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OVERALL VISCOELASTIC PROPERTIES OF FIBER-REINFORCED HIERARCHICAL COMPOSITES

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Abstract. *The work focuses on the calculation of the effective properties of non-aging linear viscoelastic and hierarchical composite materials, reinforced by wavy fibers in two directions. In particular, we investigate the effects that two different temperatures of a polymeric matrix, and the waviness of transversely isotropic fibers made from a carbon nanotube-reinforced polymer, have on the macroscopic viscoelastic behavior of a hierarchical structure. For this purpose, we consider the elastic-viscoelastic correspondence principle and the Laplace-Carson transform, and we apply the three-scale asymptotic homogenization method (AHM). The expressions for the associated local and homogenized problems, and the effective coefficients are derived at each level of organization. We solve the meso and microscale local problems in Laplace-Carson domain, by means of an analytically approach and finite element three- dimensional simulations, respectively. We obtain the effective creep and relaxation behavior of the composite. In addition, we use a numerical algorithm to invert the Laplace-Carson transform and derive the properties in the time domain. Finally, we present the results of our computations. They confirm that temperature and waviness can affect the macroscopic behavior of the hierarchical heterogeneous structure.*

1. INTRODUCTION

By natural evolution, biological systems have combined a variety of constituents with specific properties to create composites with more than two length scales. These naturally occurring materials have inspired the research into synthetic composites for a variety of engineering applications. The goal of many of these hierarchical composites is to obtain better performance materials by intentionally manipulating the complexity and inner design and by ensuring a multi-length scale property control. (see [1]). In particular, hierarchical composites with carbon nanotube additions can change the mobility of polymers or improve mechanical properties.

Recently, several studies have addressed the problem of carbon nanotubes (CNT) as reinforcing agents for a variety of polymeric structures (see [2]). For instance, in [3], the authors studied the effects of alignment orientation, agglomeration and waviness of CNT fillers on the effective stiffness. Another important application of the CNTs is to reinforce the carbon fiber (CF)-reinforced composites. In this regard, the authors in [4] predict the effective elastic properties of randomly dispersed CNT-coated CF-reinforced hybrid composites. Moreover, in [5], buckling and postbuckling analysis of CF-reinforced composite laminated plate with and without CNT reinforcement are performed.

On the other hand, a variety of phenomena induced in the manufacturing process of conventional and hierarchical composite materials can affect the macroscopic behavior of the structure. Some

examples of these are the wavy effects in the fibers and the high temperatures, mostly inevitable in the fabrication of polymer nanocomposites.

Multiscale asymptotic homogenization methods have proved to be advantageous in the description of the coarse scale mechanics of hierarchical composite materials (see [6,7]). They encode the information available at the smaller scales into the so-called effective coefficients and use them to predict the macroscopic behavior at its larger scales. The homogenization procedure requires the solution of cell problems with input data corresponding to the homogenized material properties resulting in previous steps, however the analytical solution of such problems has been derived for only a few composite. A suitable alternative comes from the numerical approaches based on the finite element method (FEM). They offer a potential alternative to solve the local problems.

In the present work, we apply the three-scale asymptotic homogenization method to investigate the effects that two different temperatures of a polymeric matrix, and the waviness of fibers made from a carbon nanotube-reinforced polymer, have on the macroscopic viscoelastic response of a hierarchical heterogeneous structure with reinforcement in orthogonal directions.

2. FORMULATION OF THE LINEAR VISCOELASTIC PROBLEM

We consider a linear viscoelastic and hierarchical composite material $\Omega \subset \mathbb{R}^3$ with periodic structure at the different levels of organization and well-separated length scales ℓ_1, ℓ_2 and L (see Fig. 1). Here, we define the dimensionless, scaling parameters, as follows,

$$\varepsilon_1 := \frac{\ell_1}{L} \ll 1 \quad \text{and} \quad \varepsilon_2 := \frac{\ell_2}{L} \ll \varepsilon_1. \tag{1}$$

In addition, x, y and z stand for the macroscopic, mesoscopic and the microscopic spatial variables, respectively and they are related through the expressions,

$$y = \frac{\rho^{(y)}(x)}{\varepsilon_1} \quad \text{and} \quad z = \frac{\rho^{(z)}(x)}{\varepsilon_2}, \tag{2}$$

wherein $\rho^{(\alpha)}$ with $\alpha = y,z$ represent the so-called stratified functions (see [8]).

