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A coated inclusion-based homogenization scheme for viscoelastic composites with interphases

Marc Schöneich[1,2,3], Florence Dinzart[3], Hafid Sabar[3], Stéphane Berbenni[2]*, Markus Stommel[1]

[1] TU Dortmund University, Mechanical Engineering, Chair of Plastics Technology, Leonhard-Euler-Str. 5, 44227 Dortmund, Germany

[2] Laboratoire d'Etude des Microstructures et de Mécanique des Matériaux, UMR CNRS 7239, Université de Lorraine, 57045 Metz, France

[3] Laboratoire de Mécanique Biomécanique Polymères Structures, Ecole Nationale Ingénieurs de Metz, Université de Lorraine, 57078 Metz, France

Abstract

A coated inclusion-based homogenization scheme is developed for three-phase viscoelastic composites in the Laplace-Carson domain. The interphase between inclusion and matrix is considered as a coating in a composite-like inclusion and shows altered viscoelastic behavior compared to the matrix. The strain concentration equations between the viscoelastic inclusion and the viscoelastic coating are derived with two different models: the double inclusion (denoted DI) model, and the reconsidered double inclusion (RDI) model. Then, the homogenization scheme is based on a modified Mori-Tanaka scheme for three-phase viscoelastic composites, which is validated with the exact analytical formulation in the case of spherical composite inclusions and isotropic behaviors for all constituents. The comparison of the proposed coated inclusion-based homogenization scheme based on the RDI model with the exact analytical solution shows a significant improvement compared to the one based on the DI model in the prediction of effective properties for composites with interphases. Finally, considering experimental dynamic mechanical analyses (DMA) in the frequency domain for a carbon-black filled styrene butadiene rubber from the literature, the effective viscoelastic behavior is estimated with a good accuracy in terms of the storage and loss moduli for different volume fractions of composite inclusions.

Keywords: Linear viscoelasticity; Homogenization; Coated Inclusion; Interphase; Polymer-Matrix Composites

* Corresponding author. Tel.: +33 (0)3 87 54 72 38
E-mail address: stephane.berbenni@univ-lorraine.fr

1 Introduction

The realistic prediction of the mechanical behavior in multi-phase composites by micromechanical approaches is essential for a holistic understanding of the fundamental underlying mechanical properties and expedient FE simulations. In literature, it has been discussed that composites consisting of a stiff inclusion phase embedded in a soft polymer matrix show the presence of a third phase (Kim et al., 2001). This interphase is defined as the region between the matrix and an inclusion and shows modified material properties with respect to the matrix phase (Schöneich et al., 2015). Still, it has to be considered that the interphase is linked to the viscous behavior of the polymer matrix material (Thomason and van Rooyen, 1990).

In the last decades, the elastic behavior of composites containing three or more phases (with different material properties) was studied with various micromechanical models. The analytical micromechanical approach of multi-coated inclusions was first given by Hervé and Zaoui (Hervé and Zaoui, 1993; Hervé and Zaoui, 1995). Starting from the "Composite Sphere Assemblage" model from Hashin (1962) and the effective moduli derived by Christensen and Lo's Generalized Self-Consistent scheme (Christensen and Lo, 1979), Hervé and Zaoui (Hervé and Zaoui, 1993) described multi coated spherical inclusions embedded in a matrix phase, which is surrounded by the homogeneous equivalent medium. The formulated exact solutions for the bulk and the shear moduli of the composite were obtained in case of isotropic materials and were restricted to spherical inclusions (Hervé and Zaoui, 1993) and to cylindrical inclusions (Hervé and Zaoui, 1995). Besides analytical solutions, the mean-field homogenization approach based on the Mori-Tanaka model (Mori and Tanaka, 1973) is frequently applied to calculate the effective moduli of the composite. For example, Benveniste et al. (Benveniste et al., 1989) started to predict the stress fields due to various boundary conditions applied on coated ellipsoidal fibers embedded in a matrix phase. Thereby, a modified Mori-Tanaka scheme was applied and the coating was considered as a separate phase with assigned mechanical properties. The double-inclusion model (DI model) introduced by Hori and Nemat-Nasser (Hori and Nemat-Nasser, 1993) is based on a coated inclusion of arbitrary ellipsoidal shape which is embedded in a homogeneous anisotropic reference medium. In case of remote homogeneous stresses or strains, the average fields in the inclusion and the coating can be analytically estimated. The DI model and its further enhancement to the multi-coated inclusion model (MI model) predict the same effective composite behavior as the Mori-Tanaka approach (Nemat-Nasser and Hori, 1993).

