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Mohamed El Bachir Seck, Mihail Garajeu, Renaud Masson. Exact solutions for the effective nonlinear viscoelastic (or elasto-viscoplastic) behaviour of particulate composites under isotropic loading. European Journal of Mechanics - A/Solids, 2018, 10.1016/j.euromechsol.2018.04.018. hal-02482763

# HAL Id: hal-02482763 https://hal.science/hal-02482763v1

Submitted on 18 Feb 2020

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# Exact solutions for the effective nonlinear viscoelastic (or elasto-viscoplastic) behaviour of particulate composites under isotropic loading

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#### Abstract

We consider a composite sphere which consists of a spherical inclusion embedded in a concentric spherical matrix, the inclusion and matrix phases obeying an isotropic nonlinear viscoelastic behaviour. For different isotropic loadings (macroscopic stress or dilatation, swelling of the inclusion phase), the general solutions are shown to depend on the shear stress distribution in the matrix. This shear stress distribution is solution of a first-order nonlinear integro-differential equation, regardless of the inclusion viscoplastic behaviour. When the viscous strain rate potential in the matrix is a power-law function of the von Mises equivalent stress, closed-form solutions are given for some special cases clearly identified. Full-field calculations of representative volume elements of particulate composites are also reported. For a moderate volume fraction of inclusions, the composite sphere model turns out to be in excellent agreement with these full-field calculations.

Keywords: Nonlinear viscoelasticity, Elasto-viscoplasticity, Voids or inclusions, Two-phase composites, Composite sphere assemblage, Stress-free strain, Isotropic loading, FFT calculations

#### 1. Introduction

Many technological matrials are particulate composites (particles embeded in a matrix) in which the phases may undergo different stress-free strains (thermal dilatation, physicochemical evolution, phase transformation, irradiation effect, ...). This differential deformation of the phases induces internal stresses in the material, even for materials which are homogeneous elastically. If relaxation mechanisms like viscous (or viscoplastic) strains appear in the phases (effect of time, temperature, irradiation,...), this internal elastic stress field will relax heterogeneously. The internal stresses level as well as their time evolutions have to be known to assess the mechanical integrity of the considered composite. Thats why, even for particular situations, it would be of great interest to derive closed-form expressions of the time evolutions of stresses and strains of a nonlinear viscoelastic (or elasto-viscoplastic)

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composite submitted to differential stress-free strains. This is precisely the goal of this paper to yield analytical results and hence to allow a better understanding of the local distribution and time-evolution of the mechanical fields inside these particulate composites. Otherwise, as explained hereafter, these closed-form expressions will provide a reference solution to challenge homogenization methods for a class of particular microstructure (the composite sphere assemblage).

The particulate composites of interest in this study have a small or moderate volume fraction of inclusions (less than 30%) and therefore the interactions between inclusions are weak. In this case, classically, it is idealized by considering spherical inclusions with a gradation in size (composite sphere assemblage, [1]) and such that the ratio of the inclusions and matrix radi remain constant for all size of inclusions (self-similar spheres). Moreover, the distribution of inclusions is such that a volume filling configuration is obtained. Following [1], the effective behaviour of such a microstructure is well approached by that of a composite sphere with the same volume fractions of the phases. The analytical results mentioned above are obtained by solving the mechanical problem on this simplified volume element.

For general microstructures, for which only some statistical informations are available, when the constituents display a linear elastic behaviour, homogenization methods provide bounds or estimates of the effective modulus (or compliance) tensor. For instance, the "Self-consistent" model [2] provides reliable estimates for polycristalline microstructures while the "Mori-Tanaka" one [3] is well-suited to particle reinforced composites with a low volume fraction of particles.

With the help of the so called correspondence principle [4], these models can be easily extend to linear viscoelastic composite materials to estimate the effective creep or relaxation functions. When (at least) one of the constituents of the composite material displays a nonlinear viscoelastic behaviour, several approaches have been proposed. The approach proposed by [5] consists in considering the viscous strains as homogeneous stress free strains, the associated first moment of the stress field in the phases being solutions of a linear thermoelastic problem. To improve these too stiff estimates, the "affine method" [6] is based on the solution of a linear thermo-viscoelastic problem. An alternative approach, using the correspondence principle and internal variables formulation was proposed by [7] in order to estimate the effective response of ageing viscoelastic composites.

More recently, variational approaches ([8], [9]) classically used for behaviours deriving from a single potential (nonlinear elasticity or viscosity) were extended by [10] to nonlinear elastic-viscoplastic (and elastic-plastic) behaviours (which derive from two potentials). Based on the first and second moments of the fields, this approach has been shown to yield satisfactory estimates unlike "classical" mean-fields approaches based on the first moment of the fields.

Otherwise, analytical results obtained for particular microstructures are useful and it can be used to improve estimates derived by other homogenization methods. This is the case for the Eshelby's result [11], which gives the localization tensor of an ellipsoidal region embedded in an infinite linear elastic matrix. Eshelby's result is used in many others models which aim to estimate the effective behaviour of composites with a particle-matrix microstructure, with a low volume fraction of particles (as in the "Mori-Tanaka" model [3]).

Composite sphere volume element is extensively used in micromechanics due to the facility to derive analytical results for this simple microstructure. When the inclusions and the matrix

obey a linear elastic behaviour, the effective properties of the composite can be bounded by solving the composite sphere problem: a spherical inclusion surrounded by a spherical layer of matrix ([1]). For voids or rigid particles surrounded by a viscoplastic matrix, bounds have been derived by considering a similar problem. In [12] or, more recently, in [13] the analytical results obtained for a hollow sphere have been used in order to improve variational bounds for viscoplastic porous media.

In porous plasticity, Gurson [14] uses the hollow sphere with a von Mises matrix, to derive its well known criterion. Gurson analysis was extended to porous materials containing align ellipsoidal voids by [15], [16], [17], [18]. Exact calculations by [19] carried out on a hollow sphere lead to a plasticity criterion which presents a third invariant effect. Additional results were obtained by [20] for elastic-plastic matrix and by [21], [22], when the matrix phase is pressure-sensitive. In addition, the composite sphere problem has also been solved for elastic-plastic constituents behaviour (see [23]). This solution is consistent with the particular one (when inclusions are voids) given by [24].

For linear viscoelasticity, estimates have been derived by using the composite sphere model and the correspondence principle ([25]). Here, we aim at solving the composite sphere problem when the two phases display a nonlinear viscoelastic behaviour. To derive exact results, we will limit these analytical developments to isotropic loading. This isotropic loading will consist in a purely mechanical loading as dilatation or tension applied to the outer surface of the composite combined with a thermo-mechanical loading as a thermal strain mismatch between the two phases, appearing with temperature variations (thermomechanical loadings of structures, fabrication processes, etc. ). The limit case of an incompressible matrix or of a composite sphere with homogeneous compressibility will be derived explicitly as well. The following theoretical developments are limited to infinitesimal strains ( $\varepsilon$  and  $\sigma$  denote the infinitesimal strain and Cauchy stress tensors, respectively).

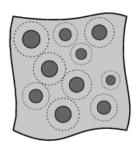
The paper is organized as follows. In section 2, the composite sphere problem is formulated. The local fields in the inclusion and the equations for the local fields in the matrix are obtained in section 3. Section 4 presents the exact solutions obtained in the particular cases mentioned above. The predictions of the model are compared to the ones given by full-field computations of random particulate composites in section 5.

#### 2. The Composite sphere model

We consider a composite sphere (domain V) of external radius b containing in its center a non linear viscoelastic spherical inclusion of radius a (domain 2), embedded in a nonlinear viscoelastic matrix (domain 1). As shown by [1], the effective elastic potential of such a simplified volume element is a bound for the elastic potential of more complex microstructures, obtained by spheres assemblage (Figure 1).

In what follows indexes (1) and (2) are used for the mechanical fields and coefficients in the matrix and in the inclusion, respectively. Hence,  $\chi_1(\boldsymbol{x})$  and  $\chi_2(\boldsymbol{x})$  are the characteristic functions of domains 1 and 2, respectively, and the volume fractions of the matrix and of the inclusion are denoted by  $c_1$  and  $c_2$ ,  $c_1 = \langle \chi_1(\boldsymbol{x}) \rangle_V$ ,  $c_2 = \langle \chi_2(\boldsymbol{x}) \rangle_V = \frac{a^3}{b^3}$  and  $c_1 + c_2 = 1$  ( $\langle \rangle_V$  is the average operator on V).

