Upscaling of viscoelastic properties of highly-filled composites: Investigation of matrix–inclusion-type morphologies with power-law viscoelastic material response

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In this paper, homogenization schemes for upscaling of elastic properties in the framework of continuum micromechanics are extended towards upscaling of viscoelastic material properties. Hereby, the Laplace–Carson transform method is applied to the Mori–Tanaka scheme, the self-consistent scheme, and the generalized self-consistent scheme and solved numerically by the Gaver–Stehfest algorithm. The performance of the so-obtained upscaling schemes is: (i) illustrated for an academic example (a 2-phase composite with Maxwellian-type creep response of the phases) and (ii) assessed considering a polyester matrix/marble dust filler composite with respective experimental data taken from the literature. Hereby, for the investigated range of volume fractions of inclusions, ranging from 29 vol.% to 55 vol.%, and a matrix/inclusion-type morphology, the transformed generalized self-consistent scheme emerged as the most suitable scheme for determination of the effective viscoelastic properties of this highly-filled composite material, resulting in a sound representation of the experimental data.

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1. Introduction

An important class of man-made composite materials are characterized by a matrix–inclusion-type morphology. As regards the determination of effective elastic properties of these composite materials, the Mori–Tanaka scheme [39] – accounting for this matrix–inclusion-morphology within the continuum micromechanics framework – may be used. However, as the volume fraction of the inclusions in the composite material increases, i.e., in case of so-called highly-filled composites, preconditions for the applicability of the Mori–Tanaka scheme are violated.1 In case the particle-size distribution of the inclusion phase is known, probably separated by a thin film of the matrix material, and the identification of a distinct continuous phase becomes difficult.

In such situations, the so-called self-consistent scheme [19,26], derived under the assumption of a polycrystalline microstructure, with no material phase acting as a continuous phase, may be more appropriate. Hereby, the determination of effective properties is based on a model configuration characterized by the different material phases embedded in the effective medium. In contrast to the Mori–Tanaka scheme, the self-consistent scheme is an implicit method which can only be solved in an iterative manner.

The mechanical response of the so-called generalized self-consistent scheme [9,10,3] lies between the Mori–Tanaka scheme and the self-consistent scheme. The underlying model configuration is characterized, in case of a 2-phase composite with spherical inclusions, by a particle of radius \( a \) surrounded by a matrix shell (inner radius \( a \), outer radius \( b \)), where \( a^2/b^3 \) is equal to the volume fraction of the inclusion phase. The particle and the matrix shell are, in turn, embedded in the effective medium.

In case the particle-size distribution of the inclusion phase is available, there are several homogenization schemes taking this information into account, including the double inclusion method [20], the Ponte Castañeda and Willis method [41], the Kuster-Toksoz model [27,43,46], and the interaction direct derivative estimate [51]. For a review of these methods, see Hu and Weng [23,24].

1.1. Literature review – previous work on upscaling of viscoelastic composites

In case of linear non-aging viscoelastic material behavior, the Laplace–Carson transform technique can be applied in order to extend the mentioned homogenization schemes towards upscaling of
viscoelastic properties. This so-called correspondence principle allows to transfer a viscoelastic problem into a symbolic elastic problem with known solution [30]. Previous applications of the correspondence principle in the scope of homogenization can be found in:

- Hashin [18] and Christensen [8] using the composite sphere assemblage model [17];
- Laws and McLaughlin [29]; Rougier et al. [43]; Beurthey and Zaoui [5] using the self-consistent scheme;
- Wang and Weng [48]; Brinson and Lin [7]; Schjedt-Thomsen and Pyrz [45]; Li et al. [35]; Le et al. [30]; Ricaud and Masson [42] using the Mori–Tanaka scheme;
- Rougier et al. [43]; Beurthey and Zaoui [5] using the generalized self-consistent scheme; Li and Weng [34] using the latter in the context of coated fiber/matrix composites; for the case of multiple coated inclusions, see Beurthey and Zaoui [5];
- Li and Hu [33] using the generalized Kuster–Toksoz model [36].

With regard to the viscoelastic behavior of the phases constituting the composite material, the existing literature deals with the Maxwell model [43], the generalized Maxwell model (Maxwell bodies in parallel) [29,5,30,42], and the 4-parameter model (Maxwell and Kelvin–Voigt bodies in series) [48,34,33].

The effective behavior in the time domain is obtained by inverse Laplace–Carson transformation of the expressions obtained from the correspondence principle applied to the respective homogenization schemes based on continuum micromechanics. However, only in limited cases an analytical solution for the inverse transformation exists, see, e.g., Rougier et al. [43] (in the form of relaxation spectra) and Section 4 of this paper for Maxwellian type behavior of the phases constituting the composite material, Beurthey and Zaoui [5] for the generalized Maxwell and Kelvin–Voigt model [30] assuming constant Poisson’s ratio or constant bulk modulus of the viscoelastic matrix, and with limited microscopic retardation times and number of phases. In most cases, the inverse Laplace–Carson transformation has to be carried out numerically. For a review of numerical methods, see Abate and Valkó [1] and references therein; Davies and Martin [13]; Narayan and Beskos [40]; Duffy [14]; Chapter 19 in Davies [12]. The most commonly employed algorithms (in various engineering fields) may be classified according to the used approach [1], i.e., (i) Fourier series expansion, (ii) Laguerre function expansion, (iii) deform of the Bromwich contour, and (iv) combinations of Gaver functionals. For homogenization of viscoelastic properties of composites in the framework of continuum micromechanics, the so-called collocation method [44] and the so-called multidata method [11] have been developed and recently improved with regard to error estimation [32]. The latter methods are based on a Dirichlet series expansion and have been employed by, e.g., Laws and McLaughlin [29]; Masson and Zaoui [38]. Similarly, an internal-variable formulation, also making use of Dirichlet series expansion of effective properties was recently employed with regard to aging viscoelastic behavior [42]. The use of a time-integration approach based on an internal-variable formulation operating in the time domain, i.e., not making use of the Laplace–Carson transform method, has originally been proposed by Lahellec and Suquet [28].