Furthermore, some connections of the DI model with the Mori-Tanaka's approach (Benveniste, 1987; Mori and Tanaka, 1973; Weng, 1984, 1990) and with the Ponte Castañeda-Willis approach (Ponte Castañeda and Willis, 1995) were provided by Hu and Weng (2000).

Micromechanical approaches considering interphases were also solved with the so-called "Hill's interfacial operators" (Hill, 1983; Laws, 1975; Walpole, 1978) together with the Green's function technique. These solutions were presented in various studies (Cherkaoui et al., 1994, 1995) and were later extended to multi-coated inclusion-reinforced elastic composites by Lipinski et al. (2006) and to multi-coated inclusion-reinforced elastic composites with eigenstrains like thermal strains by Berbenni and Cherkaoui (2010). Aboutajeddine and Neale (Aboutajeddine and Neale, 2005) also proposed a reformulated DI model in two steps. First, these authors considered a composite inclusion constituted of the inclusion and the coating embedded in an infinite reference medium. In the first step, the strain concentration problem simplifies to a classic Eshelby problem, which is exactly solved using the Eshelby tensor (Eshelby, 1957) of the composite inclusion. Second, the mechanical interaction between the inclusion and the coating, which is difficult to estimate, is simplified by considering the inclusion embedded in an infinite reference medium with the mechanical properties of the coating (Aboutajeddine and Neale, 2005). However, this promising extension of the original double-inclusion model was only performed for two-phase elastic composites. Recently, Dinzart et al. (2016) extended the model of Aboutajeddine and Neale (2005) for the development of a "n+1 phase" Self-Consistent model of multi-phase elastic composites based on a revisited formulation of the multi-coated inclusion problem. It was shown that the effective bulk moduli correspond exactly to the analytical results. This last approach appears to be an improvement of the MI model initiated by Hori and Nemat-Nasser (Nemat-Nasser and Hori, 1993) for different contrasts, volume fractions and aspect ratios of the coated inclusions.

All aforementioned approaches do not include the viscoelastic properties of the interphase coming from the modified matrix phase in the case of a linear viscoelastic generalized Maxwell model (Hashin, 1991; Diani et al., 2013; Diani and Gilormini, 2014). Compared to the diversity of existing multi-phase approaches, the viscoelastic behavior of three-phase composite materials containing an interphase was rarely investigated. Hashin (1991) described the influence of a viscoelastic interphase in a unidirectional fiber composite containing continuous cylindrical fibers. Thus, the composite exhibits transversal isotropic

effective behavior. Hashin worked out, that the viscoelastic properties of the interphase mainly influence the shear loading cases. Li and Weng (1996) studied the influence of viscoelastic interphases on the creep and stress-strain behavior of fiber-reinforced polymer matrix composites using the "Composite Cylinder Assemblage" model of Hashin and Rosen (1964). They showed that the effective creep and stress-strain behavior were stringly dependent on interphase. Recently, Schöneich et al. (2015) developed a promising experimental method for the characterization of the interphase in short glass fiber composites with high mechanical contrast between the matrix and the fibers. A summary of the interphase thickness observed by different experimental methods was given by Fisher and Brinson (Fisher and Brinson, 2001). In their work, Fisher and Brinson extended the aforementioned approach of Benveniste et al. (1989) to study the viscoelastic interphase properties in composites with unidirectional cylindrical fibers. Moreover, a comparison with the original Mori-Tanaka method was performed, where no significant difference between the proposed solutions could be found. However, supplementary two-dimensional finite element results pointed out, that the interphase should be taken into account and deserved further investigations. Additional work concerning particle-reinforced composites with the viscoelastic interphase was performed by Wei und Huang (Wei and Huang, 2004). They investigated the relaxation and damping characteristics of an elastic brittle composite with a viscoelastic interphase. Friebel et al. (2006) reported a two-step homogenization procedure where coated inclusions as described by the Hori and Nemat-Nasser's DI approach (Hori and Nemat-Nasser, 1999) were firstly homogenized, and second, the DI approach or a classic Mori-Tanaka scheme were applied to obtain the effective behavior of the composite. This two-level approach was seen to be consistent with the approach by Aboutajeddine and Neale (2005) in the particular case of coated homothetic inclusions.