The composite sphere is loaded isotropically on the outer boundary and by submitting an uniform isotropic stress-free strain to the inclusion. This stress-free strain may correspond



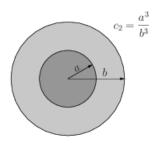


Figure 1: The composite spheres assemblage

Figure 2: The composite sphere

to a thermal strain mismatch between the two phases. On the outer boundary, two kinds of loadings, which generalise relaxation and creep tests, are considered:

- homogeneous strain :  $\boldsymbol{u}(\boldsymbol{x},t) = E_m^0(t).\boldsymbol{x}$  for  $\boldsymbol{x} \in \partial V$ ,
- homogeneous stress :  $\sigma(x,t).n(x) = \Sigma_m^0(t).n(x)$  for  $x \in \partial V$ ,

Assuming the small strain hypothesis, the decomposition of the total strain reads:

$$\boldsymbol{\varepsilon}(\boldsymbol{x},t) = \boldsymbol{\varepsilon}^{e}(\boldsymbol{x},t) + \boldsymbol{\varepsilon}^{v}(\boldsymbol{x},t) + \varepsilon_{0}^{(2)}(t)\chi_{2}(\boldsymbol{x})\boldsymbol{\delta}, \tag{1}$$

where  $\boldsymbol{\varepsilon}^e$  and  $\boldsymbol{\varepsilon}^v$  are the elastic and viscous strains and  $\varepsilon_0^{(2)}(t)\boldsymbol{\delta}$  is the uniform and spherical stress-free strain applied to the inclusion ( $\boldsymbol{\delta}$  is the second order identity tensor).

The non linear viscous behaviour considered in this paper derives from a potential  $w(x, \sigma)$ :

$$\dot{\boldsymbol{\varepsilon}}^{v} = \frac{\partial w}{\partial \boldsymbol{\sigma}} \quad \text{with} \quad w(\boldsymbol{x}, \underline{\sigma}) = w_{1}(\underline{\sigma})\chi_{1}(\boldsymbol{x}) + w_{2}(\underline{\sigma})\chi_{2}(\boldsymbol{x})$$
 (2)

where  $w_1$  and  $w_2$  are the dissipation potentials (convex functions) of the matrix and of the inclusion respectively, which depend only on the square of equivalent stress  $\tilde{\sigma} = \sigma_{eq}^2$ . The mean and equivalent stresses  $\sigma_m$  and  $\sigma_{eq}$  are defined as usually:

$$\sigma_m = \frac{1}{3} \text{Tr}(\boldsymbol{\sigma}), \quad \boldsymbol{s} = \boldsymbol{\sigma} - \sigma_m \boldsymbol{\delta}, \quad \sigma_{eq} = \sqrt{\frac{3}{2} \boldsymbol{s} : \boldsymbol{s}},$$
 (3)

where s denotes the stress deviator.

Then, by derivation of (2) we get

$$\dot{\boldsymbol{\varepsilon}}^{v}(\boldsymbol{x},t) = 3\boldsymbol{s}(\boldsymbol{x},t) \frac{\partial w}{\partial \underline{\sigma}} \left( \boldsymbol{x}, \underline{\sigma}(\boldsymbol{x},t) \right)$$
(4)

and the behaviour law in the whole domain V can be written as:

$$\dot{\boldsymbol{\varepsilon}}(\boldsymbol{x},t) = \frac{\dot{\sigma}_m(\boldsymbol{x},t)}{3\kappa(\boldsymbol{x})}\boldsymbol{\delta} + \frac{\dot{\boldsymbol{s}}(\boldsymbol{x},t)}{2\mu(\boldsymbol{x})} + 3\boldsymbol{s}(\boldsymbol{x},t)\frac{\partial w}{\partial \underline{\sigma}}(\boldsymbol{x},\underline{\sigma}(\boldsymbol{x},t)) + \dot{\varepsilon}_0^{(2)}(t)\chi_2(\boldsymbol{x})\boldsymbol{\delta}, \tag{5}$$

where  $\kappa(\mathbf{x})$  and  $\mu(\mathbf{x})$  are the elastic bulk and shear moduli, constant in each phase:

$$\kappa(\boldsymbol{x}) = \kappa_1 \chi_1(\boldsymbol{x}) + \kappa_2 \chi_2(\boldsymbol{x}), \qquad \mu(\boldsymbol{x}) = \mu_1 \chi_1(\boldsymbol{x}) + \mu_2 \chi_2(\boldsymbol{x}). \tag{6}$$

Consistently with the isotropic loading and the isotropic behaviour of the phases, we adopt spherical coordinates  $(r, \theta, \varphi)$  ( $e_r$  denotes the unit vector in a radial direction). The general form of the velocity field in the whole sphere is:

$$\dot{\boldsymbol{u}}(\boldsymbol{x},t) = \dot{\boldsymbol{u}}(r,t)\boldsymbol{e}_r \tag{7}$$

Therefore, the strain rate field and (from equation (5)) the stress field are both diagonal (in the spherical coordinates basis) and  $\dot{\varepsilon}_{\varphi\varphi}(r,t) = \dot{\varepsilon}_{\theta\theta}(r,t)$  and  $\sigma_{\varphi\varphi}(r,t) = \sigma_{\theta\theta}(r,t)$ . In that particular situation, the equilibrium equation reads:

$$\frac{\partial}{\partial r} \left( \sigma_{rr}(r,t) \right) + \frac{2}{r} \left( \sigma_{rr}(r,t) - \sigma_{\theta\theta}(r,t) \right) = 0. \tag{8}$$

The deviatoric parts of the strain rate and the stress, denoted by  $\dot{e}$  and s, are co-linear with the same second order deviatoric tensor d:

$$\dot{\boldsymbol{e}} = \frac{\dot{\varepsilon}_{rr} - \dot{\varepsilon}_{\theta\theta}}{3} \boldsymbol{d}, \qquad \boldsymbol{s} = \frac{\sigma_{rr} - \sigma_{\theta\theta}}{3} \boldsymbol{d} \qquad \text{with} \qquad \boldsymbol{d} = \begin{pmatrix} 2 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & -1 \end{pmatrix}. \tag{9}$$

In addition, the equivalent stress reads  $\sigma_{eq}(r,t) = |\sigma_{\theta\theta}(r,t) - \sigma_{rr}(r,t)|$ . Let  $\sigma_s$  denotes the signed equivalent stress,  $\sigma_s(r,t) = \sigma_{\theta\theta}(r,t) - \sigma_{rr}(r,t)$ . Then  $\sigma_s^2 = \sigma_{eq}^2 = \underline{\sigma}$  and  $s = -\frac{\sigma_s}{3}d$ .

# 2.1. Effective response

Both boundary conditions are isotropic and therefore, due to the macroscopic isotropy of the composite sphere, the effective stress and strain tensors are spherical:

$$\Sigma(t) = \Sigma_m(t)\delta, \qquad E(t) = E_m(t)\delta.$$
 (10)

Macroscopic stress and strain are usually defined by average relations, but in the case of a spherical volume element it is more convenient to use equivalent definitions:

$$\Sigma(t) = \frac{1}{|V|} \int_{\partial V} \boldsymbol{\sigma}(t) \cdot \boldsymbol{n} \otimes_s \boldsymbol{x} \, dS, \qquad \boldsymbol{E}(t) = \frac{1}{|V|} \int_{\partial V} \boldsymbol{u}(t) \otimes_s \boldsymbol{n} \, dS.$$
 (11)

For a spherical volume  $\mathbf{n} = \mathbf{e}_r$ ,  $\mathbf{x} = b\mathbf{e}_r$  and  $\int_{\partial V} \mathbf{e}_r \otimes_s \mathbf{e}_r \, \mathrm{d}S = \frac{4\pi b^2}{3} \boldsymbol{\delta}$  and in the present case  $\mathbf{u}(t) = u(b,t)\mathbf{e}_r$ ,  $\boldsymbol{\sigma}(t).\mathbf{n} = \sigma_{rr}(b,t)\mathbf{e}_r$ . Therefore,

$$\Sigma(t) = \Sigma_m(t)\delta = \sigma_{rr}(b,t)\delta$$
 and  $E(t) = E_m(t)\delta = \frac{u(b,t)}{b}\delta$ . (12)

#### 2.2. The purely elastic solution

The problem is that of a composite sphere made by two elastic phases under isotropic loadings. As shown by [1], the effective compressibility is given by:

$$\tilde{k} = \frac{3 \kappa_1 \kappa_2 + 4 \mu_1 (c_1 \kappa_1 + c_2 \kappa_2)}{3 c_2 \kappa_1 + 3 c_1 \kappa_2 + 4 \mu_1}.$$
(13)

The displacement and stress fields of this composite sphere under an isotropic traction  $E_m^0$  and with a uniform and spherical stress-free strain  $\varepsilon_0^{(2)}$  prescribed in the inclusion are well known:

• the displacement field:

$$\begin{cases} u(r) = \frac{(3\kappa_2 + 4\mu_1)E_m^0 - 3c_2\kappa_2\varepsilon_0^{(2)}}{3c_2\kappa_1 + 3c_1\kappa_2 + 4\mu_1} r + \frac{(\kappa_1 - \kappa_2)E_m^0 + \kappa_2\varepsilon_0^{(2)}}{3c_2\kappa_1 + 3c_1\kappa_2 + 4\mu_1} \frac{3a^3}{r^2} & \text{in 1} \\ u(r) = \frac{(3\kappa_1 + 4\mu_1)E_m^0 + 3c_1\kappa_2\varepsilon_0^{(2)}}{3c_2\kappa_1 + 3c_1\kappa_2 + 4\mu_1} r & \text{in 2} \end{cases}$$
(14)

