Modeling of the effective and macroscopic ageing behavior of viscoelastic composite materials

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Abstract. In this work, a mathematical modeling is developed to estimate the ageing macroscopic response and effective behavior of viscoelastic composites. The modeling is developed directly in time domain by considering the Volterra tensorial integral product describing the local behavior and governing equations. The ageing dynamic Green's tensor is introduced in order to solve the partial differential equations for the time dependent displacement and make the transition between local and macroscopic fields. The localization tensor making the scale transition is obtained through using the heterogeneity problem and the Mori-Tanaka mean field approach. Once this step is completed, averaging techniques are used to derive the effective ageing behavior of viscoelastic composites. To compute the macroscopic response and the effective behavior a numerical scheme is used. Numerical results are presented with respect to the shape and volume fraction of inclusions and for different ageing viscoelastic composites.

1 Introduction

Many composite materials are constituted with viscoelastic component to gain in weight and flexibility. These composites present a time dependent behavior and are used under different conditions leading to an ageing behavior. Modeling this ageing behavior is a challenging problem and attracted the interest of many researchers.

Many published works investigated the non ageing behavior in frequency and time domains: among other works one can cite Brinson and Lin [1], Fisher and Brinson [2], Azrar et al. [3] and Bakkali et al. [4]. Some other researchers have been interested by the ageing one. In the literature one can find two kinds of approaches to model ageing viscoelastic composites. The first approach is based on internal variables and most of them are iterative models and the second approach drive the ageing behavior using the dynamic Green's function techniques leading to effective ageing expressions that could be computed with no need to iterative procedures.

Based on the internal variables approach, Lahellec and Suquet [5] computed the ageing viscoelastic behavior of composite materials based on the discretization and solving evolution equations. Another internal variable approach developed by Ricaud and Masson [6] for non ageing and ageing behavior of viscoelastic composites. Masson et al. [7] investigated the ageing viscoelastic behavior of polycristals based on the formulation developed in [6]. Considering the approach based on the Green's function techniques, Sanahuja [8] developed a displacement formulation for ageing composites and considering spherical inclusions and isotropic constituents. Another formulation was developed by El Kouri et al. [9] predicting the overall and effective behavior of non-ageing viscoelastic composites, directly in the time domain and taking into account the general case of ellipsoidal inclusions and anisotropic medium. This later was generalized to composite materials with multi-coated inclusions by El Kouri et al. [10].

In this work, the ageing behavior of viscoelastic composites is investigated. In contrast with the internal variable approach which needs incremental procedures and the displacement approach presented in [8] and restricted to the case of isotropic behavior and spherical inclusions, the developed modeling gives straightforward expressions of the effective ageing behavior while taking into account the anisotropy of viscoelastic composites and capturing different geometries by considering ellipsoidal inclusions. In the case of spherical inclusions and isotropic medium, one can derive analytically expressions of the effective moduli which is not doable in the general case.

In this work, a new formulation and a numerical algorithm based on time discretization to predict the ageing effective and overall response of viscoelastic composites in the general case are elaborated. This article is organized as follow: First, the constitutive equations are given. The ageing behavior is described through Volterra tensorial products. Second, introducing the ageing dynamic Green's functions and based on the inclusion problem viscoelastic fields are obtained. Using

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the equivalence between the inclusion and heterogeneity problem, the localization tensors are derived. Based on the Mori-Tanaka's approach and on average techniques, the expression of the ageing effective behavior is derived through Volterra tensorial products. A numerical procedure is developed to calculate the direct and inverse of the resulting Volterra tensorial products. Finally, a parametric study is done and different parameter effects on the ageing effective behavior of viscoelastic composites are analyzed.