Wang and Weng [48] used a method based on Legendre polynomial expansion for inverse Laplace–Carson transformation [2]. In this work, the so-called Gaver–Steinhardt algorithm [46] is employed. This algorithm uses a linear convergence acceleration scheme to the series (combination of Gaver functionals) that approximates the inverse. The method has recently been improved as the so-called Gaver–Wynn–Rho algorithm which uses a nonlinear convergence scheme and multi-precision computing enabling an a priori accuracy estimation of the inverse transform [47,1].

1.2. Outline

This paper focuses on the model configuration best suited for determination of effective viscoelastic properties of highly-filled composite materials with power-law viscous behavior. Departing from three classical upsampling schemes for effective elastic properties based on continuum micromechanics reviewed in Section 2, these schemes are extended towards upsampling of viscoelastic properties in Section 3, employing the aforementioned Laplace–Carson transform method. The effective properties determined within the presented upsampling framework are illustrated in Section 4 for a 2-phase composite with Maxwellian viscoelastic material response of the phases. Finally, the developed upsampling approach is applied to a polyester matrix/marble dust filler composite material with power-law viscous behavior [22] in Section 5, giving insight into the performance of the upsampling framework when applied to a highly-filled composite. The added value of the present study is the comparison of different underlying micromechanical models spanning a wide range of microstructural configurations and featuring actual viscoelastic material behavior supported by experimental data.

2. Review of classical upsampling schemes for elastic properties based on continuum micromechanics

Homogenization schemes based on continuum micromechanics consider a representative volume element (RVE) subjected to a homogeneous strain E at its boundary. These schemes depart from the definition of the so-called strain-localization tensor A, linking the effective strain tensor E with the local strain tensor e at the location x:

\[ e(x) = A(x) : E. \]  

(1)

The effective strain tensor E represents the volume average of the local strain tensor e:

\[ E = \frac{1}{V} \int_V e(x) \, dV. \]  

(2)

Inserting Eq. (1) into Eq. (2), one gets \( E = (A(x))_V : E \) and, thus, \( (A(x))_V = 1 \). Considering an ellipsoidal inclusion “i” embedded in a reference medium characterized by the material tensor \( C_0 \), the strain-localization tensor A within the domain “i” is constant and given as [16]

\[ A_i = 1 + S_i : (C_0 - C_1) \]  

(3)

with \( C_1 \) as the material tensor of the inclusion “i”. \( S_i \) denotes the Eshelby tensor, conditioned by the geometric properties of the inclusion and the elastic properties of the reference medium.

The volume average of the local stress tensor \( \sigma(x) \) determines the effective stress tensor \( \Sigma \):

\[ \Sigma = \frac{1}{V} \int_V \sigma(x) \, dV. \]  

(4)

Considering a linear-elastic constitutive law for the rth material phase, linking the local strain tensor with the local stress tensor, \( \sigma_i(x) = C_r : e_i(x) \) and Eq. (1) in Eq. (4) one gets

\[ \Sigma = (C_r : A_r)(x)_V : E. \]  

(5)

Comparison of Eq. (6) with \( (\Sigma = C_{eff} : E)^2 \) gives access to the effective material tensor \( C_{eff} \):

\[ C_{eff} = C_r : (A_r)_V : C_r^{-1}. \]  

Levin's theorem states that the effective state equation is of the same form as the local state equation [49].
\[ C_{\text{eff}} = \langle \mathbf{A}(\mathbf{x}) : \mathbf{A}(\mathbf{x}) \rangle_v. \]  

(7)

Considering the morphology of the composite material, the unknown strain localization tensor \( \mathbf{A} \), which so far is available for the inclusion only (Eq. (3)), can be estimated based on the choice of \( C_0 \):

- In case the microstructure is characterized by a distinct matrix/inclusion-type morphology, \( C_0 \) is set equal to the material tensor of the matrix material \( C_m \). This estimation leads to the Mori–Tanaka scheme [39].
- For a polycrystalline microstructure, i.e., the material phases are equally dispersed, and none of them forms a matrix, \( C_0 \) is replaced by the effective material tensor \( C_{\text{eff}} \). The obtained implicit method is referred to as self-consistent scheme [19,26].