• the radial stress:

$$\begin{cases}
\sigma_{rr}(r) = 3\kappa_1 \frac{(3\kappa_2 + 4\mu_1) E_m^0 - 3c_2 \kappa_2 \varepsilon_0^{(2)}}{3c_2 \kappa_1 + 3c_1 \kappa_2 + 4\mu_1} - 12\mu_1 \frac{(\kappa_1 - \kappa_2) E_m^0 + \kappa_2 \varepsilon_0^{(2)}}{3c_2 \kappa_1 + 3c_1 \kappa_2 + 4\mu_1} \left(\frac{a}{r}\right)^3 & \text{in } 1 \\
\sigma_{rr}(r) = 3\kappa_2 \frac{(3\kappa_1 + 4\mu_1) E_m^0 - (3c_2 \kappa_1 + 4\mu_1) \varepsilon_0^{(2)}}{3c_2 \kappa_1 + 3c_1 \kappa_2 + 4\mu_1} & \text{in } 2
\end{cases}$$
(15)

• while the equivalent stress is nonzero only in 1:

$$\sigma_{eq}(r) = 18\mu_1 \left(\frac{a}{r}\right)^3 \frac{\left| (\kappa_1 - \kappa_2) E_m^0 + \kappa_2 \varepsilon_0^{(2)} \right|}{3 c_2 \kappa_1 + 3 c_1 \kappa_2 + 4 \mu_1}$$
(16)

It is emphasized that the stress-free strain of the composite sphere (denoted by  $\tilde{\varepsilon}_m^0$ ) is given by:

$$\sigma_{rr}(b) = 0 \qquad \to \qquad \tilde{\varepsilon}_m^0 = \frac{c_2 \kappa_2 (3 \kappa_1 + 4 \mu_1)}{3 \kappa_1 \kappa_2 + 4 \mu_1 (c_1 \kappa_1 + c_2 \kappa_2)} \varepsilon_0^{(2)}$$
 (17)

If the inclusion is still submitted to a stress-free strain  $\varepsilon_0^{(2)}$  but a traction  $\Sigma_m^0$  is now prescribed on the outer boundary of the composite sphere, the displacement and stress fields are given by substituting the overal strain  $E_m^0$  by  $(\frac{\Sigma_m^0}{3\,\tilde{k}}+\tilde{\varepsilon}_m^0)$  in the previous equations. In this case the equivalent stress in 1 reads:

$$\sigma_{eq}(r) = 6\mu_1 \left(\frac{a}{r}\right)^3 \frac{\left|3\kappa_1\kappa_2\varepsilon_0^{(2)} + \Sigma_m^0 \left(\kappa_1 - \kappa_2\right)\right|}{3\kappa_1\kappa_2 + 4\mu_1\left(c_1\kappa_1 + c_2\kappa_2\right)}.$$
(18)

#### 2.3. Initial conditions

For general loading conditions, the time t=0 is chosen such that for t<0 the outer boundary of the sphere composite is stress free and the stress-free strain is zero. However, creep (or relaxation) tests correspond to a constant stress (or strain) loading. In that particular situations, the stress (or strain) loading is applied instantaneously at t=0 and it is maintained constant for t>0.

This instantaneous loading physically corresponds to a loading applied during a time interval sufficiently small that one can neglect the viscous strain in the relation (1). As a result, the instantaneous response at  $t = 0_+$  is purely elastic and the corresponding elastic solution is given in the previous section.

#### 3. The general solution

#### 3.1. Local equations

Considering the deviatoric parts of (5) it follows:

$$(\dot{\varepsilon}_{rr} - \dot{\varepsilon}_{\theta\theta})\boldsymbol{d} = -\frac{\dot{\sigma}_s}{2\mu(r)}\boldsymbol{d} - 3\sigma_s \frac{\partial w}{\partial g}\boldsymbol{d}, \tag{19}$$

where  $\mu(r) = \mu_2$  if r < a and  $\mu(r) = \mu_1$  if r > a. Using the compatibility of the strain rate tensor, the following differential equation for the stress  $\sigma_s$  is obtained:

$$\dot{\sigma}_s(r,t) + 6\mu(r)\,\sigma_s(r,t)\,\frac{\partial w}{\partial g}\left(r,\sigma_s^2(r,t)\right) = -2\mu(r)\,\left[r\frac{\partial}{\partial r}\left(\frac{\dot{u}(r,t)}{r}\right)\right].\tag{20}$$

Considering now the spherical parts in (5) we get (after time integration):

$$\varepsilon_m(r,t) = \frac{1}{3\kappa(r)}\sigma_m(r,t) + \varepsilon_0^{(2)}(t)\chi_2(r)$$
(21)

where  $\kappa(r) = \kappa_2$  if r < a and  $\kappa(r) = \kappa_1$  if r > a. Then, the compatibility of the strain tensor and the equilibrium equation (8) leads to the following differential equation for  $\sigma_{rr}$ :

$$\frac{\partial}{\partial r} \left( r^3 \sigma_{rr}(r, t) \right) = 3\kappa(r) \left[ \frac{\partial}{\partial r} \left( r^2 u(r, t) \right) - 3r^2 \varepsilon_0^{(2)}(t) \chi_2(r) \right]. \tag{22}$$

This differential equation can be easily integrated to give the expression of the radial stress in terms of radial displacement. Taking into account that u(0,t) = 0, we obtain:

$$\sigma_{rr}(r,t) = \begin{cases} 3\kappa_1 \left(\frac{u(r,t)}{r} - \lambda(t)\frac{b^3}{r^3}\right), & \text{in } 1\\ 3\kappa_2 \left(\frac{u(r,t)}{r} - \varepsilon_0^{(2)}(t)\right). & \text{in } 2 \end{cases}$$
 (23)

The unknown time function  $\lambda(t)$  is defined by:

$$\lambda(t) = \frac{u(b,t)}{b} - \frac{\sigma_{rr}(b,t)}{3\kappa_1}. (24)$$

This dimensionless time function sounds physically as an additional strain appearing when the composite sphere is inhomogeneous  $(c_2 \neq 0)$ .

From (23) and the equilibrium equation, the radial distribution of the stress  $\sigma_s$  reads:

$$\frac{2}{3}\sigma_s(r,t) - 3\kappa_1\lambda(t)\frac{b^3}{r^3}\chi_1(r) = \kappa(r)\ r\frac{\partial}{\partial r}\left(\frac{u(r,t)}{r}\right)$$
 (25)

After time derivation of (25) and using (20) it follows:

$$\left[1 + \frac{4\mu(r)}{3\kappa(r)}\right]\dot{\sigma}_s + 6\mu(r)\frac{\partial w}{\partial g} = 6\mu(r)\dot{\lambda}(t)\frac{\kappa_1\chi_1(r)}{\kappa(r)}\left(\frac{b^3}{r^3}\right). \tag{26}$$

This is a differential equation with respect to time for the stress  $\sigma_s$ , which is non homogeneous in the matrix phase. In order to integrate this equation an initial value  $\sigma_s(r, 0_+)$  has to be prescribed. For all fields involved in this problem, their initial values are given by the corresponding elastic fields (section 2.3).

#### 3.2. Local fields in the inclusion

In the inclusion 2, the equation (26) specializes to an homogeneous ordinary differential equation

 $\left(1 + \frac{4\mu_2}{3\kappa_2}\right)\dot{\sigma}_s^{(2)}(r,t) + 6\mu_2\sigma_s^{(2)}(r,t)\frac{\partial w_2}{\partial g}\left(g^{(2)}(r,t)\right) = 0,$ (27)

with initial condition  $\sigma_s^{(2)}(r, 0_+) = 0$  (since  $\sigma_{eq} = |\sigma_s|$ ). Under appropriate hypothesis on the potential  $w_2$  (for instance if the second derivative of  $w_2$  exists and it is continuous),  $\sigma_s^{(2)}(r,t) = 0$  is the unique solution of this problem. This implies that the stress  $\boldsymbol{\sigma}$  is a spherical tensor ( $\sigma_{rr} = \sigma_{\theta\theta}$ ) and from the behaviour law (5) the strain rate and the strain are also spherical tensors. Therefore the displacement field has a linear dependence on r. Finally, in the inclusion the following relations holds:

$$\begin{cases} u^{(2)}(r,t) = r C(t) \\ \sigma_{rr}^{(2)}(r,t) = \sigma_m^{(2)}(r,t) = 3 \kappa_2 \left( C(t) - \varepsilon_0^{(2)}(t) \right) \\ \sigma_s^{(2)}(r,t) = \sigma_{eq}^{(2)}(r,t) = 0 \end{cases}$$
(28)

where C(t) is an unknown function.

It is worth noting that the nullity of the equivalent stress in the inclusion leads to a lack of nonlinear effects in the inclusion. This is due to the particular loadings considered here which are all of spherical symmetry.