2 Governing equations

A linear ageing viscoelastic domain $D \subset \mathbb{R}^3$ with a relaxation tensor $\mathbb{C}(x,t,t')$ is considered. The linear ageing viscoelastic behavior at each point x of the considered domain is modeled through the following integral-differential equation

$$\sigma(x,t) = \int_0^t \mathbb{C}(x,t,t') : d\varepsilon(x,t') \tag{1}$$

A compact form of this equation is obtained by introducing the Volterra product denoted $"\overset{\circ}{:}"$ [8] as follows

$$\sigma(x,t) = \mathbb{C}(x,t,.) \overset{\circ}{:} \varepsilon(x,.) \tag{2}$$

where σ, ε and $\mathbb{C}$ are the stress tensor, the strain and the relaxation tensors respectively.

For small deformation, the compatibility equation is given by

$$\varepsilon(x,t) = \frac{1}{2}\left[grad\left(u(x,t)\right) + {}^t grad\left(u(x,t)\right)\right] \tag{3}$$

where u is the displacement vector.

The equilibrium equations are given by the following Navier partial differential equation

$$div\left(\sigma(x,t)\right) + F = 0\,, \quad \forall x \in D,\, t \in \left[0,+\infty\right[\tag{4}$$

where F is the body vector force.

Inserting Eq. (2) into Eq. (4), the following second order partial differential equation results.

$$\mathbb{C}_{ijkl}(x,t,.) \overset{\circ}{:} u_{k,lj}(x,.) + F_i = 0 \tag{5}$$

Under given boundary conditions, the displacement field in a viscoelastic body, with a known relaxation tensor $\mathbb{C}$, in response to a body force, may be obtained in the time domain by appropriate numerical procedures.

3 Derivation of the viscoelastic field

In this section, the displacement field solution of Eq. (5) is sought in response of a concentrated body force $F_j(t) = \delta(x-x')H(t)\delta_{ij}$, where $H(t)$ is the Heaviside function, $\delta(\bullet)$ the Dirac distribution and $\delta_{..}$ the Kronecker symbol. To do so, the ageing dynamic viscoelastic Green's tensor $G_{ij}(x-x',t,t')$ of an infinite viscoelastic medium is introduced. It constitutes the elementary displacement at each time t, x and in the direction i caused by a unit force F_j applied in the direction j and in a continuous manner from the time t'. The displacement field is thus written as follow

$$u_i(x,t) = \int_{D'} G_{ij}(x-x',t,.) \overset{\circ}{:} F_j(.)\, dD' \tag{6}$$

Substituting Eq. (6) into Eq. (5) and after some mathematical procedures, the following Volterra integral equation, satisfied by the dynamic viscoelastic Green's tensor, is obtained

$$\begin{aligned} &\mathbb{C}_{kpim}(x,t,.) \overset{\circ}{:} G_{ij,mp}(x-x',.,t') \\ &\quad + \delta(x-x')\delta_{jk} H(t-t') = 0 \end{aligned} \tag{7}$$

This equation is equivalent to Eq. (5) when the body force is a delta function $F_j(t) = \delta(x-x')H(t)\delta_{ij}$.

4 Effective properties and numerical algorithm

To obtain the expression of the localization tensor making the transition between ageing local fields and macroscopic ones has to go through adjusting the eigenstrain ε^* to achieve equivalency between the inclusion problem and the heterogeneity problem. To do so, the time stress evolution inside the inclusion is taken equal to the time stress evolution inside the heterogeneity. The following relationship is thus obtained in Ω (volume of the inclusion that is a subdomain of D)

$$\mathbb{C}^*(t,.) \overset{\circ}{:} \left[\varepsilon^\infty(.) + \varepsilon^d(.)\right] = \mathbb{C}(t,.) \overset{\circ}{:} \left[\varepsilon^\infty(.) + \varepsilon^d(.) - \varepsilon^*(.)\right] \tag{8}$$

where ε^d is the disturbance ageing caused by the presence of inclusion.

Introducing the Eshelby tensor, the following equivalent inclusion equation results:

$$\begin{aligned} &\mathbb{C}^*(t,.) \overset{\circ}{:} \left[\varepsilon^\infty(.) + S^\Omega(t',.) \overset{\circ}{:} \varepsilon^*(.)\right] = \\ &\qquad \mathbb{C}(t,.) \overset{\circ}{:} \left[\varepsilon^\infty(.) + S^\Omega(t',.) \overset{\circ}{:} \varepsilon^*(.) - \varepsilon^*(.)\right] \end{aligned} \tag{9}$$

in which S^Ω is the ageing viscoelastic interior Echelby tensor related to shape of the inclusion as well as to the properties of its surrounding.