2.1. Statement of the problem

In the model, we assume the constitutive response of the phases to be linear viscoelastic, and we neglect inertia and external volume forces in Ω. Moreover, we further impose continuity conditions for displacements and tractions on both Γ^{ε_1} and Γ^{ε_2}, i.e. the matrix and the sub-phases are perfectly bonded. Then, the balance of linear momentum in Ω, reads

$$\begin{aligned} &\nabla \cdot \boldsymbol{\sigma}^{\varepsilon}(x,t) = \mathbf{0} && \text{in } \Omega \setminus (\Gamma^{\varepsilon_1} \cup \Gamma^{\varepsilon_2}) \times \mathbb{R}, \\ &\left[\!\left[\boldsymbol{u}^{\varepsilon}(x,t) \right]\!\right] = \mathbf{0}, \qquad &&\left[\!\left[\boldsymbol{\sigma}^{\varepsilon}(x,t) \cdot \boldsymbol{n}^{(\alpha)} \right]\!\right] = \mathbf{0} \qquad (\alpha = y,z). \end{aligned} \tag{3}$$

In Eq. (3), $\boldsymbol{\sigma}^{\varepsilon}$ represents the second-order stress tensor, $\boldsymbol{u}^{\varepsilon}$ stands for the displacement field, $\boldsymbol{n}^{(\alpha)}$ with $\alpha = y,z$ represent the respectively outward unit vectors to the surfaces Γ^{ε_1} and Γ^{ε_2}. In addition, the operator $\left[\!\left[\phi^{\varepsilon} \right]\!\right]$ denotes the jump of ϕ^{ε} across the interface between the constituents. We also assume boundary conditions on $\partial\Omega \times \mathbb{R}$ and initial conditions in $\Omega \times \{0\}$. From now on, we consider the notation $\phi^{\varepsilon}(x,t) = \phi(x,y,z,t)$, where ϕ^{ε} is assumed to be periodic in y and z.

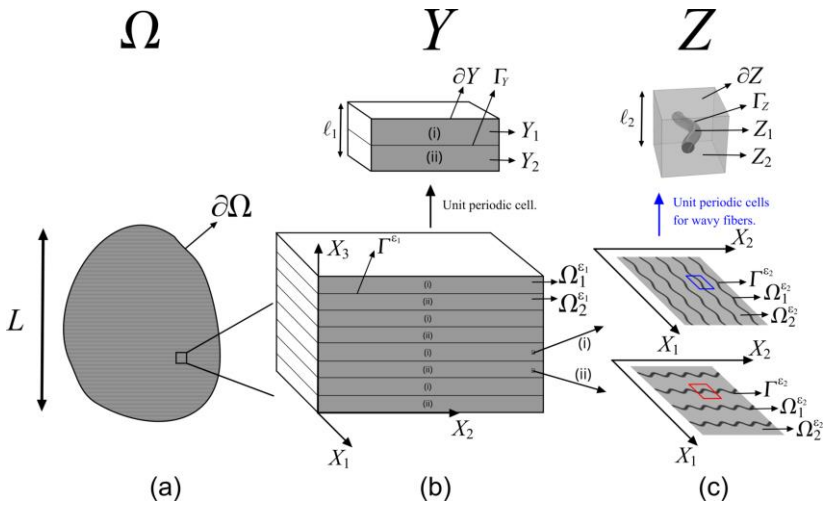

Figure 1 – (a) Macroscale: viscoelastic heterogeneous material. (b) ε_1 -structural level. Mesoscale: Unit periodic cell for the laminated composite. (c) ε_2 -structural level. Microscale: Unit periodic cell for the fiber-reinforced composite. The fibers are non-overlapped and do not intersect the boundaries.