The function C(t) occurring in (28) is related to the function  $\lambda(t)$  of (23) by the continuity conditions at the phases interface, which, using (28) and (23), read:

$$u^{(1)}(a,t) = aC(t), \qquad 3\kappa_2 \left( C(t) - \varepsilon_0^{(2)}(t) \right) = 3\kappa_1 \left( \frac{u^{(1)}(a,t)}{a} - \frac{\lambda(t)}{c_2} \right). \tag{29}$$

Finally it holds:

$$\lambda(t) = -c_2 \left[ \frac{\kappa_2 - \kappa_1}{\kappa_1} C(t) - \frac{\kappa_2}{\kappa_1} \varepsilon_0^{(2)}(t) \right]. \tag{30}$$

# 3.3. The macroscopic stress - dilatation relation

As a result of the equations (12)) and (23), the relation between the macroscopic stress and the macroscopic strain reads:

$$\Sigma_m(t) = \sigma_{rr}(b, t) = 3\kappa_1 \left( \frac{u(b, t)}{b} - \lambda(t) \right) = 3\kappa_1 \left( E_m(t) - \lambda(t) \right)$$
(31)

By integration of the equilibrium equation it follows

$$\sigma_{rr}^{(1)}(b,t) - \sigma_{rr}^{(1)}(a,t) = \int_{a}^{b} \frac{2}{r} \sigma_{s}^{(1)}(r,t) dr.$$
 (32)

The continuity of the stress vector at the interface between phases and the known radial stress in the inclusion (28) leads to the following relation:

$$3\kappa_1 \left( E_m(t) - \lambda(t) \right) = 3\kappa_2 \left( C(t) - \varepsilon_0^{(2)}(t) \right) + \int_a^b \frac{2}{r} \sigma_s^{(1)}(r, t) dr$$
 (33)

From (30), C(t) can be substituted in the previous relation and the resulting expression for  $\lambda(t)$  is:

$$\lambda(t) = \frac{c_2(\kappa_2 - \kappa_1)}{c_2\kappa_1 + c_1\kappa_2} \left\{ -E_m(t) + \frac{\kappa_2}{\kappa_2 - \kappa_1} \varepsilon_0^{(2)}(t) + \frac{1}{3\kappa_1} \int_a^b \frac{2}{r} \sigma_s^{(1)}(r, t) dr \right\}$$
(34)

Finally, from (31) and (34), the macroscopic mean stress is obtained in terms of the macroscopic mean strain  $E_m(t)$ , of the stress-free strain  $\varepsilon_0^{(2)}(t)$  prescribed in the inclusion and of the radial distribution of the stress  $\sigma_s^{(1)}$  in the matrix:

$$\Sigma_m(t) = \frac{3\kappa_1 \kappa_2}{c_2 \kappa_1 + c_1 \kappa_2} (E_m(t) - c_2 \varepsilon_0^{(2)}(t)) - \frac{c_2 (\kappa_2 - \kappa_1)}{c_2 \kappa_1 + c_1 \kappa_2} \int_a^b \frac{2}{r} \sigma_s^{(1)}(r, t) dr.$$
 (35)

Alternatively, we can introduce in this last expression the effective elastic compressibility (13) as well as the overall stress-free swelling (17) so that the macroscopic strain-stress relation reads:

$$\Sigma_m(t) = 3\,\tilde{\kappa} \left( E_m(t) - \tilde{\epsilon}_m^0(t) - \epsilon_m^v(t) \right). \tag{36}$$

The macroscopic strain  $\epsilon_m^v(t)$  is such that (35) and (36) are equivalent:

$$\epsilon_{m}^{v}(t) = \frac{\frac{4}{3}c_{2}(\kappa_{1} - \kappa_{2})}{(\kappa_{1}\kappa_{2} + \frac{4}{3}\mu_{1}(c_{1}\kappa_{1} + c_{2}\kappa_{2}))(c_{2}\kappa_{1} + c_{1}\kappa_{2})} \left(c_{1}\mu_{1}\left((\kappa_{1} - \kappa_{2})E_{m}(t) + \kappa_{2}\epsilon_{0}^{(2)}(t)\right)\right) (37)$$
$$-(c_{2}\kappa_{1} + c_{1}\kappa_{2} + \frac{4}{3}\mu_{1})\int_{a}^{b} \frac{\sigma_{s}^{(1)}(r, t)}{2r} dr\right).$$

By substituting the (signed) von Mises equivalent stress distribution by the elastic solution (16), one can easily show that this macroscopic strain vanishes for a purely elastic behaviour. This last remark makes clear the physical meaning of the macroscopic strain  $\epsilon_m^v$ : this strain is the macroscopic inelastic strain due to the viscous behaviour of the matrix phase. For a given loading, the time evolution of this inelastic strain and the overall stress-strain relation (35) depend on the radial distribution of the signed equivalent stress in the matrix phase. This distribution is given in the next section 3.4.

It is worth noting that when the two phases have the same compressibility modulus,  $\kappa_1 = \kappa_2 = \tilde{\kappa}$ , the viscous macroscopic strain  $\epsilon_m^v$  vanishes so that the macroscopic strain-stress behaviour is purely elastic<sup>1</sup>:

$$\Sigma_m(t) = 3\,\tilde{\kappa}\,(E_m(t) - \tilde{\varepsilon}_m^0(t)). \tag{38}$$

# 3.4. Radial distribution of the equivalent stress in the matrix

In the matrix, the signed equivalent stress field, as given by (26), obeys to a non homogeneous ordinary differential equation:

$$\dot{\sigma}_s^{(1)}(r,t) + \frac{1}{\eta} \sigma_s^{(1)}(r,t) \frac{\partial w_1}{\partial g} (g^{(1)}(r,t)) = \frac{1}{\eta} \dot{\lambda}(t) \left(\frac{b^3}{r^3}\right)$$
(39)

<sup>&</sup>lt;sup>1</sup>From (17), the expression of the macroscopic stress-free strain is  $\tilde{\epsilon}_m^0(t) = c_2 \, \varepsilon_0^{(2)}(t)$  when  $\kappa_1 = \kappa_2$ .

where  $\eta = \frac{3\kappa_1 + 4\mu_1}{18\kappa_1\mu_1}$  and the time function  $\lambda(t)$  is given by the relation (34). Finally, whatever the loading is, the equation (39) for the stress  $\sigma_s^{(1)}$  in the matrix is an integro-differential equation of the form

$$\dot{\sigma}_s^{(1)}(r,t) + \frac{1}{\eta} \sigma_s^{(1)}(r,t) \frac{\partial w_1}{\partial \underline{\sigma}}(\underline{\sigma}^{(1)}(r,t)) = \left(\frac{b}{r}\right)^3 \left[\frac{2\alpha}{\eta} \int_a^b \frac{\sigma_s^{(1)}(u,t)}{u} \frac{\partial w_1}{\partial \underline{\sigma}}(\underline{\sigma}^{(1)}(u,t)) du + \mathcal{F}(t)\right],\tag{40}$$

with  $\alpha$  a dimensionless coefficient depending on the elastic moduli of the phases and  $\mathcal{F}(t)$  a function depending on the loading functions (the expressions of  $\alpha$  and  $\mathcal{F}(t)$  are given in the Appendix A, for both cases of loadings). The initial condition for this differential equation is given by the elastic distribution of the equivalent stress (16).

In the section 4, exact solutions of the equation (40) are derived in the case where the dissipation potential  $w_1$  is a power-law and for some particular cases of loadings.

#### 3.5. First and second moments of the stress field

As explained previously, the deviatoric part of the averaged stresses per phases is zero in that isotropic situation. In addition, the averaged stress in the inclusion is given by (28) as a function of the scalar function of time C(t). Substituting C(t) as a function of  $\lambda(t)$  (relation (30)) gives the time evolution of the averaged stress in the inclusion:

$$\sigma_m^{(2)}(t) = \frac{3 \kappa_1 \kappa_2}{\kappa_2 - \kappa_1} \left( \varepsilon_0^{(2)}(t) - \frac{1}{c_2} \lambda(t) \right) \tag{41}$$

where  $\lambda(t)$  is given by (34) for a macroscopic strain prescribed on the outer boundary  $\partial V$  of the composite sphere. Finally, we obtain:

$$\sigma_m^{(2)}(t) = \frac{3 \kappa_1 \kappa_2}{c_2 \kappa_1 + c_1 \kappa_2} \left( E_m(t) - c_2 \varepsilon_0^{(2)}(t) - \frac{1}{3 \kappa_1} \int_a^b \frac{2}{u} \sigma_s^{(1)}(u, t) du \right) \tag{42}$$

the radial distribution of the signed equivalent stress being solution of the differential equation (40). If a stress is now prescribed to the outer boundary of the composite sphere, the time evolution of the averaged stress in the inclusion is given by substituting in this last relation the macroscopic strain  $E_m(t)$  by  $\frac{1}{3\tilde{\kappa}} \Sigma_m(t) + \tilde{\varepsilon}_m^0(t) + \varepsilon_m^v(t)$  (relation (36)). It's worth emphasizing that the time evolution of the averaged strain in the inclusion is

It's worth emphasizing that the time evolution of the averaged strain in the inclusion is also purely hydrostatic and can be easily deduced from the time evolution of the averaged stress in the inclusion from (28):

$$\varepsilon_m^{(2)}(t) = \frac{1}{3\kappa_2} \sigma_m^{(2)}(t) + \varepsilon_0^{(2)}(t). \tag{43}$$

The time evolution of the averaged stress in the matrix is deduced by the usual average relation as a function of the macroscopic stress as well as the averaged stress in the inclusion as follows:

$$\sigma_m^{(1)}(t) = \frac{1}{c_1} \Big( \Sigma_m(t) - c_2 \, \sigma_m^{(2)}(t) \Big). \tag{44}$$

Obviously, a similar relation can be stated for the averaged strain in the matrix.