The relationship between the eigenstrain ε^* and the applied strain at the boundary of the infinite medium, ε^∞, is obtained by

$$\varepsilon^*(t) = \Lambda(t,.) \overset{\circ}{:} \varepsilon^\infty(.) \tag{10}$$

with $\Lambda(t,t') = \left[\left(\mathbb{C}(t,.) - \mathbb{C}^*(t,.) \right)^{-1} \overset{\circ}{:} \mathbb{C}(.,t') - S^{\Omega}(t,t') \right]^{-1}$.

Using the expression of the total time dependent strain in the inclusion, the transition is made between the averaged time dependant strain inside the inclusion and the applied one at the boundary of the infinite medium:

$$\varepsilon^I(t) = A^I(t,.) \overset{\circ}{:} \varepsilon^{\infty}(.) \quad (11)$$

where $A^I(t,t')$ is the time dependent localization tensor. Then, one can express the localization tensor as follow:

$$A^I(t,t') = H(t,t')I + S^{\Omega}(t,.) \overset{\circ}{:} \Lambda(.,t') \quad (12)$$

where I is the fourth order identity tensor and $H(t,t')$ is the Heaviside function and is denoted as a function of two variables for the sake of commodity such as $H(t,t') = H(t-t')$.

Eq. (12) is the dilute approximation of the localization tensor. In this paper, the Mori-Tanaka micromechanical mean field approach is used. In the Mori-Tanaka's approximation, an inclusion with the relaxation tensor $\mathbb{C}^I(t,t')$ is considered to be embedded in a matrix with the relaxation tensor $\mathbb{C}^M(t,t')$. The medium is subjected to the time strain evolution $E(t)$ at its boundary. The interaction between the matrix and the inclusion is taken into account by considering that the time strain evolution in the matrix equals to the average of the time strain evolution over the volume of the matrix. To get the associated Mori-Tanaka's localization tensor, ε^{∞} and the relaxation tensor $\mathbb{C}^*(t,t')$ in Eq. (11) are replaced by ε^M, averaged ageing strain over volume of the matrix, and $\mathbb{C}^M(t,t')$ as well as $\mathbb{C}(t,t')$ is replaced by $\mathbb{C}^I(t,t')$. The following equation is thus obtained:

$$\varepsilon^I(t) = A^I(t,.) \overset{\circ}{:} \varepsilon^M(.) \quad (13)$$

Using the assumption

$$E(t) = f^I \varepsilon^I(t) + f^M \varepsilon^M(t) \quad (14)$$

where f^I and f^M are volume fractions of the inclusion and matrix respectively, and based on the presented equations, the following relationship is formulated

$$\varepsilon^I(t) = \left[f^M H(t,.)I + f^I A^I(t,.) \right]^{-1} \overset{\circ}{:} A^I(.,.) \overset{\circ}{:} E(.) \quad (15)$$

From this equation, one can deduce the resulting time dependent Mori-Tanaka's localization tensor

$$A^{MT}(t,t') = \left[f^M H(t,.)I + f^I A^I(t,.) \right]^{-1} \overset{\circ}{:} A^I(.,t') \quad (16)$$

Based on the averaging techniques [8] and on the localization relationship given by Eq. (15), the ageing effective behavior of the two phase viscoelastic composite is formulated by

$$\mathbb{C}^{eff}(t,t') = \mathbb{C}^M(t,t') + f^I \left[\mathbb{C}^I(t,.) - \mathbb{C}^M(t,.) \right] \overset{\circ}{:} A^{MT}(.,t') \quad (17)$$

It should be noted that this new formulation is time dependent with Volterra tensorial products and presents a closed form formulation for computing the ageing effective behavior of the considered reinforced viscoelastic composites.