In addition, the scale-dependent constitutive law is stated as follows,

$$\boldsymbol{\sigma}^{\varepsilon}(x,t)=\int_0^t \mathcal{R}^{\varepsilon}(x,t-\tau):\frac{\partial \boldsymbol{\xi}(\boldsymbol{u}^{\varepsilon}(x,\tau))}{\partial \tau}\,d\tau, \tag{4}$$

where $\mathcal{R}^{\varepsilon}$ represents the fourth-order tensor of relaxation moduli, equipped with symmetry properties $\mathcal{R}^{\varepsilon}_{ijkl}=\mathcal{R}^{\varepsilon}_{jikl}=\mathcal{R}^{\varepsilon}_{ijlk}=\mathcal{R}^{\varepsilon}_{klij}$. Furthermore, $\boldsymbol{\xi}$ denotes the second-order strain tensor and it is determined by the formula

$$\boldsymbol{\xi}\left(\boldsymbol{u}^{\varepsilon}(x,t)\right)=\frac{1}{2}\left(\nabla\,\boldsymbol{u}^{\varepsilon}(x,t)+(\nabla\,\boldsymbol{u}^{\varepsilon}(x,t))^{T}\right). \tag{5}$$

Finally, we rely on the elastic-viscoelastic correspondence principle and we rewrite the problem of Eq. (3) in the Laplace-Carson domain (see [8, 9]),

$$\begin{aligned}
&\nabla\cdot\left[\mathcal{R}^{\varepsilon}(x,s):\boldsymbol{\xi}\left(\boldsymbol{u}^{\varepsilon}(x,s)\right)\right]=\mathbf{0} \qquad \text{in } (\Omega\setminus(\Gamma^{\varepsilon_1}\cup\Gamma^{\varepsilon_2}))\times[0,+\infty),\\
&\left[\!\left[\boldsymbol{u}^{\varepsilon}(x,s)\right]\!\right]=\mathbf{0}, \quad \left[\!\left[\mathcal{R}^{\varepsilon}(x,s):\boldsymbol{\xi}\left(\boldsymbol{u}^{\varepsilon}(x,s)\right)\cdot\boldsymbol{n}^{(\alpha)}\right]\!\right]=\mathbf{0} \qquad (\alpha=y,z),
\end{aligned} \tag{6}$$

Here, the variable s represents the Laplace-Carson space. Additionally, we derive the boundary conditions on $\partial\Omega\times[0,+\infty)$ and the initial condition in $\Omega\times\{0\}$.

3. BRIEF DESCRIPTION OF THE METHOD

In this section, we jointly apply the methodologies described in [8, 9, 10] to compute the effective properties of a hierarchical composite material similar to the one shown in Fig. 1. As starting point, we obtain the main theoretical results of the three scale asymptotic homogenization method. Then, we solve the local problems and compute the effective coefficients at the different structural levels.

3.1. Three-scale asymptotic homogenization approach

Since each field and material property $\phi^{\varepsilon}(x,s)$ is assumed to be regular in x and periodic in y and z, according to the chain rule, the following relation is obtained

$$\frac{\partial \phi_i^{\varepsilon}(x,s)}{\partial x_j} = \frac{\partial \phi_i(x,y,z,s)}{\partial x_j} + \frac{1}{\varepsilon_1}\frac{\partial \rho_l^{(y)}(x)}{\partial x_j}\frac{\partial \phi_i(x,y,z,s)}{\partial y_l} + \frac{1}{\varepsilon_2}\frac{\partial \rho_m^{(z)}(x)}{\partial x_j}\frac{\partial \phi_i(x,y,z,s)}{\partial z_m}. \tag{7}$$