Finally, the time-evolution of the second moment of the stress field is non-zero in the matrix phase and given by:

$$<\sigma_{eq}^{2}(t)>_{1}=\frac{4\pi}{c_{1}V}\int_{a}^{b}\left(\sigma_{s}(r,t)\right)^{2}r^{2}dr.$$
 (45)

#### 4. Exact solutions for particular cases

For further developments, the dissipation potential of the matrix is supposed to be a power-law:

$$w_1(\underline{\sigma}) = \frac{\dot{e}_0 \,\sigma_0}{n+1} \left(\frac{\underline{\sigma}}{\sigma_0^2}\right)^{\frac{n+1}{2}} \tag{46}$$

with  $\sigma = \sigma_{eq}^2 = \sigma_s^2$  and material coefficients  $\dot{e}_0, \sigma_0$  and  $n \ge 1$  which characterize the intensity of the creep rate. It's emphasized that the particular situation n = 1 corresponds to a linear viscoelastic behaviour.

The equation (40) takes the form

$$\dot{\sigma}_{s}^{(1)}(r,t) + \frac{\dot{e}_{0}}{2\sigma_{0}\eta}\sigma_{s}^{(1)}(r,t) \left(\frac{\left|\sigma_{s}^{(1)}(r,t)\right|}{\sigma_{0}}\right)^{n-1} = \left(\frac{b}{r}\right)^{3} \left[\frac{\alpha\dot{e}_{0}}{\eta\sigma_{0}} \int_{a}^{b} \frac{\sigma_{s}^{(1)}(u,t)}{u} \left(\frac{\left|\sigma_{s}^{(1)}(u,t)\right|}{\sigma_{0}}\right)^{n-1} du + \mathcal{F}(t)\right],\tag{47}$$

Since  $|\sigma_s| = \sigma_{eq}$  and supposing that the sign  $\xi$  of  $\sigma_s^{(1)}$  is independent<sup>2</sup> of r and t, this equation becomes a differential equation for the equivalent stress  $\sigma_{eq}^{(1)}$ . Denoting  $\tau_Y = \frac{2\eta\sigma_0}{\dot{e}_0}$  ( $\tau_Y$  has a time dimension), equation (47) reads:

$$\frac{\dot{\sigma}_{eq}^{(1)}(r,t)}{\sigma_0} + \frac{1}{\tau_Y} \left(\frac{\sigma_{eq}^{(1)}(r,t)}{\sigma_0}\right)^n = \left(\frac{b}{r}\right)^3 \left[\frac{2\alpha}{\tau_Y} \int_a^b \frac{1}{u} \left[\frac{\sigma_{eq}^{(1)}(u,t)}{\sigma_0}\right]^n du + \xi \frac{\mathcal{F}(t)}{\sigma_0}\right]. \tag{48}$$

Closed-form solutions of the equation (48) can be obtained only when the right hand side of (48) vanishes or is constant with respect to the time variable. Moreover, if the integral term is present in (48) only numerical solutions can be obtained. The table 1 resumes the values taken by the coefficient  $\alpha$  and the function  $\mathcal{F}(t)$  (as defined by relations (A.5) and (A.4)) for different hypothesis about the behaviour of the phases.

In what follows all the loading functions have the form of an Heaviside step function,

$$E_m^0(t) = E_m^0 H(t) \quad \text{or} \quad \Sigma_m^0(t) = \Sigma_m^0 H(t), \qquad \varepsilon_0^{(2)}(t) = \varepsilon_0^{(2)} H(t),$$
 (49)

where H(t) is the Heaviside function. At t = 0, the instantaneous answer is purely elastic. As the macroscopic inelastic strain is zero at t = 0, its subsequent time evolution (relation (37)) can be written as follows:

$$\epsilon_m^v(t) = \frac{\frac{4}{3}c_2\left(\kappa_1 - \kappa_2\right)\left(c_2\kappa_1 + c_1\kappa_2 + \frac{4}{3}\mu_1\right)}{\left(\kappa_1\kappa_2 + \frac{4}{3}\mu_1\left(c_1\kappa_1 + c_2\kappa_2\right)\right)\left(c_2\kappa_1 + c_1\kappa_2\right)} \int_a^b \frac{\left(\sigma_s^{(1)}(r,0) - \sigma_s^{(1)}(r,t)\right)}{2r} dr, \tag{50}$$

<sup>&</sup>lt;sup>2</sup>This hypothesis can be verified a posteriori due to the uniqueness of the solution of (47).

	$\alpha_p$	$\mathcal{F}_p(t)$	$\alpha_d$	$\mathcal{F}_d(t)$
$\kappa_1 = \kappa_2$	0	$\frac{18c_2\kappa_1\mu_1}{3\kappa_1 + 4\mu_1}\dot{\varepsilon}_0^{(2)}(t)$	0	$\frac{18c_2\kappa_1\mu_1}{3\kappa_1 + 4\mu_1}\dot{\varepsilon}_0^{(2)}(t)$
$\kappa_1 \to \infty$	$\frac{6\mu_1c_2}{3\kappa_2 + 4\mu_1c_1}$	$\alpha_p \left( \dot{\Sigma}_m^0(t) + 3\kappa_2 \dot{\varepsilon}_0^{(2)}(t) \right)$	0	$6\mu_1  \dot{E}_m^0(t)$
$\kappa_2 \to \infty$ $\mu_2 \to \infty$ rigid inclusion	$-\frac{6\mu_1c_2}{3\kappa_1+4\mu_1c_2}$	$\alpha_p \left( \dot{\Sigma}_m^0(t) - 3\kappa_1 \dot{\varepsilon}_0^{(2)}(t) \right)$	$-\frac{2c_2\mu_1}{c_1\kappa_1}$	$6\mu_1 \frac{c_2}{c_1} \left( \dot{\varepsilon}_0^{(2)}(t) - \dot{E}_m^0(t) \right)$
$\kappa_2 = 0$ $\mu_2 = 0$ voided inclusion	$\frac{3c_2}{2c_1}$	$\alpha_p \dot{\Sigma}_m^0(t)$	$\frac{6\mu_1c_2}{3c_2\kappa_1 + 4\mu_1}$	$3\alpha_d\kappa_1\dot{E}_m^0(t)$

Table 1: Values of  $\alpha$  and  $\mathcal{F}(t)$  for different particular situations; indice d is used for dilatation and indice p is used for pressure (homogeneous strain or stress prescribed on the sphere composite outer boundary, respectively)

where the elastic field  $\sigma_{eq}^{(1)}(r,0_+)$  is given by (16) in the case of a homogeneous strain applied on the boundary or by (18) if a traction is applied on the boundary. In addition, time derivatives of the loading functions are zero for time t > 0 ( $\dot{E}_m^0(t) = 0$  or  $\dot{\Sigma}_m^0(t) = 0$ ,  $\dot{\varepsilon}_0^{(2)}(t) = 0$ ) so that the time functions  $\mathcal{F}_d(t)$  and  $\mathcal{F}_p(t)$  vanishes (see table 1). Then, the in-homogeneous term of equation (48) is constant or zero. Exact solutions can be obtained in the case where the phases have the same compressibility modulus or in the case of an incompressible matrix under a relaxation test. The cases of a voided or a rigid inclusion are of particular interest. In all other cases, including the cases of a voided or a rigid inclusion, a numerical integration must be performed, but as illustrated in section 4.2.2, this integration is quite straightforward and can be easily implemented.

#### 4.1. Composite sphere with homogeneous compressibility

As remarked at the end of section 3.3, the macroscopic inelastic strain  $\epsilon_m^v(t)$  vanishes and the macroscopic stress-strain relation is purely elastic (relation (38)) when the compressibility is homogeneous ( $\kappa_1 = \kappa_2$ ). As a result, when the loading functions obeys an Heaviside step function, the macroscopic strain and stress are constant while time dependent relaxation phenomena are limited to the microscopic fields. For instance the averaged stress in the inclusion can be easily expressed as a function of the macroscopic stress and the radial distribution of the shear stress in the matrix:

$$\sigma_m^{(2)}(t) = \Sigma_m(0) - \int_a^b \frac{2}{u} \, \sigma_s^{(1)}(u, t) \, du \tag{51}$$

An explicit expression of the radial distribution of the shear stress can be derived in that particular situation. Indeed, the equation (48) simplifies to:

$$\frac{\dot{\sigma}_{eq}^{(1)}(r,t)}{\sigma_0} + \frac{1}{\tau_Y} \left( \frac{\sigma_{eq}^{(1)}(r,t)}{\sigma_0} \right)^n = 0$$
 (52)

and the initial (elastic) distribution of the equivalent stress reads:

$$\frac{\sigma_{eq}^{(1)}(r,0_{+})}{\sigma_{0}} = \frac{18\mu_{1}\kappa_{1}}{\sigma_{0}(3\kappa_{1}+4\mu_{1})} \frac{a^{3}}{r^{3}} \left| \varepsilon_{0}^{(2)} \right| = \frac{2\left| \varepsilon_{0}^{(2)} \right|}{\dot{e}_{0}\tau_{Y}} \frac{a^{3}}{r^{3}}.$$
 (53)

The solution of this ordinary differential problem is:

$$\frac{\sigma_{eq}^{(1)}(r,t)}{\sigma_{0}} = \begin{cases}
\left[ (n-1)\frac{t}{\tau_{Y}} + \left(\frac{2\left|\varepsilon_{0}^{(2)}\right|}{\dot{e}_{0}\tau_{Y}}\right)^{1-n} \left(\frac{a^{3}}{r^{3}}\right)^{1-n} \right]^{\frac{1}{1-n}}, & \text{if } n > 1 \\
\frac{2\left|\varepsilon_{0}^{(2)}\right|}{\dot{e}_{0}\tau_{Y}} \frac{a^{3}}{r^{3}} \exp\left(-\frac{t}{\tau_{Y}}\right), & \text{if } n = 1
\end{cases}$$
(54)

As expected, if a traction is prescribed to the sphere model outer boundary, the strain and stress fields are uniform. Only a differential swelling between the two phases will induce non uniform mechanical fields and relaxation phenomena will be driven by a non-zero equivalent stress field in the matrix. Note also that the equivalent stress doesn't vanishes for any r and t and therefore the stress  $\sigma_s$  has constant sign which is that of the stress-free strain  $\varepsilon_0^{(2)}$ .