Elementary algebraic manipulations (see Appendix A) give access to the effective material tensor as

\[ C_{\text{eff}} = f_0 C_0 : \langle \mathbf{A}(\mathbf{x}) \rangle_v + f_i C_i : \mathbf{A}_i \]

\[ = \left\{ \sum_{r \neq i} f_r C_r : \left[ 1 + \mathbf{S}_r : \left( C_0^{-1} : \mathbf{S}_r - 1 \right) \right]^{-1} \right\} \]

\[ + \left\{ \sum_{r \neq i} f_r C_r : \left[ 1 + \mathbf{S}_r : \left( C_0^{-1} : \mathbf{S}_r - 1 \right) \right]^{-1} \right\}, \]  

(8)

where \( C_0 \) is set equal to the matrix material tensor \( C_m \) and the effective material tensor \( C_{\text{eff}} \) for the Mori–Tanaka scheme and the self-consistent scheme, respectively. In the following, Eq. (8) is specialized for isotropic material behavior given in terms of the effective volumetric compliance and deviatoric compliance, \( J_{\text{eff}} \) and \( \nu_{\text{eff}} \), respectively.

- **Self-consistent scheme:**

\[ J_{\text{eff}} = \frac{\sum_{r \neq i} f_r}{J_0 \sum_{r \neq i} f_r} \left[ 1 + \frac{\nu_{\text{eff}} - 1}{J_0 \nu_{\text{eff}} - 1} \right]^{-1} \]  

with \( \alpha = \frac{3 \nu_{\text{eff}}}{3 \nu_{\text{eff}} + 4 \nu_{\text{eff}}} \),

\[ \nu_{\text{eff}} = \frac{6 (\nu_{\text{eff}} + 2 J_{\text{eff}})}{5 (3 \nu_{\text{eff}} + 4 \nu_{\text{eff}})}. \]  

(9)

where \( \alpha \) and \( \beta \) represent the volumetric and deviatoric part of the Eshelby tensor \( \mathbf{S} \) specialized for spherical inclusions;

- **Mori–Tanaka scheme:**

\[ J_{\text{eff}} = \frac{f_m + f_i}{f_m + f_i} \left[ 1 + \frac{\nu_{\text{eff}} - 1}{J_0 \nu_{\text{eff}} - 1} \right]^{-1} \]  

with \( \alpha = \frac{3 \nu_{\text{eff}}}{3 \nu_{\text{eff}} + 4 \nu_{\text{eff}}} \),

\[ \nu_{\text{eff}} = \frac{6 (\nu_{\text{eff}} + 2 J_{\text{eff}})}{5 (3 \nu_{\text{eff}} + 4 \nu_{\text{eff}})}. \]  

(10)

### 2.1. Generalized self-consistent scheme

Similar to the self-consistent scheme and the Mori–Tanaka scheme, the generalized self-consistent scheme is based on a basic result of Eshelby: the relation between the strain energy \( U \) for a homogeneous medium containing an inclusion, and the strain energy \( U_0 \) for this homogeneous medium with no inclusion reads

\[ U = U_0 - \frac{1}{2} \int_S \left( T_{\mu} u_{\mu} - T_{\mu} u_0^\mu \right) dS, \]  

(11)

where \( S \) represents the surface of the inclusion, \( T_{\mu} \) and \( u_0^\mu \) are traction and displacements in the medium containing no inclusions, and \( T_{\mu} \) and \( u_{\mu} \) are the corresponding quantities at the same point in the medium containing the inclusion. \( U \) is equal to the strain energy of the effective medium \( U_{\text{eff}} \) (see Fig. 1a).

For the configuration of a coated inclusion surrounded by the effective medium, \( U_0 = U_{\text{eff}} \) (see Fig. 1b). Since \( U_{\text{eff}} = U \), one gets \( U = U_0 \), reducing Eq. (11) to

\[ \int_S \left( T_{\mu} u_{\mu} - T_{\mu} u_0^\mu \right) dS = 0. \]  

(12)

The resulting effective elastic compliance for a 2-phase composite determined by the generalized self-consistent scheme is summarized in Appendix B. For the application of the generalized self-consistent scheme towards multi-phase composites see Benveniste [3].

### 3. The Laplace–Carson transform method for the solution of the viscous upscaling problem

Viscous material response is characterized by: (i) an increase of deformation during constant loading (creep) and (ii) a decrease of stress for constrained deformation (relaxation). The viscous response is commonly described by the creep compliance \( J_0 (\text{Pa}^{-1}) \) and the relaxation modulus \( R_0 (\text{Pa}) \), both dependent on time. The creep compliance associated with uniaxial loading is determined as

\[ J(t) = \frac{e(t)}{\sigma_0} \]  

(13)

with \( e(t) \) denoting the measured strain and \( \sigma_0 \) representing the applied constant stress. The relaxation modulus, on the other hand, is determined from the measured stress decrease \( \sigma(t) \) in consequence of a constant strain \( e_0 \) as

\[ R(t) = \frac{\sigma(t)}{e_0}. \]  

(14)

Introducing the Boltzmann convolution integral, Eqs. (13) and (14) can be expanded towards variable (non-constant) stress or strain histories, respectively:

\[ e(t) = \int_0^t J(t - \tau) \frac{\partial \sigma(\tau)}{\partial \tau} d\tau \]  

and \( \sigma(t) = \int_0^t R(t - \tau) \frac{\partial e(\tau)}{\partial \tau} d\tau. \)  