Using a similar idea, Eq. (5) becomes,

$$\xi_{kl}\left(\phi^{\varepsilon}(x,s)\right) = \xi_{kl}\left(\phi(x,y,z,s)\right) + \frac{1}{\varepsilon_1}\xi_{kl}^{(y)}\left(\phi(x,y,z,s)\right) + \frac{1}{\varepsilon_2}\xi_{kl}^{(z)}\left(\phi(x,y,z,s)\right), \tag{8}$$

where

$$\xi_{kl}^{(\alpha)}\left(\phi(x,y,z,s)\right) = \frac{1}{2}\frac{\partial}{\partial \alpha_m}\left(\frac{\partial \rho_m^{(\alpha)}(x)}{\partial x_l}\phi_k(x,y,z,s) + \frac{\partial \rho_m^{(\alpha)}(x)}{\partial x_k}\phi_l(x,y,z,s)\right) \quad (\alpha = y,z). \tag{9}$$

Moreover, the solution of the problem of Eq. (6) is proposed as follows

$$\begin{aligned} \boldsymbol{u}^{\varepsilon}(x,s) &= \boldsymbol{u}^{(0)}(x,y,z,s) + \sum_{i=1}^{+\infty}\boldsymbol{u}^{(i)}(x,y,z,s)\varepsilon_1^i + \sum_{i=1}^{+\infty}\tilde{\boldsymbol{u}}^{(i)}(x,y,z,s)\varepsilon_2^i, \\ &= \tilde{\boldsymbol{u}}^{(0)}(x,y,z,s) + \sum_{i=1}^{+\infty}\tilde{\boldsymbol{u}}^{(i)}(x,y,z,s)\varepsilon_2^i. \end{aligned} \tag{10}$$

At this point, as a consequence of the application of the three-scale asymptotic homogenization method, the problems for the parameters ε_2 and ε_1 are analyzed in that order. The results are summarized as follows,

$$\begin{aligned} &\tilde{u}_m^{(1)}(x,y,z,s) = \tilde{\chi}_{klm}(x,y,z,s)\tilde{U}_{kl}^{(0)}(x,y,s), \\ &\tilde{U}_{kl}^{(0)}(x,y,s) = \xi_{kl}\left(\tilde{\boldsymbol{u}}^{(0)}(x,y,s)\right) + \varepsilon_1^{-1}\xi_{kl}^{(y)}\left(\tilde{\boldsymbol{u}}^{(0)}(x,y,s)\right). \\ &\boldsymbol{u}^{(0)} = \boldsymbol{u}^{(0)}(x,s), \quad u_m^{(1)}(x,y,s) = \chi_{klm}(x,y,s)\xi_{kl}\left(\boldsymbol{u}^{(0)}(x,s)\right). \end{aligned} \tag{11}$$

In addition, we derive the homogenized problems at ε_1-structural level and at the macroscale,

$$\left(\frac{\partial}{\partial x_j} + \varepsilon_1^{-1}\frac{\partial \rho_n^{(y)}}{\partial x_j}\frac{\partial}{\partial y_n}\right)\breve{\mathcal{R}}_{ijkl}\tilde{U}_{kl}^{(0)} = 0 \text{ and } \frac{\partial}{\partial x_j}\left[\mathcal{R}_{ijkl}^{(*)}\xi_{kl}\left(\boldsymbol{u}^{(0)}\right)\right] = 0, \tag{12}$$

which are defined in $\Omega_1^{h_{\varepsilon_1}}\times[0,+\infty)$ and $\Omega^h\times[0,+\infty)$, respectively. For the latter, we also obtain boundary conditions on $\partial\Omega^h\times[0,+\infty)$ and the initial condition in $\Omega^h\times\{0\}$. The expressions for the effective coefficients shown in Eq. (12) are given as follows,

$$\breve{\mathcal{R}}_{ijkl} = \left\langle \mathcal{R}_{ijkl} + \mathcal{R}_{ijpq}\xi_{pq}^{(z)}\left(\tilde{\boldsymbol{\chi}}_{kl}\right)\right\rangle_z \text{ and } \mathcal{R}_{ijkl}^{(*)} = \left\langle \breve{\mathcal{R}}_{ijkl} + \breve{\mathcal{R}}_{ijpq}\xi_{pq}^{(y)}\left(\boldsymbol{\chi}_{kl}\right)\right\rangle_y, \tag{13}$$

where the notation $\langle\bullet\rangle_{\alpha}$ with $\alpha = y,z$ defines the cell average operator.