Within the described framework of multi-phase mean-field homogenization approaches considering viscoelastic interphases, this work adopts the advantages of a two-level scheme leading to better predictions than the classic DI model. Therefore, the reconsidered double-inclusion scheme by Aboutajeddine and Neale (Aboutajeddine and Neale, 2005) is extended to study the effective linear viscoelastic behavior of three-phase composites. The considered composite is described by general ellipsoidal inclusions with anisotropic behavior, surrounded by viscoelastic interphases forming coated inclusions embedded in a viscoelastic matrix. Furthermore, an extended Mori-Tanaka scheme for three-phase composites with coated inclusions is considered as homogenization scheme. In the following section 2, the strain

concentration equations between the viscoelastic inclusion and the viscoelastic coating are derived with two different approximations: the double inclusion (denoted DI) model, and the reconsidered double inclusion (RDI) model. These strain concentration equations are incorporated in a homogenization scheme based on a modified Mori-Tanaka scheme for three-phase viscoelastic composites. In section 3, the proposed coated inclusion-based homogenization schemes based on the DI and RDI models are discussed in comparison with the exact solution derived by Hervé and Zaoui (Hervé and Zaoui, 1990) for both spherical inclusions and coated inclusions. Finally, considering experimental DMA data in the frequency domain for a carbon-black filled styrene butadiene rubber from Diani et al. (2013), the effective viscoelastic behavior is estimated with a good accuracy with the RDI model in terms of the storage and loss moduli for different volume fractions of composite coated inclusions embedded in the matrix phase (unfilled rubber).

2 Micromechanical model

2.1 Constitutive equations

For the development of a micromechanical model including linear viscoelasticity, the constitutive equation for linear viscoelastic materials, also denoted as Stieltjes integral equation is recalled in Eq. (1) (Christensen, 1969). For brevity, the tensorial notation is used for the subsequently stated equations. In the following, the appearing tensors are denoted in bold type. With the Boltzmann principle for non-ageing linear viscoelastic behavior, the following local constitutive law is introduced:

$$\boldsymbol{\sigma}(\boldsymbol{r},t) = \int_{-\infty}^{t} \boldsymbol{C}(\boldsymbol{r},t-t'):\frac{d\boldsymbol{\varepsilon}(\boldsymbol{r},t')}{dt'}dt' \tag{1}$$

Thereby, $\boldsymbol{\sigma}$ and $\boldsymbol{\varepsilon}$ are the stress and strain second order tensors, $\boldsymbol{C}$ is the viscoelastic relaxation fourth order tensor. Based on various studies of viscoelastic material modeling (Hashin, 1965; Mandel, 1966), the Laplace-Carson (LC) transformation utilizing the correspondence principle is introduced. The general form of the transformation is given by the following expression, where p represents the complex variable:

$$\mathcal{L}C\{f\}(p) = \hat{f}(p) = p\int_{0}^{\infty} f(t)e^{-pt}dt \tag{2}$$

This transformation is applied to Eq. (1) leading to the symbolic linear elastic constitutive equation:

$$\hat{\boldsymbol{\sigma}}(\boldsymbol{r},p) = \hat{\boldsymbol{C}}(\boldsymbol{r},p):\hat{\boldsymbol{\varepsilon}}(\boldsymbol{r},p) \tag{3}$$

Thus, the subsequently developed equations are presented in the transformed domain. Thereby, the notation $\hat{f}$ represents the LC transformation of f as illustrated in Eq. (3).

Considering a viscoelastic heterogeneous medium V with the boundary ∂V, the field equations in the transformed LC domain are elaborated. The stress equilibrium condition for the stress tensor $\hat{\boldsymbol{\sigma}}$ without body forces or inertia effects is defined as:

$$\boldsymbol{div}\hat{\boldsymbol{\sigma}} = 0 \tag{4}$$

The strain field $\hat{\boldsymbol{\varepsilon}}$ is obtained from the symmetric part of the displacement gradient:

$$\hat{\boldsymbol{\varepsilon}} = \frac{1}{2}(\boldsymbol{\nabla}\hat{\boldsymbol{u}} + (\boldsymbol{\nabla}\hat{\boldsymbol{u}})^{\boldsymbol{t}}) \tag{5}$$

where $\hat{\boldsymbol{u}}$ represents the displacement field in the transformed domain. As boundary conditions, a uniform strain field $\hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}}$ is imposed on ∂V:

$$\hat{\boldsymbol{u}} = \hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}} \cdot \boldsymbol{r} \text{ on } \partial\mathrm{V} \tag{6}$$

From Eqs. (3), (4) and (5) and using the major symmetry of $\hat{\boldsymbol{C}}$ ($\hat{C}_{ijkl} = \hat{C}_{ijlk} = \hat{C}_{jikl} = \hat{C}_{klij}$), the following equilibrium equation is obtained:

$$\boldsymbol{div}\left(\hat{\boldsymbol{C}}: \boldsymbol{\nabla}\hat{\boldsymbol{u}}\right) = 0 \tag{7}$$