The overall response of the composite can be obtained based on the relationship relating the macroscopic stress to the macroscopic strain applied to the composite through the effective ageing relaxation tensor by the following equation

$$\Sigma(t) = \mathbb{C}^{eff}(t,.) \overset{\circ}{:} E(.) \quad (18)$$

Based on the key formulations, Eq. (17) and Eq. (18), the effective ageing behavior and the overall response of two-phase viscoelastic composite could be predicted by taking into account the constituents properties, such as the volume fraction and shape of inclusions as well as the ageing effect. The main difficulty of this work lies in the calculation of the direct and inverse Volterra tensorial products. A numerical procedure is elaborated. Discretizing times t and t' are based on a trapezoidal algorithm [9].

5 Numerical results

In this part of the work, the ageing effective and overall response of viscoelastic composite are computed for different volume fractions, shape of inclusions and ageing time. The considered composite is composed of elastic inclusions embedded in an ageing viscoelastic matrix. The behavior of the matrix and inclusions are isotropic. The linear ageing law without the temperature effect describing the ageing behavior of the viscoelastic constituent is modeled in [8] by

$$J_0^E(t,t') = \frac{1}{E_0} + f(t') \sum_{p=1}^{n} J_0^p \left[1 - \exp\left(-\frac{t-t'}{\tau_0^p} \right) \right] \quad (19)$$

in which J_0^E is the compliance function, E_0 is the Young's modulus ($1/E_0$ corresponds to the elastic part of the compliance function). J_0^p and τ_0^p, $p \in \{1,...,n\}$ are viscoelastic parameters. $f(t')$ is the ageing function taken as

$$f(t') = \exp\left(-\frac{t'}{\tau} \right) \quad (20)$$

where is the characteristic ageing time.

Thus, the ageing viscoelastic behavior of the constituent is given by the ageing viscoelastic compliance function $J_0^E(t,t')$ and by the associated Poisson ratio ν_0 that is considered time independent. The elastic constituent behavior is described by the Young modulus E_1 and the associated Poisson ratio ν_1. The used properties are listed in Table 1.

In the considered general case, the shape of inclusions are ellipsoids with the semi-axes a, b and c. In the numerical results ($a=b=c=1$) are taken for spherical, ($a=b=1$ and $c=10$) for ellipsoidal and ($a=b=1$ and $c=1000$) for fibrous composites.

For the sake of comparison and validation of the developed formulation, a comparison is done between the obtained results based on the presented formulation and the ones based on the formulation developed in [8] for the case of ageing viscoelastic composites constituted of spherical elastic inclusions embedded in an ageing viscoelastic matrix. The properties of the constituents of the considered composite are given in Table 1. The normalized effective ageing bulk modulus with respect to the normalized ageing is given in Fig. 1 for spherical elastic inclusions and $f^I = 0.113$.

Table 1. Elastic, viscoelastic and ageing coefficients for the considered materials ([8]).

Elastic material properties	Poisson's ratio	ν_1	0.3
	Young's modulus	E_1/E_0	10
Viscoelastic material properties	Poisson's ratio	ν_0	0.25
	Viscoelastic parameters of Eq. (19)	$J_0^1 E_0$	2
		$J_0^2 E_0$	3
		τ_0^1/τ	0.1
		τ_0^2/τ	0.5

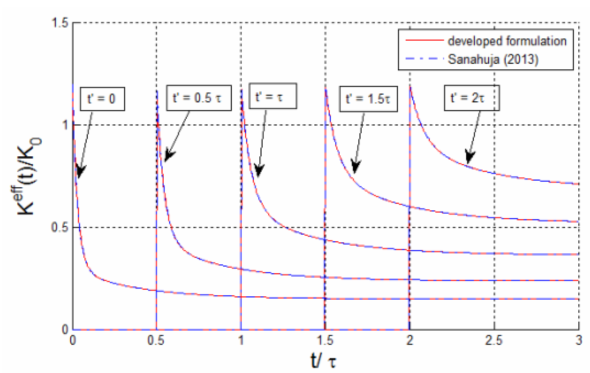

Fig. 1. Normalized effective bulk modulus, by K_0 bulk modulus of the matrix, for an ageing viscoelastic composites constituted of spherical elastic inclusions embedded in an ageing viscoelastic matrix, with the volume fraction of the inclusion $f^I = 0.113$.