3.2. Effective coefficients at ε_1-structural level. Computational approach

The ε_2-local problems obtained via the three-scale asymptotic homogenization method are solved by means of finite element three-dimensional simulations. Specifically, we use the finite element software COMSOL Multiphysics® and LiveLink™ for Matlab® scripting. As starting point, we assume a z- constant expression for the relaxation moduli ($\mathcal{R}_{ijpq}$) in each phase of the periodic cell Z, i.e.

$$\mathcal{R}_{ijpq}(z,s)=\begin{cases}\mathcal{R}^{(1)}_{ijpq}(s) & : \quad z\in Z_1,\\ \mathcal{R}^{(2)}_{ijpq}(s) & : \quad z\in Z_2,\end{cases} \tag{14}$$

Thus, ε_2-local problems become,

$$\begin{aligned}
&\frac{\partial}{\partial z_j}\Big[\mathcal{R}^{(\alpha)}_{ijpq}(s)\xi^{(z)}_{pq}(\tilde{\chi}^{(\alpha)}_{kl}(z,s))\Big]=0 \quad \text{in } Z_\alpha\times[0,+\infty) \ \ (\alpha=1,2),\\
&\tilde{\chi}^{(1)}_{klm}(z,s)=\tilde{\chi}^{(2)}_{klm}(z,s) \quad \text{on } \Gamma_Z\times[0,+\infty),\\
&\Big(\mathcal{R}^{(1)}_{ijpq}(s)\xi^{(z)}_{pq}(\tilde{\chi}^{(1)}_{kl}(z,s))\Big)n^{(z)}_j-\Big(\mathcal{R}^{(2)}_{ijpq}(s)\xi^{(z)}_{pq}(\tilde{\chi}^{(2)}_{kl}(z,s))\Big)n^{(z)}_j=f^{(z)}_{ikl}(s) \ \text{ on } \Gamma_Z\times[0,+\infty).\\
&\text{Initial condition in } Z\times\{0\},\\
&\tilde{\chi}_{klm}(z,s)=0.
\end{aligned} \tag{15}$$

We notice that, the stress jump conditions obtained in Eq. (15.3) led to the interface loads $f^{(z)}_{ikl}(s):=\Big(\mathcal{R}^{(2)}_{ijkl}(s)-\mathcal{R}^{(1)}_{ijkl}(s)\Big)n^{(z)}_j$. They occur as a consequence of the discontinuities of the coefficients between the host medium (matrix) and the sub-phases, and represent the driving force to obtain nontrivial local problems solutions.

3.3. Effective coefficients at the macroscale. Laminated composites with generalized periodicity

Taking into account Voigt's notation, the effective coefficients of Eq. (13)-(right) are written,

$$\begin{aligned}
\mathcal{R}^{(*)}_{ij}=\Big\langle &\breve{\mathcal{R}}_{ij}+\Big(\breve{\mathcal{R}}_{i1}\frac{\partial\rho^{(y)}}{\partial x_1}+\breve{\mathcal{R}}_{i6}\frac{\partial\rho^{(y)}}{\partial x_2}+\breve{\mathcal{R}}_{i5}\frac{\partial\rho^{(y)}}{\partial x_3}\Big)\frac{\partial\chi_{j1}}{\partial y}+\Big(\breve{\mathcal{R}}_{i6}\frac{\partial\rho^{(y)}}{\partial x_1}\\
&+\breve{\mathcal{R}}_{i2}\frac{\partial\rho^{(y)}}{\partial x_2}+\breve{\mathcal{R}}_{i4}\frac{\partial\rho^{(y)}}{\partial x_3}\Big)\frac{\partial\chi_{j2}}{\partial y}+\Big(\breve{\mathcal{R}}_{i5}\frac{\partial\rho^{(y)}}{\partial x_1}+\breve{\mathcal{R}}_{i4}\frac{\partial\rho^{(y)}}{\partial x_2}+\breve{\mathcal{R}}_{i3}\frac{\partial\rho^{(y)}}{\partial x_3}\Big)\frac{\partial\chi_{j3}}{\partial y}\Big\rangle_y.
\end{aligned} \tag{16}$$