2.2 Linear viscoelastic Lippmann-Schwinger equation

With the definition of the reference homogeneous viscoelastic moduli $\hat{\boldsymbol{C}}^{\boldsymbol{0}}$ of the infinite reference medium, the local viscoelastic relaxation tensor $\hat{\boldsymbol{C}}$ is expressed as a function of spatial fluctuations $\boldsymbol{\delta}\hat{\boldsymbol{C}}$ and $\hat{\boldsymbol{C}}^{\boldsymbol{0}}$:

$$\hat{\boldsymbol{C}}(\boldsymbol{r}) = \hat{\boldsymbol{C}}^{\boldsymbol{0}} + \boldsymbol{\delta}\hat{\boldsymbol{C}}(\boldsymbol{r}) \tag{8}$$

Eqs. (7) and (8) lead to the following Navier-type equation:

$$\boldsymbol{div}\left[\left(\hat{\boldsymbol{C}}^{\boldsymbol{0}}: \boldsymbol{\nabla}\hat{\boldsymbol{u}}\right) + \left(\boldsymbol{\delta}\hat{\boldsymbol{C}}: \boldsymbol{\nabla}\hat{\boldsymbol{u}}\right)\right] = 0 \tag{9}$$

Introducing the linear viscoelastic Green tensor $\widehat{\boldsymbol{G}}^{\boldsymbol{0}}$ associated with the homogeneous reference medium (HRM) with viscoelastic moduli $\widehat{\boldsymbol{C}}^{\boldsymbol{0}}$ defined by:

$$\hat{C}^0_{ijkl}\,\hat{G}^0_{km,lj}(\boldsymbol{r}-\boldsymbol{r}') + \delta_{im}\delta(\boldsymbol{r}-\boldsymbol{r}') = 0 \tag{10}$$

with $\hat{G}^0_{km}(\boldsymbol{r}-\boldsymbol{r}') = 0$ on ∂V, $\delta(\boldsymbol{r}-\boldsymbol{r}')$ represents the three-dimensional Dirac delta function and δ_{im} is the Kronecker delta.

From Eqs. (9) and (10), the integral equation of the Lippmann-Schwinger type in the transformed domain can be classically derived as in (Dederichs and Zeller, 1973; Kröner, 1977):

$$\hat{\boldsymbol{\varepsilon}}(\boldsymbol{r}) = \hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}} - \int_V \hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}(\boldsymbol{r}-\boldsymbol{r}') : \boldsymbol{\delta}\widehat{\boldsymbol{C}}(\boldsymbol{r}') : \hat{\boldsymbol{\varepsilon}}(\boldsymbol{r}')\, dV' \tag{11}$$

In Eq. (11), the fourth-order tensor $\hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}$ represents the modified Green tensor of the reference medium $\widehat{\boldsymbol{C}}^{\boldsymbol{0}}$ associated with the boundary conditions and is defined by (Kröner, 1977; Kröner, 1990:

$$\hat{\Gamma}^0_{ijkl}(\boldsymbol{r}-\boldsymbol{r}') = -\frac{1}{2}\left(\hat{G}^0_{ik,jl}(\boldsymbol{r}-\boldsymbol{r}') + \hat{G}^0_{jk,il}(\boldsymbol{r}-\boldsymbol{r}')\right) \tag{12}$$

In the next section, the integral equation (Eq. (11)) is used to estimate the average strains in a coated inclusion embedded in the infinite homogeneous reference medium.

2.3 Coated inclusion problem and strain concentration equations

The viscoelastic coated inclusion problem is described in Figure 1. The composite coated inclusion denoted "DI" with volume V^{DI} is constituted of an inclusion I with volume V^I and

