}}\right)} \tag{12}$$

where the elastic part $\widetilde{b}^{(2)}_{d(e)}$ and two numerator roots $\left(\tau^{(2)}_{d(i)}\right)_{i=1,2}$ read :

$$\widetilde{b}^{(2)}_{d(e)} = \frac{5\,\mu_e^{(2)}(4\,\mu_e^{(1)} + 3\,\kappa_e^{(1)})\left(\mu_e^{(1)}(8\,\mu_e^{(1)} + 12\,\mu_e^{(3)}) + \kappa_e^{(1)}(9\,\mu_e^{(1)} + 6\,\mu_e^{(3)})\right)}{\mathrm{denom}(\widetilde{b}^{(2)}_{d(e)})},$$

$$\begin{aligned} \mathrm{denom}(\widetilde{b}^{(2)}_{d(e)}) = {} & 6\,\mu_e^{(2)}\mu_e^{(3)}\,(2\,\mu_e^{(1)} + \kappa_e^{(1)})((20 - 8\,c^{(1)})\mu_e^{(1)} + (15 - 9\,c^{(1)})\kappa_e^{(1)}) \\ & + \mu_e^{(1)}(8\,\mu_e^{(1)} + 9\,\kappa_e^{(1)})(6\,c^{(1)}(2\,\mu_e^{(1)} + \kappa_e^{(1)})(\mu_e^{(2)} + \mu_e^{(3)}) \\ & + (20\,\mu_e^{(1)} + 15\,\kappa_e^{(1)})(c^{(2)}\mu_e^{(2)} + c^{(3)}\mu_e^{(3)}) + c^{(1)}\mu_e^{(1)}(8\,\mu_e^{(1)} + 9\,\kappa_e^{(1)})), \end{aligned}$$

$$\tau^{(2)}_{d(1)} = \tau^{(1)} \left(1 + \frac{4\,\mu^{(1)}_e}{3\,\kappa^{(1)}_e}\right),\ \tau^{(2)}_{d(2)} = \tau^{(1)} \quad \text{with} \quad \tau^{(1)} = \frac{\mu^{(1)}_v}{\mu^{(1)}_e},$$

while $\left(-1/\tau^{(2)}_{d(i)}\right)_{i=3,4}$ are the two real roots of the quadratic polynomial :

$$\begin{aligned}\left(\mu^{(1)}_v\right)^2 \mu^{(3)}_v \left[(8\,\mu^{(1)}_e + 9\,\kappa^{(1)}_e)\,\mu^{(1)}_e + (12\,\mu^{(1)}_e + 6\,\kappa^{(1)}_e)\,\mu^{(3)}_e\right] p^2 \\ +\mu^{(1)}_v \mu^{(1)}_e \left[9\,\mu^{(1)}_e\,\kappa^{(1)}_e\,\mu^{(3)}_v + \left((8\,\mu^{(1)}_e + 9\,\kappa^{(1)}_e)\,\mu^{(1)}_v + 12\,(\mu^{(1)}_e + \kappa^{(1)}_e)\,\mu^{(3)}_v\right)\mu^{(3)}_e\right] p \\ +\kappa^{(1)}_e \left(\mu^{(1)}_e\right)^2 \mu^{(3)}_e \left(9\,\mu^{(1)}_v + 6\,\mu^{(3)}_v\right) = 0.\end{aligned}$$

The four relaxation times are deduced from the four real roots $\left(-1/\widetilde{\tau}_{d(i)}\right)_{i=1..4}$ of a quartic polynomial which is not given here because of its complexity but which can be given on demand. Decomposing the rational fraction $\widetilde{b}^{(2)*}_d(p)$ in simple poles as in (5) we can then deduce the coefficients $\left(\widetilde{b}^{(2)}_{d(i)}\right)_{i=1..4}$.