(15)

where \( \tau \) denotes the time instant of loading. Applying the Laplace transformation to Eq. (15) gives

\[ \hat{e}(p) = p \hat{J}(p) \hat{e}(p) \quad \text{and} \quad \hat{\sigma}(p) = p \hat{R}(p) \hat{e}(p), \quad \text{with} \quad \hat{p} \hat{J}(p) = \frac{1}{p \hat{R}(p)}. \]  

(16)
considering that the Laplace transform of the convolution integral becomes a multiplication and \( \partial$\cprime$\cprime/\partial t \) turns into $pi$. Considering the definition of the Laplace–Carson transformation\(^3\) as $f' = pf$ in Eq. (16) yields

$$e'(x) = f'(x)\sigma'(x) \quad \text{and} \quad \sigma'(x) = R'(x)e'(x), \quad \text{with} \quad f'(x) = \frac{1}{R(x)}.$$ (20)

The analogous form of $\sigma' = R'e'$ in Eq. (20) and the elastic constitutive law $\sigma = Ee$ is the basis for the “correspondence principle” \([31,6,18,37]\). According to this principle, viscoelastic problems are solved using the respective solution of the elastic problem in the Laplace–Carson domain.

The Laplace–Carson transform method \([31]\) for the solution of linear viscoelastic boundary value problems (BVPs) is characterized by the elimination of the time dependence by applying the Laplace–Carson transform to the field equation (which contains the time dependent moduli) as well as the boundary conditions,\(^4\) and solving the “corresponding” elastic problem in the Laplace–Carson domain. For upscaling, the elastic material parameters are replaced by the Laplace–Carson transform of the respective viscoelastic material parameters. For example the shear compliance functions for different rheological models in both time and Laplace–Carson domain.

\(^3\) Whereas the Laplace transformation of $f(t)$ is defined as

$$\mathcal{L}[f(t)] = \int_0^\infty f(t)e^{-st}dt,$$

with $s$ as the complex variable, the Laplace–Carson transformation of $f(t)$ is given as

$$\mathcal{L}^c[f(t)] = \int_0^\infty f(t)e^{-st}dt.$$ (17)

Hence, $f' = pf$. The inverse Laplace–Carson transformation is defined in the complex plane as

$$\mathcal{L}^{-1}[f(t)] = \frac{1}{2\pi i} \int_{\gamma} f(s)e^{st}d\gamma,$$ (19)

where $\gamma$ is a parallel to the imaginary axis having all poles of $f'(x)$ to the left.

\(^4\) Hence, this method is restricted to BVPs with boundary conditions admitting such an operation, thus with boundary conditions in tractions and displacements fixed in time, as is the case for an inclusion embedded in an (infinite) matrix within the theory of continuum micromechanics.

**Fig. 1.** Strain energy consideration for composite materials.

Applying the Laplace–Carson transform method to the Mori–Tanaka scheme (Eq. (10)) gives

$$f'_{\text{eff}} = \frac{f_m + f_t}{1 + \alpha' \left( \frac{\rho_{\text{ef}}}{\rho_m} - 1 \right)}$$ with $\alpha' = \frac{3f_{\text{dev}}^{\text{vol}}}{3f_{\text{dev}}^{\text{eff}} + 4f_{\text{rot}}^{\text{vol}}},$ and

$$f_{\text{dev}}^{\text{vol}} = \frac{f_m + f_t}{1 + \beta' \left( \frac{\rho_{\text{dev}}^{\text{vol}}}{\rho_m} - 1 \right)}$$ with $\beta' = \frac{6(f_{\text{dev}}^{\text{vol}} + 2f_{\text{rot}}^{\text{vol}})}{5(3f_{\text{dev}}^{\text{vol}} + 4f_{\text{rot}}^{\text{vol}})}.$$ (21)

The solution of the viscoelastic upscaling problem in the time domain is obtained by the inverse Laplace–Carson transformation of Eq. (21). When employing simple rheological models (e.g., the Maxwell model) for the different phases of the composite material, this inverse transformation can be performed analytically. For more complex rheological models (see Table 1), the inverse transformation has to be performed numerically, e.g., in a pointwise manner (for discrete values of $t > 0$) by applying the Gaver–Stehfest algorithm \([46]\). On the other hand, one has to solely rely on numerical methods when applying the Laplace–Carson transform method to the implicit self-consistent scheme (Eq. (9))

$$f_{\text{eff}}^{\text{dev}} = \frac{\sum_{\text{comp}} f_{\text{dev}}^{\text{vol}} \left[ 1 + \alpha' \left( \frac{\rho_{\text{dev}}^{\text{vol}}}{\rho_m} - 1 \right) \right]}{\sum_{\text{comp}} f_{\text{rot}}^{\text{vol}} \left[ 1 + \alpha' \left( \frac{\rho_{\text{rot}}^{\text{vol}}}{\rho_m} - 1 \right) \right]}$$ with $\alpha' = \frac{3f_{\text{dev}}^{\text{vol}}}{3f_{\text{dev}}^{\text{eff}} + 4f_{\text{rot}}^{\text{vol}}},$ and

$$f_{\text{dev}}^{\text{rot}} = \frac{\sum_{\text{comp}} f_{\text{dev}}^{\text{vol}} \left[ 1 + \beta' \left( \frac{\rho_{\text{dev}}^{\text{vol}}}{\rho_m} - 1 \right) \right]}{\sum_{\text{comp}} f_{\text{rot}}^{\text{vol}} \left[ 1 + \beta' \left( \frac{\rho_{\text{rot}}^{\text{vol}}}{\rho_m} - 1 \right) \right]}$$ with $\beta' = \frac{6(f_{\text{dev}}^{\text{vol}} + 2f_{\text{rot}}^{\text{vol}})}{5(3f_{\text{dev}}^{\text{vol}} + 4f_{\text{rot}}^{\text{vol}})}.$$ (22)