#### 4.2. Composite sphere with an isochoric matrix

For an incompressible matrix  $(\kappa_1 \to \infty)$ , the macroscopic strain-stress relation is still given by the relation (36) but the expressions of the effective elastic compressibility and the macroscopic stress-free free strain reduce to :

$$\tilde{k} = \frac{1}{c_2} \left( \kappa_2 + \frac{4}{3} \mu_1 c_1 \right) \quad \text{and} \quad \tilde{\varepsilon}_m^0 = \frac{3 c_2 \kappa_2}{3 \kappa_2 + 4 \mu_1 c_1} \varepsilon_0^{(2)}.$$
 (55)

As compared to the previous situation (homogeneous compressibility), the inelastic strain is non-zero and its time evolution is given by (50). In this last relation, the right-hand side pre-factor reduces to  $\frac{4c_2}{3\kappa_2+4\mu_1 c_1}$ .

Equation (48) is homogeneous in the case where a strain is applied on the outer boundary (relaxation loading) and non homogeneous in the case where a traction is applied (creep loading). It is therefore more appropriate to consider each type of loading separately. It's remarked that the constant  $\tau_Y$  in (48) reads  $\tau_Y = \frac{\sigma_0}{3\mu_1 \dot{e}_0}$  when  $\kappa_1 \to \infty$ .

# 4.2.1. Relaxation loading

In this case the equation (48) reduce to (52) which has a solution of the form (54) where the initial (elastic) radial distribution of the equivalent stress is hereby given by:

$$\frac{\sigma_{eq}^{(1)}(r,0_{+})}{\sigma_{0}} = \frac{6\mu_{1}}{\sigma_{0}c_{2}}\frac{a^{3}}{r^{3}}\left|E_{m}^{0}\right| = \frac{2\left|E_{m}^{0}\right|}{\tau_{Y}\dot{e}_{0}}\frac{b^{3}}{r^{3}}.$$
(56)

It's noted that, as a result of the isochoric behaviour of the matrix, the equivalent stress does not depend on the prescribed stress-free strain in the inclusion  $\varepsilon_0^{(2)}$ . Again, the equivalent stress doesn't vanish for any r and t and the stress  $\sigma_s$  has a constant sign, that of the applied strain strain  $E_m^0$ .

# 4.2.2. Creep loading

As relation (48) is non homogeneous, this case differs significantly from those considered before. This relation reads:

$$\frac{\dot{\sigma}_{eq}^{(1)}(r,t)}{\sigma_0} + \frac{1}{\tau_Y} \left(\frac{\sigma_{eq}^{(1)}(r,t)}{\sigma_0}\right)^n = \frac{b^3}{r^3} \frac{2\alpha_p}{\tau_Y} \int_a^b \frac{1}{u} \left[\frac{\sigma_{eq}^{(1)}(u,t)}{\sigma_0}\right]^n du, \tag{57}$$

with the initial value given by (18):

$$\frac{\sigma_{eq}^{(1)}(r,0_{+})}{\sigma_{0}} = \frac{6\mu_{1}}{\sigma_{0}} \frac{a^{3}}{r^{3}} \frac{\left| 3\kappa_{2}\varepsilon_{0}^{(2)} + \Sigma_{m}^{0} \right|}{3\kappa_{2} + 4c_{1}\mu_{1}} = \frac{\alpha_{p}}{\sigma_{0}} \frac{b^{3}}{r^{3}} \left| 3\kappa_{2}\varepsilon_{0}^{(2)} + \Sigma_{m}^{0} \right|.$$
 (58)

Using the change of variable  $\check{\sigma}(r,\tau) = \frac{\sigma_{eq}^{(1)}(r,\tau\tau_Y)}{\sigma_0} = \frac{\sigma_{eq}^{(1)}(r,t)}{\sigma_0}$ , equation (57) reads:

$$\frac{\partial \check{\sigma}}{\partial \tau}(r,\tau) + \check{\sigma}^n(r,\tau) = 2\alpha_p \frac{b^3}{r^3} \int_a^b \frac{1}{u} \check{\sigma}^n(u,\tau) du.$$
 (59)

This integro-differential equation has no analytical solution and it must be integrated numerically, which implies a numerical estimate of the integral coupled with a integration method of ordinary differential equations. In order to compute the integral in (59) we choose a discretization of the interval [a, b],  $r_1 = a < r_2 < ... < r_{N+1} = b$  and using a quadrature formula we get:

$$\int_{a}^{b} \frac{1}{u} \check{\sigma}^{n}(u,\tau) du = \sum_{k=1}^{N+1} w_{k} \frac{\sigma_{k}^{n}(\tau)}{r_{k}}, \tag{60}$$

where  $\sigma_k(\tau) = \check{\sigma}(r_k, \tau)$ . Then, writing (59) for  $r = r_i$ , i = 1, ..., N + 1 a system of ordinary differential equations of first order for the unknowns  $\sigma_k(\tau)$  is obtained:

$$\frac{\mathrm{d}\sigma_i}{\mathrm{d}\tau}(\tau) + \sigma_i^n(\tau) = 2\alpha_p \frac{b^3}{r_i^3} \sum_{k=1}^{N+1} w_k \frac{\sigma_k^n(\tau)}{r_k}.$$
 (61)

This system of non linear differential equations is solved with and adaptive Runge-Kutta method. One example of solution is shown in figure 3 which was obtained using a regular discretization and the trapezoidal rule  $(w_i = \frac{b-a}{N}, i = 2, ..., N)$  and  $w_1 = w_{N+1} = \frac{b-a}{2N}$ . The different constants involved in the model are set as follows: b = 1,  $c_2 = 0.1$ ,  $\varepsilon_0^2 = 0.01$ , n = 4,  $\kappa_2/\sigma_0 = 166$ ,  $\mu_1/\sigma_0 = 77$  and  $\Sigma_m^0 = 0$ .

At  $\tau = 0$  (elastic response), the equivalent stress is a decreasing function of the radius  $(\propto r^{-3})$  and reaches its maximum at the inner surface of the matrix. As a result the creep rate (and the stress relaxation kinetic) is the highest near the inner boundary. Finally and as expected, the radial stress distribution tends progressively toward zero.

#### 5. Full field simulations

The model proposed in the previous sections is intended to be applied to particulate composites with a moderate volume fraction of inclusions ( $\leq 30\%$ ), whose microstructures

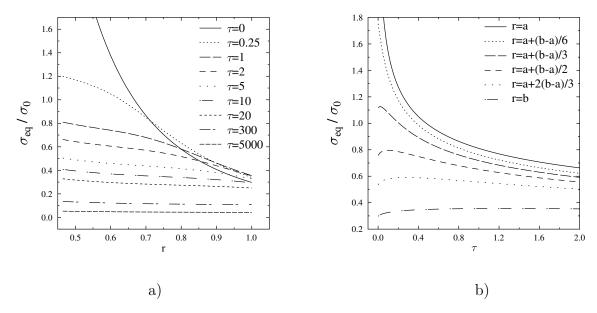


Figure 3: Equivalent stress in the matrix phase: a) distribution of the equivalent stress in the matrix at different moments; b) time evolution of the equivalent stress in the matrix for different radii.

can be idealized by the Hashin's spheres assemblage. Full-field computations with periodic boundary conditions are used to assess the composite sphere model. Hereafter we consider a representative volume element (RVE) of a particulate composite. The particles have spherical shape with the same radius and are isotropically distributed in the RVE. No forces are applied to the outer surface of this RVE while particles are submitted to the same stress-free strain  $\varepsilon_0^{(2)}(t) > 0$ . The time evolution of the stress-free strain follows a Heaviside step function,  $\varepsilon_0^{(2)}(t) = \varepsilon_0^{(2)}H(t)$ . In addition, the elastic compressibility is chosen uniform  $(\kappa_1 = \kappa_2)$ .

The isotropic loadings applied to the RVE lead to a nill average shear stress in the matrix phase while the shear stress field is clearly non zero in this phase. This is typically a case which challenges the classical homogenization theories.