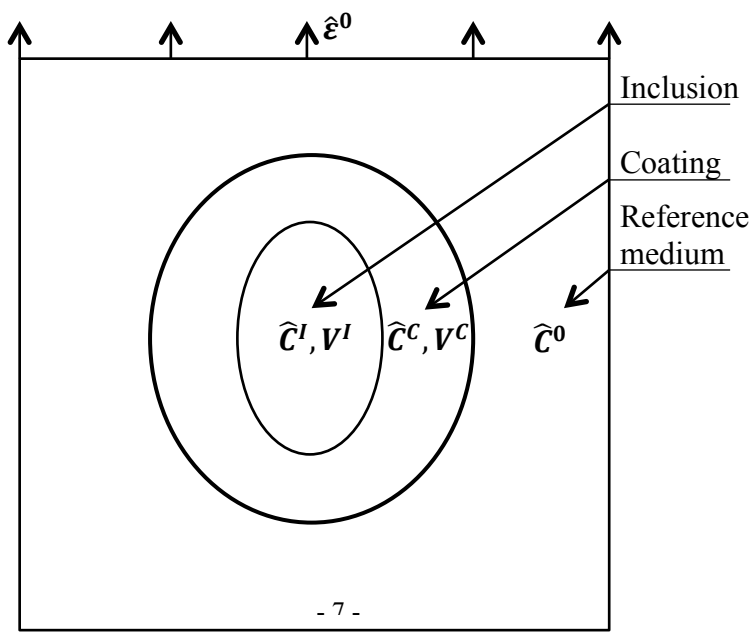


Figure 1: Composite coated inclusion embedded in a homogeneous reference medium (HRM)

symbolic elastic moduli $\widehat{\boldsymbol{C}}^{\boldsymbol{I}}$, and a coating C with volume V^C and symbolic elastic moduli $\widehat{\boldsymbol{C}}^{\boldsymbol{C}}$. The coated inclusion is embedded in an infinite homogeneous reference medium (HRM) of symbolic elastic moduli $\widehat{\boldsymbol{C}}^{\boldsymbol{0}}$ and subjected to a homogeneous symbolic strain at infinity $\hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}}$. Using Figure 1, the spatial fluctuation of symbolic elastic moduli $\boldsymbol{\delta}\widehat{\boldsymbol{C}}$ defined in Eq. (8) reads:

$$\boldsymbol{\delta}\widehat{\boldsymbol{C}}(\boldsymbol{r}) = \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I}/\boldsymbol{0}}\,\phi^I(\boldsymbol{r}) + \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{C}/\boldsymbol{0}}\,\phi^C(\boldsymbol{r}) \tag{13}$$

where $\boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I}/\boldsymbol{0}} = \widehat{\boldsymbol{C}}^{\boldsymbol{I}} - \widehat{\boldsymbol{C}}^{\boldsymbol{0}}$ and $\boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{C}/\boldsymbol{0}} = \widehat{\boldsymbol{C}}^{\boldsymbol{C}} - \widehat{\boldsymbol{C}}^{\boldsymbol{0}}$. The characteristic function $\phi^i(\boldsymbol{r})$ with $i = I, C$ is defined as:

$$\phi^i(\boldsymbol{r}) = \begin{cases} 1, & r \in V^i \\ 0, & r \notin V^i \end{cases} \tag{14}$$

Using Eq. (13) together with Eq. (11) leads to:

$$\begin{aligned} \hat{\boldsymbol{\varepsilon}}(\boldsymbol{r}) = \hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}} &- \int_{V^I} \hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}(\boldsymbol{r}-\boldsymbol{r}') : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I}/\boldsymbol{0}} : \hat{\boldsymbol{\varepsilon}}(\boldsymbol{r}')dV' \\ &- \int_{V^C} \hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}(\boldsymbol{r}-\boldsymbol{r}') : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{C}/\boldsymbol{0}} : \hat{\boldsymbol{\varepsilon}}(\boldsymbol{r}')dV' \end{aligned} \tag{15}$$

In order to determine the average strains over the inclusion, the coating and the coated inclusion, the overbar sign is introduced to denote the average value of a given mechanical field. Therefore, the average of $\hat{\boldsymbol{\varepsilon}}^{\boldsymbol{\alpha}}$ over the volume V^α is defined as:

$$\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{\alpha}} = \frac{1}{V^\alpha}\int_{V^\alpha} \hat{\boldsymbol{\varepsilon}}(\boldsymbol{r})\, dV^\alpha \tag{16}$$

where $\alpha = I, C, DI$ in the following. Using Eq. (16) together with Eq. (15), the average strain in the double inclusion $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{DI}}$ is obtained after simple manipulations:

$$\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{DI}} = \hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}} - \frac{V^I}{V^{DI}}\widehat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I}/\boldsymbol{0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} - \frac{V^C}{V^{DI}}\widehat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{C}/\boldsymbol{0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}} \tag{17}$$

where the fourth order tensor $\widehat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}}(\widehat{\boldsymbol{C}}^{\boldsymbol{0}}) = \int_{V^{DI}} \hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}(\boldsymbol{r}-\boldsymbol{r}')dV$ is uniform if $\boldsymbol{r} \in V^{DI}$and $\boldsymbol{r}' \in V^I, V^C \subset V^{DI}$ for ellipsoidal-shaped coated inclusion DI with volume V^{DI} (Eshelby, 1957). Thereby, the tensor $\widehat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}}$ depends on the viscoelastic modulus $\widehat{\boldsymbol{C}}^{\boldsymbol{0}}$ of the HRM medium

as well as the outer ellipsoidal geometry of the inclusion with volume V^{DI} (Mura, 1987; Nemat-Nasser and Hori, 1993).