Similarly, ε_1-cell problems obtained via AHM, are stated as follows,

$$\frac{\partial}{\partial y}\left(B_{ij}+A_{ik}\frac{\partial\chi_{jk}}{\partial y}\right)=0, \tag{17}$$

where

$$\begin{aligned}
&B_{1j}=\frac{\partial\rho^{(y)}}{\partial x_1}\breve{\mathcal{R}}_{1j}+\frac{\partial\rho^{(y)}}{\partial x_2}\breve{\mathcal{R}}_{6j}+\frac{\partial\rho^{(y)}}{\partial x_3}\breve{\mathcal{R}}_{5j}, \quad B_{2j}=\frac{\partial\rho^{(y)}}{\partial x_1}\breve{\mathcal{R}}_{6j}+\frac{\partial\rho^{(y)}}{\partial x_2}\breve{\mathcal{R}}_{2j}+\frac{\partial\rho^{(y)}}{\partial x_3}\breve{\mathcal{R}}_{4j},\\
&B_{3j}=\frac{\partial\rho^{(y)}}{\partial x_1}\breve{\mathcal{R}}_{5j}+\frac{\partial\rho^{(y)}}{\partial x_2}\breve{\mathcal{R}}_{4j}+\frac{\partial\rho^{(y)}}{\partial x_3}\breve{\mathcal{R}}_{3j} \quad \text{and} \quad A_{ik}=\frac{\partial\rho^{(y)}}{\partial x_j}\breve{\mathcal{R}}_{ijkl}\frac{\partial\rho^{(y)}}{\partial x_l}.
\end{aligned}$$

4. NUMERICAL RESULTS AND DISCUSSION

In this section, we take inspiration from [11] and consider a hierarchical structure with two different levels of organization as shown in Fig. 1. In our particular case, Ω is a two-phases composite consisting of a viscoelastic laminated structure (matrix), in which each layer is reinforced by wavy fibers. It is worth to point out that layers (i) and (ii) in Fig. 1 possess orthogonal directions.

Regarding the fibers, the authors in [12] present a technique for developing constitutive models for carbon nanotube-reinforced polymer. They consider the nanotube, the local polymer near the nanotube, and the nanotube/polymer interface to be an effective continuum fiber. Here, we assume the properties of those fibers as input values for the present model. Such properties are listed in Tab. 1. In addition, we deal with the waviness (w) of the fiber in the unit periodic cell (see Fig.1 (c)) by means of the function $f(t) = H\sin(2\pi t / L)$ with $t \in [-1/4,\ 3/4]$. We notice that the waviness is defined as follows $w := H / L$. Additionally, we consider the same value of w for both, layers (i) and (ii).

Table 1 – Elastic moduli of transversely isotropic fibers.

$E_L^{(f)}$ (GPa)	$\mu_T^{(f)}$ (GPa)	$\mu_L^{(f)}$ (GPa)	$K_T^{(f)}$ (GPa)	$\nu_{LT}^{(f)}$
450.4	4.4	27.0	9.9	0.4242

On the other hand, a thermoplastic polyimide LaRc-SI with a 3% molecular weight offset is taken as the isotropic viscoelastic matrix (see [13]). The polymeric matrix creep response is well modeled by the power-law,

$$S(t) = D_0 + D_1 t^n, \tag{18}$$

where the properties are shown in Tab. 2 for two different temperatures.

Table 2 – Viscoelastic power-law parameters and Poisson's ratios of isotropic matrix at two temperatures.