Injecting $\widetilde{b}^{(2)*}_d$ and $\widetilde{b}^{(3)*}_d$ in (9), we obtain by identification with equation (2) the expression of the $\left(\widetilde{m}_{d(i)}\right)_{i=1..4}$:

$$i = 1\ldots 4 : \widetilde{m}_{d(i)} = \frac{1}{2}\sum_{r=2}^{r=3} c^{(r)}\widetilde{b}^{(r)}_{d(i)} \left(\frac{1}{\widetilde{\tau}_{d(i)}}\left(\frac{1}{\mu^{(r)}_e} - \frac{1}{\mu^{(1)}_e}\right) - \left(\frac{1}{\mu^{(r)}_v} - \frac{1}{\mu^{(1)}_v}\right)\right).$$

2.2 Three-phase isotropic composites, case of a nonlinear viscoelastic behavior

For a nonlinear behavior, we adopt a secant linearization procedure in the three phases, the linearized behavior is defined by the shear modulus of the matrix and the inclusion phases:

$$\frac{1}{\mu^{(1)}_v(t)} = \frac{3}{\overline{\overline{\sigma}}^{(1)}_{eq}(t)} \frac{\partial U^{(1)}_v}{\partial \sigma_{eq}}(t,\, \overline{\overline{\sigma}}^{(1)}_{eq}(t)) \quad \text{and} \quad r = 2,3 : \ \frac{1}{\mu^{(r)}_v(t)} = \frac{3}{\overline{\sigma}^{(r)}_{eq}(t)} \frac{\partial U^{(r)}_v}{\partial \sigma_{eq}}(t,\, \overline{\sigma}^{(r)}_{eq}(t)). \tag{13}$$

It's emphasized that the shear modulus of the matrix phase is computed for the second-order stress moment over this phase. Additionally, as the dissipation potential depends only on the equivalent stress in the considered isotropic situation, only the $(ijij)$ trace of this second order moment is needed to calculate the shear modulus in the matrix. As the Hashin-Shtrikman model is used, the stress in the inclusion phases is homogeneous ($r = 2,3 :\ \overline{\overline{\sigma}}^{(r)}_{eq}(t) = \overline{\sigma}^{(r)}_{eq}(t)$). Therefore, the shear modulus of the inclusion phases can simply be computed for their averaged stresses. This property attached to Hashin-Shtrikman estimates have been used by [1] to compute the second-order stress moment in the matrix phase for two-phase incompressible Maxwellian composites by assuming that the hydrostatic stress field fluctuations can be neglected in the matrix. This former result can be simply extended when two families of inclusions are under consideration. Indeed, the scalar quantity $S^{(1)}$ related to the second-order stress moment

$$S^{(1)}(t) = \langle \boldsymbol{s}(\boldsymbol{x}\,,t) : \boldsymbol{s}(\boldsymbol{x}\,,t)\rangle^{(1)} = \frac{2}{3}\left(\overline{\overline{\sigma}}^{(1)}_{\text{eq}}(t)\right)^2 \tag{14}$$

is solution of the following first-order time differential equation :

$$\frac{1}{4\,\mu_e^{(1)}}\dot{S}^{(1)}(t)+\frac{1}{2\mu_v^{(1)}}\,S^{(1)}(t)=M(t), \tag{15}$$

where the right-hand member $M(t)$ is the following scalar time function :

$$M(t)=\frac{1}{c^{(1)}}\Bigg(\dot{\overline{\boldsymbol{\varepsilon}}}(t):\overline{\boldsymbol{\sigma}}(t)-\sum_{r=2}^{r=3}c^{(r)}\Big(\frac{1}{2\,\mu_e^{(r)}}\dot{\overline{\boldsymbol{s}}}^{(r)}(t):\overline{\boldsymbol{s}}^{(r)}(t)+\frac{1}{2\mu_v^{(r)}}\overline{\boldsymbol{s}}^{(r)}(t):\overline{\boldsymbol{s}}^{(r)}(t)\Big)$$
$$-\sum_{r=1}^{r=3}c^{(r)}\Big(\frac{1}{\kappa_e^{(r)}}\dot{\overline{\sigma}}_m^{(r)}(t)\,\overline{\sigma}_m^{(r)}(t)\Big)\Bigg). \tag{16}$$