Furthermore, $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{DI}}$ is a function of the average strain over the inclusion $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}}$ and the average strain over the coating $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}}$ from simple volume averaging as follows:

$$\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{DI}} = \frac{V^I}{V^{DI}}\,\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} + \frac{V^C}{V^{DI}}\,\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}} \tag{18}$$

Second, the average symbolic strain over the inclusion V^I denoted $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}}$ can be obtained from the application of Eqs. (15) and (16) and after a straightforward simplification using the Eshelby (1957)'s property for ellipsoidal-shaped inclusion I as follows :

$$\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} = \hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}} - \hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}} : \boldsymbol{\Delta}\hat{\boldsymbol{C}}^{\boldsymbol{I/0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} - \frac{1}{V^I}\int_{V^I}\int_{V^C} \hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}(\boldsymbol{r}-\boldsymbol{r}') : \boldsymbol{\Delta}\hat{\boldsymbol{C}}^{\boldsymbol{C/0}} : \hat{\boldsymbol{\varepsilon}}(\boldsymbol{r}')dV'dV \tag{19}$$

where the fourth order tensor $\hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}}(\hat{\boldsymbol{C}}^{\boldsymbol{0}}) = \int_{V^I} \hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}(\boldsymbol{r}-\boldsymbol{r}')dV'$ is uniform if $\boldsymbol{r} \in V^I$and $\boldsymbol{r}' \in V^I$ for ellipsoidal-shaped inclusion with volume V^I (Eshelby, 1957). Let us recall that the tensor $\hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{\alpha}}$ $(\alpha = I, DI)$ is linked to the Eshelby tensor $\hat{\boldsymbol{S}}^{\boldsymbol{\alpha}}$ by the relation: $\hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{\alpha}} = \hat{\boldsymbol{S}}^{\boldsymbol{\alpha}} : (\hat{\boldsymbol{C}}^{\boldsymbol{0}})^{-\boldsymbol{1}}$ in the transformed domain.

Eqs. (17), (18) and (19) form a set of three exact equations for both ellipsoidal inclusion and coated inclusion to derive the average symbolic strains $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}}$ and $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}}$ in the coating and in the inclusion, respectively. Therefore, the symbolic strain concentration tensors $\hat{\boldsymbol{A}}^{\boldsymbol{IC}}$ and $\hat{\boldsymbol{A}}^{\boldsymbol{I0}}$ can be determined as follows:

$$\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} = \hat{\boldsymbol{A}}^{\boldsymbol{IC}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}} \tag{20}$$

$$\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} = \hat{\boldsymbol{A}}^{\boldsymbol{I0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{0}}$$

First, it is straightforward to find the relationship between $\hat{\boldsymbol{A}}^{\boldsymbol{I0}}$ and $\hat{\boldsymbol{A}}^{\boldsymbol{IC}}$ using Eq. (17) together with Eq. (18) and eliminating $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{DI}}$:

$$\hat{\boldsymbol{A}}^{\boldsymbol{I0}} = \left[\frac{V^I}{V^{DI}}\left(\boldsymbol{I} + \hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}} : \boldsymbol{\Delta}\hat{\boldsymbol{C}}^{I/0}\right) + \frac{V^C}{V^{DI}}\left(\boldsymbol{I} + \hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}} : \boldsymbol{\Delta}\hat{\boldsymbol{C}}^{C/0}\right) : \left(\hat{\boldsymbol{A}}^{\boldsymbol{IC}}\right)^{-\boldsymbol{1}}\right]^{-\boldsymbol{1}} \tag{21}$$

However, the difficulty to determine $\hat{\boldsymbol{A}}^{\boldsymbol{IC}}$ (and then $\hat{\boldsymbol{A}}^{\boldsymbol{I0}}$) lies in an accurate enough estimate of the double volume integral term in Eq. (19). Therefore, two different models are developed in the following to derive these strain concentration tensors.