#### 5.1. RVE generation

The RVE is generated using the random sequential addition (or adsorption, RSA) algorithm ([26]): the spherical particles are progressively added in the RVE. To avoid overlapping inclusions, a new particle is added only if it does not intersect any of the already existing particles. If the inclusion intersect the boundary of the RVE, it is duplicated on the opposite face. With this method, several RVEs were generated with a volume fraction of inclusions ranging from 1% to 30% (intermediate values: 2%, 5% and 10%). Pictures of these microstructures are reported on Figure 4.

Moreover, for the microstructure with 5% volume fraction of inclusions three realizations were generated in order to ensure that the dispersion of the simulated responses (macroscopic behaviour, first and second order moments of the mechanical fields) is less than 0.5%.

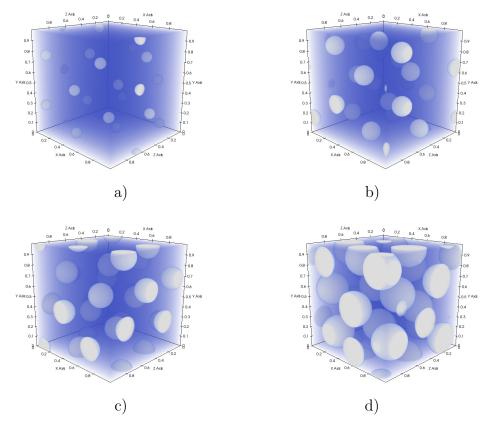


Figure 4: RVE microstructures for a) 1% b) 5% c) 10% and d) 30% with 256<sup>3</sup> voxels

### 5.2. FFT computations

The computational method used for this analysis is based on fast Fourier transforms, originally proposed by [27] and implemented in the CraFT freeware (a software freely available at http://craft.lma.cnrs-mrs.fr). The basic scheme implemented in CraFT is a fixed point algorithm which evaluates at each time step the strain field  $\varepsilon$  for a given stress field  $\sigma$  using fast Fourier transforms and the exact expression of the Green function for a linear comparision medium. The fixed point algorithm is described in [27], the local behavior (equation (5)) being computed by an implicit Euler method.

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% (n=4,  $c_2=5\%$ ). The results as presented in this paper are obtained with  $256^3$  voxels which is a good compromise between computation times and accuracy.

#### 5.3. Results

For an homogeneous compressibility and Heaviside loading functions, the solution of the composite sphere is analytical (see relation (54)). The macroscopic strain is given by:

$$E_m(t) = c_2 \,\varepsilon_0^{(2)} \tag{62}$$

while the averaged stresses in the two phases are purely hydrostatic and given by:

$$\sigma_m^{(2)}(t) = -\int_a^b \frac{2}{u} \,\sigma_s^{(1)}(u,t) \,du \qquad \sigma_m^{(1)}(t) = -\frac{c_2}{c_1} \sigma_m^{(2)}(t). \tag{63}$$

Due to the instantaneous stress-free strain of the inclusions, the inclusions phases will be in compression while the matrix is in traction. The intensity of these hydrostatic stresses depends on the time evolution of the shear stress distribution in the matrix. Hereafter, the time and the stresses are normalized as follows:

$$\hat{t} = \frac{t}{\tau_R} \quad \text{with} \quad \tau_R = \frac{\tau_Y}{n-1} \left( \frac{\left| \varepsilon_0^{(2)} \right|}{\eta} \sigma_0 \right)^{1-n} \quad \text{and} \quad \hat{\boldsymbol{\sigma}} = \frac{\boldsymbol{\sigma}}{\sigma_R} \quad \text{with} \quad \sigma_R = \frac{\left| \varepsilon_0^{(2)} \right|}{\eta}$$
 (64)

Substituting the expression of the signed equivalent stress (54) in relation (63) and adopting the substitution  $v = \frac{u}{a}$  in the integral on the right hand side of this last relation, the evolution with  $\hat{t}$  of the averaged normalized stress in the inclusions can be easily shown to depend only on the power-law exponent and the volume fraction of inclusions:

$$\hat{\sigma}_m^{(2)}(\hat{t}) = -2 \int_1^{c_2^{-\frac{1}{3}}} \left[ \hat{t} + v^{3(1-n)} \right]^{\frac{1}{1-n}} \frac{dv}{v}. \tag{65}$$

A similar result can be derived for the second-moment of the stress field in the matrix phase:

$$<\hat{\sigma}_{eq}^2(\hat{t})>_1 = \frac{3c_2}{c_1} \int_1^{c_2^{-\frac{1}{3}}} \left[\hat{t} + v^{3(1-n)}\right]^{\frac{2}{1-n}} v^2 dv.$$
 (66)

The normalized results (65) and (66), as derived by the composite sphere model, depend only on the power-law exponent and the volume fraction of inclusions. Does this normalization apply to the results as predicted by full-field computations? To answer this question, we have studied the effect of the stress-free strain intensity on the evolution of the averaged stress in the inclusions with  $\hat{t}$  as predicted by full-field computations ( $n = 8, c_2 = 10\%$ ): as shown in Figure 5, even if the stress-free strain intensity is increased by an order of magnitude, the normalized averaged stress in the inclusion remains constant. This suggests that the distribution of the equivalent stress in the composite sphere should be close to the one in the RVE. This aspect is analyzed in details at the end of this section.

We have reported on Figures 6 the (normalized) time evolution of the first and second moments per phase of the stress field for a moderate power-law exponent (n=4) and a moderate volume fraction of inclusions  $(c^{(2)}=10\%)$ . As expected, the stress intensity is maximum at  $\hat{t}=0$  (elastic response) and relaxes progressively. In addition, for this moderate volume fraction of inclusions, the composite sphere model agrees well with the full-field computations. A similar result was observed in the cases of an elastic behaviour or a non linear viscoplastic behaviour, and this is due to the fact that when the volume fraction of inclusions is moderate, the interactions between inclusions are weak and the composite sphere asemblage is a good approximation of the composite microstructure.

In Figure 7 are reported the same long time evolution of the first and second moments per phase of the normalized stress as in Figure 6, but for a higher power-law exponent

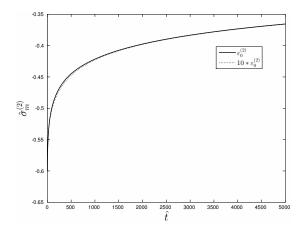


Figure 5: Evolution with  $\hat{t}$  of the (normalized) stress first moment in the inclusion as predicted by the full-field calculations for two stress-free strains (n = 8,  $c_2 = 10\%$ ,  $\varepsilon_0^{(2)} = 3.3 \times 10^{-4}$ )

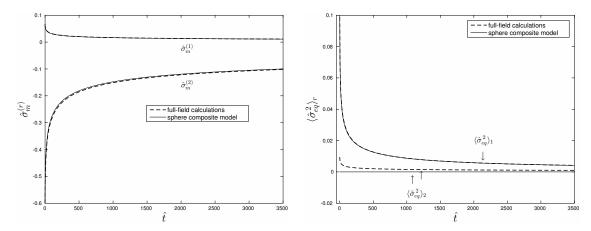


Figure 6: Evolution with  $\hat{t}$  of the (normalized) stress first and second moments in both phases as predicted by the sphere composite model and the full-field calculations ( $n = 4, c_2 = 10\%$ )

(n = 10) and a higher volume fraction of inclusions  $(c^{(2)} = 30\%)$ . As expected, when the volume fraction of inclusions increases the agreement between the predictions of the composite sphere model and the results of the full-field simulations is getting worse. The curves ploted in Figure 8 show that this effect is mostly dependent on the volume fraction of inclusions and far less on the power-law exponent.

It's also remarked in Figure 7 that the full-field computation predict a non-zero second-order moment in the inclusions. However, this second-order moment remains small as compared to the ones computed in the matrix.

One of the goal of this study is to assess the pertinence of the composite sphere model in describing the spatial distribution of the stress field around the inclusions of the RVE. In Figure 9 the distribution of the equivalent stress in the matrix (54) in the composite sphere (continuous line) is compared with that observed around one of the inclusions of the RVE (error bars). We observed that choosing any other inclusion leads to similar results since the RVE generation algorithm gives a sufficiently homogeneous distribution of the centers of the inclusions. The volume fraction of inclusions is 30% and the power-law exponent is n = 10.

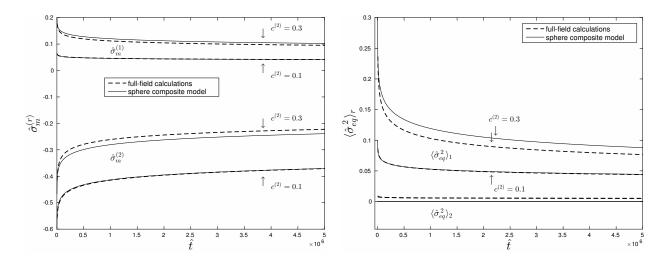


Figure 7: Evolution with  $\hat{t}$  of the (normalized) stress first and second moments in both phases as predicted by the sphere composite model and the full-field calculations (n = 10,  $c_2 = 10\%$  and  $c_2 = 30\%$ )

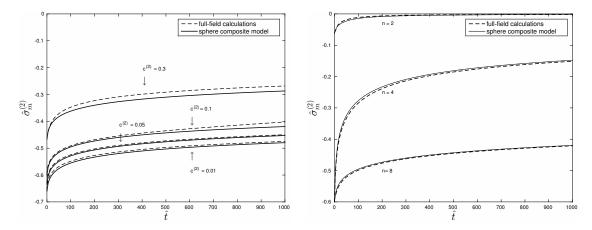


Figure 8: Evolution with  $\hat{t}$  of the (normalized) stress moments in the inclusion as predicted by the sphere composite model and the full-field calculations: effect of the inclusions volumic fraction  $c_2$  (left, n=8) as well as the power-law exponent n (right,  $c_2=10\%$ )

The error bars was obtained by considering concentric spherical shells of constant thickness around the inclusion. For each shell, the minimal and the maximal of the values of the equivalent stress associated to the voxels of the shell give the ends of the corresponding error bar. The average of these values gives the dot on the error bar. These values are associated to a radius which corresponds to the half of the tickness of the shell.