2.3 Numerical implementation

The loading is defined by the time evolution of the macroscopic stress $\overline{\boldsymbol{\sigma}}(t)$ on the interval of time $[0;T]$. At $t=0$, this time function is nil. As a result, the mechanical fields as well as the three unknown time functions $\left(\overline{\overline{\sigma}}_{eq}^{(1)}(t),\,\overline{\boldsymbol{s}}^{(2)}(t),\,\overline{\boldsymbol{s}}^{(3)}(t)\right)$ are also nil at $t=0$. This system of three nonlinear differential equations can be solved by classical methods like the Euler-implicit scheme. The TFEL/MFRONT software (http://tfel.sourceforge.net/) was used for this integration.

3. RESULTS

The model proposed in the previous section is now applied to three-phase particulate composites with a moderate volume fraction of inclusions ($\leq 30\%$), whose microstructures can be idealized by the Hashin-Shtrikman model.

3.1 Material data

As [1] and [8], we consider a rate-dependent matrix reinforced by elastic inclusions. The dissipation potentials of the matrix is supposed to be a power-law:

$$U_v^{(1)}(\boldsymbol{s})=\frac{\dot{e}_0^{(1)}\,\sigma_0^{(1)}}{n^{(1)}+1}\left(\frac{[\sigma_{eq}-\sigma_Y^{(1)}]^+}{\sigma_0^{(1)}}\right)^{n^{(1)}+1} \tag{17}$$

where $[x]^+$ denotes the positive part of the scalar x while $\dot{e}_0^{(1)},\sigma_0^{(1)}$ and $n^{(1)}\geqslant 1$ are material coefficients which characterize the intensity of the creep rate. But with this new contribution, we take in consideration an additional phase. Due to the fabrication process or the aging of the composites, the elastic moduli of this third phase are significantly lower as compared to the ones of the reinforcements. These inclusions are called hereafter defective inclusions. The total volume fraction of inclusions (reinforcements and defective inclusions) equals 17% but we consider different configurations ranging from a perfect fabrication process (no defective inclusions, 17% of reinforcements) to a deficient one where all the reinforcements display defective elastic properties (no reinforcements, 17% of the defective inclusions). The predictions of the three-phase approach is therefore investigated when the reinforcements coexists with defective inclusions in the composite.

We are considering two cases with two distinct behaviors, a linear viscoelastic one with material data reported on table 1 as well as an elasto-viscoplastic with material data reported on table 2. In both cases, the behavior of the third phase (index (3)) is purely elastic with elastic moduli four times lower as compared to the ones of the reinforcements (index (2)). The particles (elastic reinforcements and defective inclusions) have a spherical shape with the same radius and are distributed isotropically.

$c^{(2)}+c^{(3)}$	$\mu_e^{(1)}$	$\kappa_e^{(1)}$	$\mu_e^{(2)}/\mu_e^{(1)} = \kappa_e^{(2)}/\kappa_e^{(1)}$	$\mu_e^{(3)}/\mu_e^{(1)} = \kappa_e^{(3)}/\kappa_e^{(1)}$	$\dot{e}_0^{(1)}$	$\sigma_Y^{(1)}$	$n^{(1)}$
0.17	3 GPa	10 GPa	2	0.5	$1\,s^{-1}$	0	1

Table 1. Data used for linear viscoelastic simulations.

$c^{(2)}+c^{(3)}$	$\mu_e^{(1)}$	$\kappa_e^{(1)}$	$\mu_e^{(2)}/\mu_e^{(1)} = \kappa_e^{(2)}/\kappa_e^{(1)}$	$\mu_e^{(3)}/\mu_e^{(1)} = \kappa_e^{(3)}/\kappa_e^{(1)}$	$\dot{e}_0^{(1)}$	$\sigma_Y^{(1)} = \sigma_0^{(1)}$	$n^{(1)}$
0.17	3 GPa	10 GPa	2	0.5	$1\,s^{-1}$	100 MPa	10/3

Table 2. Data used for elasto-viscoplastic simulations.