1. Double-inclusion (DI) model:

The average strain relation in the inclusion I can be estimated with the original approach introduced by Hori and Nemat-Nasser (1993). Following this approach, the second double volume integral term of Eq. (19) is decomposed in two parts. This leads to the following equation for $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}}$:

$$\begin{aligned}\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} = \hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}} - \hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}} : \Delta\hat{\boldsymbol{C}}^{\boldsymbol{I/0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} - \frac{1}{V^I}\int_{V^I}\int_{V^C} \hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}(\boldsymbol{r}-\boldsymbol{r}') : \Delta\hat{\boldsymbol{C}}^{\boldsymbol{C/0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}} dV' dV \\ - \frac{1}{V^I}\int_{V^I}\int_{V^C} \hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}(\boldsymbol{r}-\boldsymbol{r}') : \Delta\hat{\boldsymbol{C}}^{\boldsymbol{C/0}} : \left(\hat{\boldsymbol{\varepsilon}}(\boldsymbol{r}') - \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}}\right) dV' dV\end{aligned} \tag{22}$$

As it is approximated by Hori and Nemat-Nasser (Hori and Nemat-Nasser, 1993), the last term of Eq. (22) involving $\left(\hat{\boldsymbol{\varepsilon}}(\boldsymbol{r}') - \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}}\right)$ if $\boldsymbol{r}' \in V^C$ can be omitted. In the present case of a composite inclusion containing the inclusion and the coating, the integral expression over V^{DI} is described by the sum of integrals over V^C and V^I. Subsequently, the previous Eq. (22) is rewritten as:

$$\begin{aligned}\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} = \hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}} - \hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}} : \Delta\hat{\boldsymbol{C}}^{\boldsymbol{I/0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} \\ - \frac{1}{V^I}\int_{V^I}\int_{V^{DI}} \hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}(\boldsymbol{r}-\boldsymbol{r}') : \Delta\hat{\boldsymbol{C}}^{\boldsymbol{C/0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}} dV' dV + \frac{1}{V^I}\int_{V^I}\int_{V^I} \hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}(\boldsymbol{r}-\boldsymbol{r}') : \Delta\hat{\boldsymbol{C}}^{\boldsymbol{C/0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}} dV' dV\end{aligned} \tag{23}$$

Using the Eshelby (1957)'s result for ellipsoidal inclusion and coated inclusion, the first double volume integral term in Eq. (23) is simplified with the uniform tensor $\hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}}$ because $r \in V^I \subset V^{DI}$, $r' \in V^{DI}$ and the second double volume integral term in Eq. (23) is simplified with the uniform tensor $\hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}}$ because r and $r' \in V^I$. Introducing the uniform tensor $\Delta\hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}} = \hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}} - \hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}}$, Eq. (23) is further recast as:

$$\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} = \hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}} - \hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}} : \Delta\hat{\boldsymbol{C}}^{\boldsymbol{I/0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} - \Delta\hat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}} : \Delta\hat{\boldsymbol{C}}^{\boldsymbol{C/0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}} \tag{24}$$

Then, the strain concentration tensor $\widehat{\boldsymbol{A}}^{\boldsymbol{IC}}$ is obtained by eliminating $\hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}}$ in Eq. (24) together with Eqs. (17) and (18) to find the relationship between $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}}$ and $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}}$. After a few algebraic manipulations, the strain concentration tensor for the DI model denoted $\widehat{\boldsymbol{A}}^{\boldsymbol{CI,1}}$yields:

$$\widehat{\boldsymbol{A}}^{\boldsymbol{IC,1}} = \left[\boldsymbol{I} + \widehat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I/0}} - \frac{V^I}{V^{DI}}\left(\boldsymbol{I} + \widehat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I/0}}\right)\right]^{-1} : \left[\frac{V^C}{V^{DI}}\left(\boldsymbol{I} + \widehat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{C/0}}\right) - \Delta\widehat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{C/0}}\right] \tag{25}$$

Therefore, the strain concentration tensor $\widehat{\boldsymbol{A}}^{\boldsymbol{I0}}$ for the DI model, denoted $\widehat{\boldsymbol{A}}^{\boldsymbol{I0,1}}$, follows from Eq. (21) together with Eq. (25).

2. Reconsidered double-inclusion (RDI) model:

Starting from the average symbolic strain in the inclusion $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}}$ derived in Eq. (19), a reconsidered double-inclusion model (RDI) originally introduced by Aboutajeddine and Neale (2005) for two-phase elastic composites within a self-consistent scheme is now extended to the case of the viscoelastic coated inclusion problem.