Applying the Laplace–Carson transform method to the (explicit) generalized self-consistent scheme (Eqs. (39) and (43) in Appendix B) gives
In order to illustrate the performance of the upscaling framework presented in the previous section, a simple 2-phase composite material is analyzed. Whereas deviatoric Maxwell-type behavior is assigned to the phases, volumetric creep of the two phases is omitted. Table 2 summarizes the employed volume fractions and properties of the material phases.

### 4.1. Upscaling using the Mori–Tanaka scheme

In the first step, the effective creep compliance of a matrix-inclusion-type morphology is determined using the Mori–Tanaka scheme. Inserting the creep compliance of the phases in the Laplace–Carson transform of the volumetric part of the Eshelby tensor \( \chi’ \) (see Eq. (21.1)) implies that deviatoric creep of the matrix material leads to volumetric creep of the composite material. The result from numerical inverse transformation of Eq. (21.1) is shown in Fig. 3. The long-term asymptote in Fig. 3 (thick solid line) can be determined from Eq. (10.1) by setting \( J_\text{dev}^{\text{eff}} \to \infty \) (hence, setting \( \tau = 1 \)) giving \( J_\text{dev}^{\text{eff}}(t \to \infty) = 0.09 \text{ MPa}^{-1} \).

For the present situation of a particle-reinforced composite, the creep compliance corresponding to a uniaxial loading situation, \( f(t) \), may be approximated by

\[
J_\text{eff}(t) = \frac{1}{\mu_\text{eff}} + \frac{1}{\mu_\text{eff}} \exp \left(-\frac{\mu_\text{eff}}{\mu_\text{eff}} t \right) \quad \text{and} \quad f(t) = \frac{J_\text{dev}^{\text{eff}}(t)}{J_\text{dev}^{\text{eff}}(t)} \exp \left(-\frac{\mu_\text{eff}}{\mu_\text{eff}} t \right)
\]

for a 2-phase composite material, where the subscripts “m” and “i” refer to matrix and inclusion, respectively. For determination of \( \Lambda’, B’, \) and \( D’ \) appearing in Eq. (23.1) see Appendix B.

### 4.2. Upscaling using the self-consistent scheme

The situation becomes more complex when investigating a polycrystalline microstructure of the composite material using the self-consistent scheme. As the self-consistent scheme is an implicit scheme with the effective deviatoric compliance \( J_\text{dev}^{\text{eff}} \) appearing on both sides of Eq. (22.2), the ansatz describing \( f(t) \) has to be set a priori and the effective material parameters have to be determined iteratively.

Inspired by the result of the previous subsections, the 4-parameter model (see Table 1) is taken to describe the deviatoric creep compliance of the effective material, with

\[
J_\text{dev}^{\text{eff}}(t) = \frac{1}{\mu_\text{eff}} + \frac{1}{\mu_\text{eff}} \exp \left(-\frac{\mu_\text{eff}}{\mu_\text{eff}} t \right) \quad \text{and} \quad f(t) = \frac{J_\text{dev}^{\text{eff}}(t)}{J_\text{dev}^{\text{eff}}(t)} \exp \left(-\frac{\mu_\text{eff}}{\mu_\text{eff}} t \right)
\]

The effective material parameters are obtained as \( \mu_\text{eff} = 374 \text{ GPa} \), \( \mu_\text{eff} = 11.7 \text{ GPa} \), and \( \mu_\text{eff} = 185 \text{ GPa} \) when setting \( f(t) = f(t) = 1 \) for backtransformation.

The effective response obtained from backtransformation (circles in Fig. 4) is well approximated by the response of the assumed 4-parameter model

\[
J_\text{dev}^{\text{eff}}(t) = \frac{1}{\mu_\text{eff}} + \frac{1}{\mu_\text{eff}} \exp \left(-\frac{\mu_\text{eff}}{\mu_\text{eff}} t \right) \quad \text{and} \quad f(t) = \frac{J_\text{dev}^{\text{eff}}(t)}{J_\text{dev}^{\text{eff}}(t)} \exp \left(-\frac{\mu_\text{eff}}{\mu_\text{eff}} t \right)
\]