This 3-phases composite is submitted to the following strain-controlled isochoric loading:

$$\overline{\boldsymbol{\varepsilon}} = \overline{\varepsilon}_{33}(t)\left(-\frac{1}{2}\boldsymbol{e}_1 \otimes \boldsymbol{e}_1 - \frac{1}{2}\boldsymbol{e}_2 \otimes \boldsymbol{e}_2 + \boldsymbol{e}_3 \otimes \boldsymbol{e}_3\right)$$

As explained above, the time $t = 0$ is chosen such that for $t < 0$ the outer boundary of the RVE is stress free.

3.2 Full-field computations

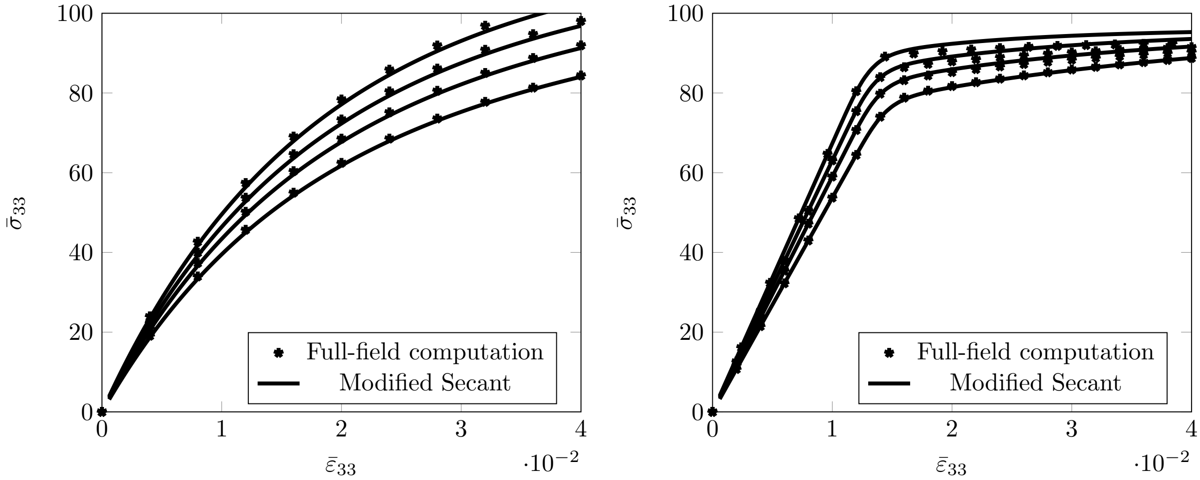
Full-field computations with periodic boundary conditions are used to assess the model. The RVE is a cubic domain which is generated using the random sequential addition (or adsorption, RSA) algorithm [9]. With this method, RVEs have been generated with a volume fraction of particles (elastic reinforcements and defective inclusions) equals to 17%.

Moreover, three realizations of this microstructure were generated in order to ensure that the dispersion of the simulated responses (macroscopic behavior, first and second order moments of the mechanical fields) is less than 0.5%. The computational method used for this analysis is based on fast Fourier transforms, originally proposed by [10]. Next simulated results are weakly dependent on the spatial resolution: when the number of voxels are increased from 64^3 to 512^3, the relative deviations between the simulated responses never exceed 1%. The results presented in this paper are obtained with 256^3 voxels which is a good compromise between computation times and accuracy.

3.3 Results

For a nil volume fraction of defective inclusions, as the inclusions are purely elastic with elastic moduli higher than the one of the matrix, it acts as reinforcements such that the effective response displays higher elastic properties as well as a higher yield stress when compared to the ones of the matrix. Conversely, substituting reinforcements by defective inclusions will decrease the overall answer of the composite.