Temperature	$D_0^{(m)}$ (GPa^{-1})	$D_1^{(m)}$ (GPa^{-1}hour^{-1})	$n^{(m)}$	$\nu^{(m)}$
213 °C	0.375	0.051606	0.4103	0.367
223 °C	0.313	0.077678	0.4718	0.382

The procedure to compute the effective viscoelastic properties of the hierarchical composite is summarized as follows (see [8, 9, 10]):

(I) We solve the problems in Eq. (15). Then, we substitute the data of $\tilde{\chi}_{kl}$ into Eq. (13)-(left) and we calculate the effective coefficients at the ε_1-structural level.

(II) At this point, it is possible to solve analytically the system of Eq. (17) and obtain the expressions for $\partial\chi_{jk} / \partial y$.

(III) Finally, the effective properties are obtained at the macroscale by means of Eq. (16).

In Fig. 2, we show the results for the effective Young ($E_1^{(*)}$) and shear modulus ($\mu_{12}^{(*)}$), respectively. In the computations, we consider a fiber volume fraction of 10%. Furthermore, we deal with two different temperatures for the matrix, i.e. $T = [213\ ^{o}C,\ 223\ ^{o}C]$ and several values for the waviness of the fibers, i.e. $w = [0,\ 0.05,\ 0.1,\ 0.15]$.

The results confirm that these two manufacturing agents (temperature and waviness) have an influence on the macroscopic viscoelastic response of the hierarchical heterogeneous structure. As illustrated in Fig. 2, the Young (shear) modulus decreases (increases) evenly as the waviness rises, and in both cases, an increase in temperature leads to a decrease in the properties.

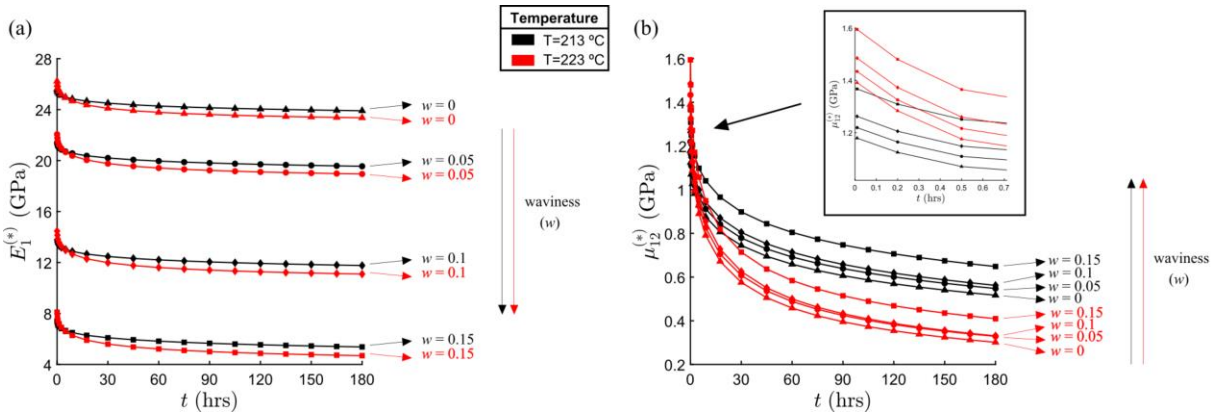

Figure 2 – Computations of the effective (a) Young and (b) shear modulus for the hierarchical composite.

5. CONCLUSIONS

In this work, we studied the overall viscoelastic response of fiber-reinforced hierarchical composites. We obtained an effective characterization of the heterogeneous system by means of the three-scale asymptotic homogenization method. We solved the local problems and computed the effective coefficients at each level of organization. Finally, we addressed the effects that two different temperatures of a polymeric matrix, and the waviness of reinforced fibers in orthogonal directions, have on the effective response of a hierarchical heterogeneous structure.

Further generalizations of the present work include the consideration of imperfect contact conditions between the interfaces of the constituents. In addition, the approach can be extended to the case of ageing viscoelastic composite materials.

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