### Table 2

<table>
<thead>
<tr>
<th>Material phases r</th>
<th>Phase 1</th>
<th>Phase 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume fraction ( f_r )</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>Elastic properties</td>
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<td></td>
</tr>
<tr>
<td>Shear modulus ( \mu_\text{r} ) (GPa)</td>
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<td>5</td>
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<tr>
<td>Poisson’s ratio ( v_\text{r} ) (-)</td>
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<td>0.25</td>
</tr>
<tr>
<td>Young’s modulus ( E_\text{r} = 2\mu_\text{r}(1 + v_\text{r}) ) (GPa)</td>
<td>25</td>
<td>12.5</td>
</tr>
<tr>
<td>Bulk modulus ( k_\text{r} = E_\text{r}/(1 - 2v_\text{r}) ) (GPa)</td>
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<td>8.33</td>
</tr>
<tr>
<td>Viscous properties</td>
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<tr>
<td>Viscosity ( \eta_\text{r} ) (GPa s)</td>
<td>500</td>
<td>50</td>
</tr>
</tbody>
</table>

5 When setting \( J_\text{dev}^{\text{eff}} = J_\text{dev}^{\text{eff}} = 1 \) in Eq. (22.2), the effective material parameters are obtained as \( \mu_\text{eff} = 374 \text{ GPa} \), \( \mu_\text{eff} = 11.7 \text{ GPa} \), and \( \mu_\text{eff} = 185 \text{ GPa} \) when setting \( f(t) = f(t) = 1 \) for backtransformation.

The obtained result is insensitive to the value of \( J_\text{dev}^{\text{eff}} \).
4.3. Upscaling using the generalized self-consistent scheme

Finally, the two possible model morphologies are investigated using the generalized self-consistent scheme, i.e., (i) phase 1 serving as the matrix material (stiff matrix shell, compliant inclusion) or (ii) phase 2 serving as the matrix material (compliant matrix shell, stiff inclusions). The result for numerical inverse Laplace–Carson transformation of Eq. (23.1), where the deviatoric matrix compliance $J_{\text{dev}}^m/C^3_m$ was set to

\[
\left(i\right) \quad J_{\text{dev}}^m/C^3_m = \frac{1}{\mu_1} + \frac{1}{\mu_{\text{max},1}} \quad \text{and} \quad \left(ii\right) \quad J_{\text{dev}}^m/C^3_m = \frac{1}{\mu_2} + \frac{1}{\mu_{\text{max},2}},
\]

respectively, is shown in Fig. 5. The effective compliance predicted by the self-consistent scheme lies between the two results obtained from the generalized self-consistent scheme. On the other hand, the Mori–Tanaka responses bound the response obtained from the generalized self-consistent scheme.

5. Application to polyester matrix/marble dust filler composite material

In this section the developed upscaling approach is applied to a polyester matrix/marble dust filler composite material tested in Hristova et al. [22], with the size of the filler particles in the range from 2 to 30 $\mu$m. Uniaxial compressive creep tests on prismatic specimens were performed for different filler contents ranging from $f_i = 0$ (polyester matrix only) to $f_i = 0.55$. Table 3 summarizes the employed material properties employed within the upscaling procedure. Hereby, viscous properties of the polyester matrix were determined by backcalculation of the experimentally available
uniaxial compliance for $f_i = 0$ using a deviatoric power-law (PL), reading

$$J^1_{\text{D}}(t) = \frac{1}{3} J^\text{dev}_m(t) + \frac{1}{9} J^\text{vol}_m(t)$$

where volumetric creep was omitted. Fig. 6 depicts the employed experimental data and the backcalculated compliance function according to Eq. (29). For upscaling, i.e., numerical backtransformation using the Mori–Tanaka scheme (Eq. (21)), the self-consistent scheme (Eq. (22)), and the generalized self-consistent scheme (Eq. (23)) the phase compliances where set to
Table 3
Volume fractions and material properties for composite material tested in Hristova and coworkers [22,21].

<table>
<thead>
<tr>
<th>Material phases</th>
<th>Matrix “m”</th>
<th>Inclusion “i”</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>thermoset</td>
<td>filler (marble dust)</td>
</tr>
<tr>
<td>Volume fraction</td>
<td>1.00;0.71;0.62;0.55;0.45</td>
<td>0.00;0.29;0.38;0.45;0.55</td>
</tr>
</tbody>
</table>

Elastic properties

- **Shear modulus** \( \mu_r \) (GPa): 1.845
- **Poisson’s ratio** \( \nu_r \) (-): 0.382
- **Young’s modulus** \( E_r \): 5.1
- **Bulk modulus** \( K_r \): 7.203

**Viscous properties**

- **Deviatoric compliance** \( J_{dev}^{\text{PL},r} \) (GPa\(^{-1}\)): 1.1720
- **Poisson’s ratio** \( \nu_r \): 0.2717

* Determined from backcalculation of uniaxial creep compliance.