The considered composite in Fig. 2 is constituted of elastic inclusions embedded in an ageing viscoelastic matrix.

The harmonic uniaxail strain is given by

$$E_{11}(t) = E_{\max}\cos(\omega t) \tag{21}$$

The amplitude of the harmonic uniaxial strain is fixed in all the figures at $E_{\max} = 10^{-4}$.

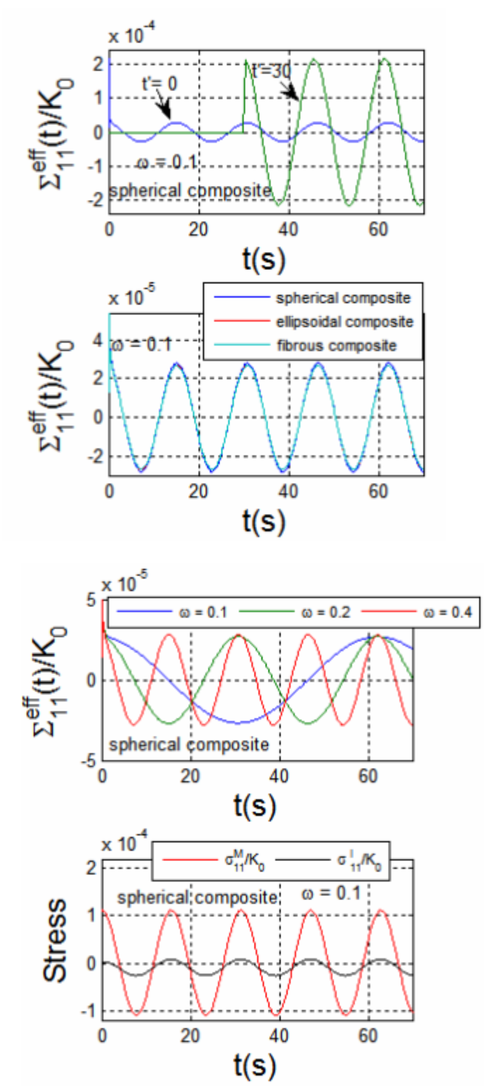

Fig. 2. Overall response of a viscoelastic composite consisting of elastic inclusions embedded in an ageing viscoelastic matrix subjected to a harmonic uniaxial strain. The volume fraction of the matrix is fixed at $f^I = 0.113$ and the ageing characteristic time is fixed at $\tau = 1s$.

In Fig. 2, the overall and local responses of the considered composites are illustrated when the harmonic uniaxial strain is applied on them. The overall response is presented for different frequencies, shapes of inclusion as well as for various ageing times. The local response inside the inclusion and the matrix is presented for the case of spherical inclusions and ageing time equal to $t'=0$. One can see the effect of frequencies as well as the ageing behavior. It is seen that the shape of inclusions does not have an effect on the overall response in the case when the composite is constituted of elastic inclusions embedded in an ageing viscoelastic matrix.

6 Conclusions

A straightforward methodological approach is developed for the computation of the effective time behavior and overall response of ageing viscoelastic composites. The effective behavior is obtained straightforwardly in the time space for general cases of ellipsoidal inclusions and anisotropic composite materials. The modeling is elaborated through Volterra tensorial integrals, the Green time function techniques as well as by using the equivalent inclusion problem and the Mori-Tanaka model. A numerical scheme is used to numerically compute Volterra tensorial products. In the numerical results section, the considered composite is constituted of an ageing viscoelastic matrix reinforced by elastic inclusions. Many numerical results are presented for different ageing times and shapes of inclusions. The overall response is predicted when harmonic uniaxial strains are applied on the composite. The analysis of the ageing behavior and macroscopic response can be easily done and the effect of some parameters is shown.

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