For all instants, in each spherical shell, the dispersion (the difference between the maximal and the minimal values) of the equivalent stress is significant and seems to increase with the radius. As the voxels located at the inclusion's boundary are ill defined, results computed for the corresponding limit layer are not reported on Figure 9. However, the average values are in very good agreement with the analytical results. This shows that the composite sphere model gives pertinent estimates of the stress field inside the particulate composite.

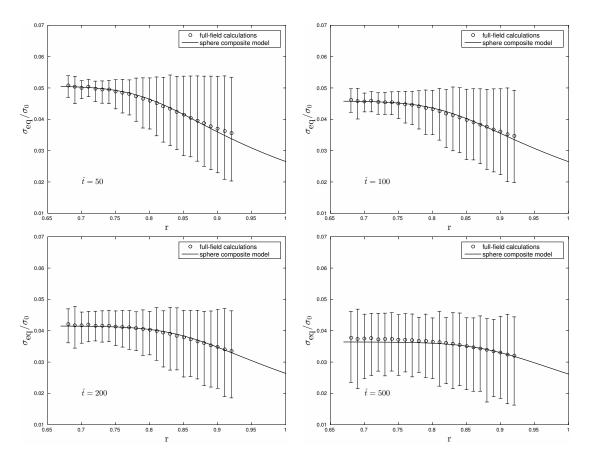


Figure 9: Spatial distribution of the equivalent stress in the matrix phase, in the composite sphere (continuous line) and around one of the inclusions of the RVE (error bars), at different instants ( $c_2 = 30\%$ , n = 10)

#### 6. Conclusions

For an isotropic loading, the composite sphere problem has been solved theoretically when the two phases obey a non linear viscoelastic behaviour. The loading is given by a time history of dilatation or traction prescribed on the outer surface of the composite sphere as well a stress-free strain homogeneously applied into the inclusions (thermal strain mismatch for instance). Denoting by  $\tilde{k}$  the effective elastic compressibility (relation 13) and by  $\tilde{\epsilon}_m^0(t)$  the stress-free macroscopic swelling (elastic solution, relation (17)), the macroscopic strain-stress behaviour of the composite sphere reads:

$$\Sigma_m(t) = 3 \,\tilde{\kappa} \left( E_m(t) - \tilde{\epsilon}_m^0(t) - \epsilon_m^v(t) \right).$$

where the macroscopic strain  $\epsilon_m^v(t)$  is an inelastic strain due to the viscous behaviour of the matrix phase (relation (38)). The time evolution of the macroscopic inelastic strain has been shown to depend on the radial distribution of the shear stress in the matrix. As expected, this radial distribution of the shear stress is the driving force of relaxation phenomena in the composite sphere. Expressions of the first and second moments of the stress field have been also derived (relations (42) and (45)).

For general time evolutions of the loading, this field is solution of a non linear integrodifferential equation (relation (40)). An efficient numerical method has been proposed to solve this equation. When the viscoplastic strain in the matrix obeys a Norton law, closed-form solutions of this equation can also be derived for some special cases (homogeneous compressibility for instance). A similar model should be easily extended to the (2D) composite cylinder model. However, extension to shear loadings appears prohibitively difficult.

Full-field calculations performed on random microstructures of particulate composites have been carried out to assess the accuracy of this model. Power-law viscoelastic behaviours of the phases have been adopted for these computations. For a volume fraction of inclusions and a power-law exponent up to 10% and 10 respectively, the effective behaviour as well as the first and second moments of the stress field were shown to be in good agreement with the predictions of the sphere composite model. However, for higher volume fraction of inclusions (30%) the agreement between the model and the full-field calculations starts to deteriorate. As a result, this model will deserve to be used for engineering studies such as the estimates of process induced residual stresses in particulate composites for moderate volume fraction of inclusions. These theoretical results will be used in future works to improve homogenization methods applied to nonlinear viscoelastic (or elasto-viscoplastic) particulate composites.

# Acknowledgements

The authors acknowledge the financial support of the French Alternative Energies and Atomic Energy Commission (CEA), Aix\*Marseille Université (AMU) and the National Center for Scientific Research (CNRS) via Grant CEA-AMU-CNRS 4251. Dr. J.M. Gatt is also gratefully acknowledged for his helpful comments. The authors also acknowledge Dr. Ing. H. Moulinec for his advices in working with CraFT software.

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# Appendix A. Expressions of $\alpha$ and $\mathcal{F}(t)$ for prescribed dilatation or pressure

A first expression of  $\dot{\lambda}(t)$  is obtained by derivating with time the relation (34):

$$\dot{\lambda}(t) = \frac{c_2(\kappa_2 - \kappa_1)}{c_2\kappa_1 + c_1\kappa_2} \left\{ -\dot{E}_m(t) + \frac{\kappa_2}{\kappa_2 - \kappa_1} \dot{\varepsilon}_0^{(2)}(t) + \frac{1}{3\kappa_1} \int_a^b \frac{2}{r} \dot{\sigma}_s^{(1)}(r, t) dr \right\}. \tag{A.1}$$

As the fields  $\dot{\sigma}_s^{(1)}(r,t)$  and  $\sigma_s^{(1)}(r,t)\frac{\partial w_1}{\partial \tilde{\varphi}}(\tilde{\varphi}^{(1)}(r,t))$  as well as the time function  $\dot{\lambda}(t)$  are non independent (relation (39)), an equivalent relation can be obtained

$$\frac{\dot{\lambda}(t)}{\eta} = \frac{18\mu_1 c_2(\kappa_1 - \kappa_2)}{3(c_1 \kappa_2 + c_2 \kappa_1) + 4\mu_1} \left[ \dot{E}_m(t) - \frac{\kappa_2}{\kappa_2 - \kappa_1} \dot{\varepsilon}_0^{(2)}(t) + \frac{2}{3\kappa_1 \eta} \int_a^b \frac{\sigma_s^{(1)}(u, t)}{u} \frac{\partial w_1}{\partial \underline{\sigma}} (\underline{\sigma}^{(1)}(u, t)) du \right]. \tag{A.2}$$

This last relation is more appropriate to be used in the differential equation (39).

As  $\dot{E}_m(t) = \frac{1}{3\kappa_1}\dot{\Sigma}_m + \dot{\lambda}(t)$  (relation (31)),  $\dot{\lambda}(t)$  can alternatively be expressed as a function of  $\dot{\Sigma}_m$ :

$$\frac{\dot{\lambda}(t)}{\eta} = \frac{18\kappa_1 \mu_1 c_2(\kappa_2 - \kappa_1)}{3\kappa_1 \kappa_2 + 4\mu_1 (c_1 \kappa_1 + c_2 \kappa_2)} \left( -\frac{\dot{\Sigma}_m(t)}{3\kappa_1} + \frac{\kappa_2}{\kappa_2 - \kappa_1} \dot{\varepsilon}_0^{(2)}(t) - \frac{2}{3\eta \kappa_1} \int_a^b \frac{\sigma_s^{(1)}(u, t)}{u} \frac{\partial w_1}{\partial \sigma} (\sigma_s^{(1)}(u, t)) du \right). \tag{A.3}$$

As a result, in the case where the composite sphere is loaded by a spherical and homogeneous strain  $E_m^0(t)$  on its outer boundary, the coefficient  $\alpha$  and the function  $\mathcal{F}(t)$  of (40) are:

$$\alpha_d = \frac{1}{\kappa_1} \frac{6\mu_1 c_2(\kappa_1 - \kappa_2)}{3(c_1 \kappa_2 + c_2 \kappa_1) + 4\mu_1} \quad \text{and} \quad \mathcal{F}_d(t) = 3 \alpha_d \kappa_1 \left( \dot{E}_m^0(t) - \frac{\kappa_2}{\kappa_2 - \kappa_1} \dot{\varepsilon}_0^{(2)}(t) \right)$$
(A.4)

while their expressions are :

$$\alpha_p = -\frac{6\mu_1 c_2(\kappa_2 - \kappa_1)}{3\kappa_1 \kappa_2 + 4\mu_1 (c_1 \kappa_1 + c_2 \kappa_2)} \quad \text{and} \quad \mathcal{F}_p(t) = \alpha_p \left( \dot{\Sigma}_m^0(t) - \frac{3\kappa_1 \kappa_2}{\kappa_2 - \kappa_1} \dot{\varepsilon}_0^{(2)}(t) \right)$$
(A.5)

if an homogeneous hydrostatic stress  $\Sigma_m^0(t)$  is applied on the outer boundary.