Fig. 6. Experimentally obtained uniaxial creep compliance \( J_{1D}^{\text{dev}}(t) \) for matrix material and composite material characterized by different volume fractions of filler \( f_i \).

\[
\begin{align*}
J_{\text{dev}}^m & = \frac{1}{\mu_m} + \frac{J_{\text{dev}}^{\text{PL},m}}{\mu_m^2} \left( \frac{1}{\mu_m^2} \right) J^m_1 \left[ 1 + \frac{J_{\text{dev}}^{\text{PL},m}}{\mu_m^2} \right], \\
J_{\text{vol}}^m & = \frac{1}{K_m},
\end{align*}
\]  

Thus, elastic material response is assigned to the inclusion phase “i” and the volumetric part of the matrix compliance. Fig. 7 shows the result from numerical inverse transformation for the deviatoric compliance. Hereby the discrete points for the (explicit) Mori–Tanaka and generalized self-consistent scheme where approximated by the function

\[
J_{\text{dev}}^{\text{eff}}(t) = \frac{1}{\mu_{\text{eff}}} + J_{\text{dev}}^{\text{PL},\text{eff}} \left( \frac{1}{\mu_{\text{eff}}} \right) J^m_{1,\text{eff}} \left[ 1 + \frac{J_{\text{dev}}^{\text{PL},\text{eff}}}{\mu_{\text{eff}}} \right],
\]  

For the self-consistent scheme, on the other hand, the effective deviatoric compliance function \( J_{\text{dev}}^{\text{eff}}(t) \) was, a priori, assumed to be of power-law type, with \( \mu_{\text{eff}} \) taken as

\[
J_{\text{dev}}^{\text{eff}}(t) = \frac{1}{\mu_{\text{eff}}} + J_{\text{dev}}^{\text{PL},\text{eff}} \left( \frac{1}{\mu_{\text{eff}}} \right) J^m_{1,\text{eff}} \left[ 1 + \frac{J_{\text{dev}}^{\text{PL},\text{eff}}}{\mu_{\text{eff}}} \right].
\]  

and \( J_{\text{dev}}^{\text{eff}} \) and \( \mu_{\text{eff}} \) were determined in an iterative manner. Table 4 summarizes \( J_{\text{dev}}^{\text{eff}} \) and \( \mu_{\text{eff}} \) obtained from the different upscaling schemes for the investigated range of the volume fraction of filler \( f_i \).

Fig. 8 shows the uniaxial compliance given by

\[
J_{\text{1D}}^{\text{eff}}(t) = \frac{1}{\mu_{\text{eff}}} + \frac{1}{\mu_{\text{vol}}} J_{\text{eff}}^{\text{vol}}(t),
\]  

with the Mori–Tanaka scheme providing the largest, the self-consistent scheme the lowest effective compliance. The generalized
self-consistent response resides between these two, reasonably well approximating the experimental data. The effective material properties $J_{\text{dev}}^{\text{eff}}$ and $\lambda_{\text{dev}}^{\text{eff}}$ were also determined by backcalculation using the test data (dash-dotted curves in Fig. 8), where the experimental uniaxial compliance was approximated by

$$J_{1D}^{\text{eff}}(t) = \frac{1}{3} \left[ \frac{1}{\lambda_{\text{dev}}^{\text{eff}}} + J_{\text{dev}}^{\text{eff}}(t) \right] + \frac{1}{3} \frac{1}{\kappa_{\text{dev}}^{\text{eff}}}. \quad (34)$$

Fig. 9 shows the prediction of effective creep parameters $J_{\text{dev}}^{\text{eff}}$ and $\lambda_{\text{dev}}^{\text{eff}}$ for the different upscaling schemes as a function of the volume fraction of filler $f_i$. The results from upscaling is compared to the result from backcalculation of test data in Fig. 8, highlighting the generalized self-consistent scheme as the appropriate upscaling method.

Brinsson and Lin [7] investigated storage and loss moduli of viscoelastic composites by means of: (i) the Fourier-transformed Mori–Tanaka method and (ii) a unit-cell model discretized by finite elements. Hereby, the authors observed, similar to the results in the present study, that with increasing volume fraction of inclusions, compliance predicted by the Mori–Tanaka scheme is progressively larger than the respective result obtained by the unit-cell approach, which supposedly depicts the reality more consistently. This result corroborates the finding, that the Mori–Tanaka scheme is inappropriate for prediction of effective behavior of composite materials with an inclusion fraction of $f_i > \approx 0.25$. The same result was obtained by Li and Hu [33] who compared the Mori–Tanaka scheme to the generalized Kuster–Toksoz model [36] for modelling of viscoelastic properties of highly-filled composite materials.

6. Summary and outlook

In this paper, classical homogenization schemes for upscaling of elastic properties in the framework of continuum micromechanics, i.e., Mori–Tanaka scheme, self-consistent scheme, and generalized self-consistent scheme, were extended towards upscaling of viscoelastic material properties. Hereby, the underlying linear viscoelastic boundary value problem is solved by the so-called Laplace–Carson transform method. The solution of the viscoelastic upscaling problem in the time domain is obtained by inverse transformation by means of the so-called Gaver–Steifert algorithm. In contrast to the explicit Mori–Tanaka and generalized self-consistent scheme, the solution for the self-consistent scheme has to be obtained in an iterative manner, with the necessity to monitor the consistency of the underlain and resulting viscoelastic behavior, respectively. Finally, the developed upscaling schemes were applied to a polyester matrix/marble dust filler composite. Hereby, for the investigated range of volume fractions of inclusions, ranging from $f_i = 0.29$ to $f_i = 0.55$, and a matrix/inclusion-type morphology, the extended generalized self-consistent scheme emerged as the most suitable scheme for determination of the effective viscoelastic properties of this highly-filled composite material. Future work will be devoted to application of the developed upscaling framework to material systems with an even