We have reported on figures 1.a and 1.b, the overall strain - overall stress response at a given speed load (the prescribed strain rates equals $12\,10^{-3}$) for a linear viscoelastic and an elasto-viscoplastic composite behavior. For these two figures, the volume fraction of particle



a. Linear elastoplastic case b. Elasto-viscoplastic case

Figure 1. Macroscopic stress evolution with the prescribed macroscopic strain as predicted by the modified secant model and the full-field calculations as a function of defective inclusions. Material data are respectively given in table 1 and 2.

is kept constant (17%) but the fraction of defective inclusions varies from 0 (reinforcements only) to 17% (no more reinforcements). For a nil volume fraction of defective inclusions, the results agree perfectly well with the former results as given initially by [8] (FFT results) and [1] (the proposed model for a two-phase composite). As expected and for a linear or a nonlinear behavior, FFT simulations show that increasing the volume fraction of defective inclusions decreases the strain-stress response.

In addition and as expected, the predictions of the model agree remarkably well with the reference results when considering a linear viscoelastic behavior. For an elasto-viscoplastic behavior, the model predicts correctly the decrease of the strain-stress response as a function of the volume fraction of the defective inclusions. However, small deviations inherent to the chosen linearization procedure remains, especially for the full reinforcements case.

4. CONCLUSIONS

With this new contribution, we have extended the theoretical developments of [1] to the case of a dispersion of two families of particles in a matrix. In the particular case of non aging linear viscoelastic constituents (Maxwellian behaviour), the closed-form (temporal) expressions are the exact solution provided by the correspondence principle and the Hashin-Shtrikman estimate.

The model predictions have been compared to full-field computations by considering partially reinforced composites where a given fraction of the elastic reinforcements are defective and modelled by inclusions with lower elastic moduli. These estimates well predict the effective behaviour of the composite when the volume fraction of the defective inclusions increases.

Future works will be devoted to the computation of the full components of the stress fluctuations in the matrix in order to make use of an improved linearization procedure like the one proposed in [11]. This improvement seems mandatory to consider extreme situations like partially voided composites.

Appendix A Hashin-Shtrikman model for a three-phase linear viscoelastic composite

If the Hashin–Shtrikman model is used, the Laplace-Carson transforms of the shear and bulk components of the time relaxation function read:

$$\widetilde{\mu}^*(p) = \mu^{(1)*}(p) + \frac{\displaystyle\sum_{r=2}^{r=3} \frac{c^{(r)}\ \left(\mu^{(r)*}(p) - \mu^{(1)*}(p)\right)}{1 + 2\,\beta^*(p)\ \left(\mu^{(r)*}(p) - \mu^{(1)*}(p)\right)}}{c^{(1)} + \displaystyle\sum_{r=2}^{r=3} \frac{c^{(r)}}{1 + 2\,\beta^*(p)\ \left(\mu^{(r)*}(p) - \mu^{(1)*}(p)\right)}},\ \beta^*(p) = \frac{3\ \left(\kappa_e^{(1)} + 2\mu^{(1)*}(p)\right)}{5\,\mu^{(1)*}(p)\ \left(3\,\kappa_e^{(1)} + 4\,\mu^{(1)*}(p)\right)}$$

$$\widetilde{\kappa}^*(p) = \kappa_e^{(1)} + \frac{\displaystyle\sum_{r=2}^{r=3} \frac{c^{(r)}\ \left(\kappa_e^{(r)} - \kappa_e^{(1)}\right)}{1 + 3\,\alpha^*(p)\ \left(\kappa_e^{(r)} - \kappa_e^{(1)}\right)}}{c^{(1)} + \displaystyle\sum_{r=2}^{r=3} \frac{c^{(r)}}{1 + 3\,\alpha^*(p)\ \left(\kappa_e^{(r)} - \kappa_e^{(1)}\right)}},\ \alpha^*(p) = \frac{1}{3\,\kappa_e^{(1)} + 4\,\mu^{(1)*}(p)}$$

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