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<th>Table 4</th>
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<tr>
<td>Effective material parameters $J_{\text{dev}}^{\text{eff}}$ and $\lambda_{\text{dev}}^{\text{eff}}$, obtained from upscaling employing the Mori–Tanaka scheme (MT), the self-consistent scheme (SCS), and the generalized self-consistent scheme (GSCS) for different values of the volume fraction of filler $f_i$ and investigated time range of $t = 10^2$ min to $10^6$ min.</td>
</tr>
<tr>
<td>MT</td>
</tr>
<tr>
<td>---</td>
</tr>
<tr>
<td>$J_{\text{dev}}^{\text{eff}}(f_i = 0)$ (GPa)</td>
</tr>
<tr>
<td>$J_{\text{dev}}^{\text{eff}}(f_i = 0.29)$ (GPa)</td>
</tr>
<tr>
<td>$J_{\text{dev}}^{\text{eff}}(f_i = 0.38)$ (GPa)</td>
</tr>
<tr>
<td>$\lambda_{\text{dev}}^{\text{eff}}(f_i = 0.45)$ (GPa)</td>
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<tr>
<td>$\lambda_{\text{dev}}^{\text{eff}}(f_i = 0.55)$ (GPa)</td>
</tr>
<tr>
<td>$\lambda_{\text{dev}}^{\text{eff}}(f_i = 0)$ (-)</td>
</tr>
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higher volume fraction of inclusions, resulting from well-designed grading curves for the inclusions as used in construction materials.

Acknowledgement

This research was carried out during the former research post of the first author at the Institute for Mechanics of Materials and Structures at Vienna University of Technology. Financial support by the Hochschuljubiläumstiftung der Stadt Wien (Grant No. H-1858/2008) during this post is gratefully acknowledged.

Appendix A. Elementary algebraic manipulations for derivation of Mori–Tanaka and self-consistent scheme

Using
\[ \langle A(x) \rangle_{\mathbf{V}} = \frac{\int V \langle A(x) \rangle_{\mathbf{V}}}{\int V} = \mathbf{A} \]
\[ \Rightarrow f_0 \langle A(x) \rangle_{\mathbf{V}} = 0 - f_i A_i, \]
where \( \langle A(x) \rangle_{\mathbf{V}} = A_i = \text{const.} \) was used and \( f_i \) and \( f_0 \) denote the volume fractions of the inclusion and reference medium, respectively, and Eq. (3), one gets the volume average of the localization tensor over the reference medium as
\[ \langle A(x) \rangle_{\mathbf{V}} = \left\{ \sum_{i=0}^{\mathbf{c}_0^3} f_i [1 + S_i_i : (c_0^1 : c_i - 1)] \right\}^{-1}. \]
(36)
Hereby,
\[ f_0 \langle A(x) \rangle_{\mathbf{V}} = \mathbf{e} - f_i \mathbf{e} \]
\[ = \langle \mathbf{e} \rangle : \langle \mathbf{e} \rangle^{-1} \]
\[ = \langle \mathbf{e} \rangle - f_i \langle \mathbf{e} \rangle^{-1} \]
\[ = \left\{ \frac{f_0}{f_0 + S_i_i : (c_0^1 : c_i - 1) - 1} + f_i \mathbf{e} \right\}^{-1} \]
\[ = \langle \mathbf{e} \rangle^{-1} \]
(37)
with the abbreviations
\[ \langle \mathbf{e} \rangle = \left\{ \sum_{i=0}^{\mathbf{c}_0^3} f_i [1 + S_i_i : (c_0^1 : c_i - 1)] \right\}^{-1} \] and \( [\mathbf{c}_0^1] = [1 + S_i_i : (c_0^1 : c_i - 1)] \) was employed. Considering Eqs. (36) and (3) in Eq. (7) gives access to the effective material tensor (see Eq. (8)).

Appendix B. Generalized self-consistent scheme for determination of effective elastic properties of a 2-phase composite material according to Christensen and Lo [9,10]

The effective deviatoric elastic compliance \( f_{\text{dev}}^{\text{eff}} = 1/\mu_{\text{eff}} \) of a matrix/inclusion-type composite (indices “m” and “i”, respectively) is obtained as the solution of the quadratic equation [9]
The effective volumetric elastic compliance for the generalized self-consistent scheme is given as [25] (see also Benveniste [3]):

\[
\begin{align*}
J_{\text{eff}}^{vol} &= \frac{1}{k_{\text{eff}}} f_m \left( \frac{4(J_{\text{m}} + 3k_3)}{f_m k_3 (4J_{\text{m}} + 3k_3) + f_i k_i (4J_{\text{i}} + 3k_3)} \right) \\
&= \frac{f_m}{J_{\text{m}}} \frac{4K_{\text{m}}^3 + 3\alpha K_{\text{m}}}{4K_{\text{m}}^3 + 3\alpha K_{\text{m}} + f_i K_i} + \frac{f_i}{J_{\text{i}}} \frac{4K_{\text{i}}^3 + 3\beta K_{\text{i}}}{4K_{\text{i}}^3 + 3\beta K_{\text{i}} + f_k K_k},
\end{align*}
\]

(43)

which coincides with the effective volumetric elastic compliance of the Mori–Tanaka scheme (see Eq. (10.1)).

References