First of all, the difference between Eq. (17) and Eq. (19) yields:

$$\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{DI}} - \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} = \left[\widehat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{I}} - \frac{V^I}{V^{DI}}\widehat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}}\right] : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I/0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} - \frac{V^C}{V^{DI}}\widehat{\boldsymbol{T}}_{\boldsymbol{0}}^{\boldsymbol{DI}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{C/0}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}} + \frac{1}{V^I}\int_{V^I}\int_{V^C}\hat{\boldsymbol{\Gamma}}^{\boldsymbol{0}}(\boldsymbol{r} - \boldsymbol{r}') : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{C/0}} : \hat{\boldsymbol{\varepsilon}}(\boldsymbol{r}')dV'dV \tag{26}$$

In order to eliminate the double volume integral term in Eq. (26), a natural choice as performed by Aboutajeddine and Neale (2005), is to set $\widehat{\boldsymbol{C}}^{\boldsymbol{0}} = \widehat{\boldsymbol{C}}^{\boldsymbol{C}}$. Thus, the coating C is chosen as the reference medium so that Eq. (26) is simplified to:

$$\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{DI}} - \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} = \left[\widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{I}} - \frac{V^I}{V^{DI}}\widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{DI}}\right] : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I/C}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} \tag{27}$$

Replacing $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{DI}}$ in Eq. (27) by its definition given in Eq. (18), the relationship between $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}}$ and $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}}$ is found and the strain concentration tensor $\widehat{\boldsymbol{A}}^{\boldsymbol{IC}}$ (see Eq. (20)) for the RDI model denoted $\widehat{\boldsymbol{A}}^{\boldsymbol{IC,2}}$yields:

$$\widehat{\boldsymbol{A}}^{\boldsymbol{IC,2}} = \left[\boldsymbol{I} + \widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{C}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I/C}}\right]^{-1} \tag{28}$$

where the tensor $\widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{C}}$ is defined as:

$$\widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{C}} = \widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{DI}} - \frac{V^{DI}}{V^{C}}\left(\widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{DI}} - \widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{I}}\right) \tag{29}$$

Furthermore, the strain concentration tensor $\widehat{\boldsymbol{A}}^{\boldsymbol{I}}$ for the RDI model, denoted $\widehat{\boldsymbol{A}}^{\boldsymbol{I,2}}$, follows from Eq. (21) together with Eqs. (28) and (29). In the next section, the derived strain concentration equations are incorporated in a homogenization scheme based on an extended Mori-Tanaka scheme for three-phase viscoelastic composites with coated inclusions.

2.4 Homogenization

The corresponding representative volume element (RVE) is constituted of coated inclusions embedded in a matrix phase *M*. Thereby, the phase volume fractions in the three-phase composite are defined as $f^{\alpha} = V^{\alpha}/V$ where $\alpha = I, C, M$ and V is the total volume of the RVE with $f^{M} + f^{I} + f^{C} = 1$. The homogenization procedure consists in the determination of the effective viscoelastic moduli tensor $\widehat{\boldsymbol{C}}^{\boldsymbol{eff}}$, which relies the macroscopic symbolic stress $\widehat{\boldsymbol{\Sigma}}$ to the macroscopic applied symbolic strain $\widehat{\mathbf{E}}$:

$$\widehat{\boldsymbol{\Sigma}} = \widehat{\boldsymbol{C}}^{\boldsymbol{eff}} : \widehat{\mathbf{E}} \tag{30}$$

The strain concentration tensors link the average strains of the inclusions, coatings and matrix to the macroscopic applied strain by:

$$\overline{\widehat{\boldsymbol{\varepsilon}}}^{\boldsymbol{\alpha}} = \widehat{\boldsymbol{A}}^{\boldsymbol{\alpha}} : \widehat{\mathbf{E}} \ \ with \ \ \alpha = I, C, M \tag{31}$$

Using Eq. (31) and the constitutive equation (Eq. (3)) for each phase yields:

$$\overline{\widehat{\boldsymbol{\sigma}}}^{\boldsymbol{\alpha}} = \widehat{\boldsymbol{C}}^{\boldsymbol{\alpha}} : \overline{\widehat{\boldsymbol{\varepsilon}}}^{\boldsymbol{\alpha}} = \widehat{\boldsymbol{C}}^{\boldsymbol{\alpha}} : \widehat{\boldsymbol{A}}^{\boldsymbol{\alpha}} : \widehat{\mathbf{E}} \ \ with \ \ \alpha = I, C, M \tag{32}$$

The overall symbolic stress and strain tensors are respectively given by:

$$\widehat{\boldsymbol{\Sigma}} = f^{M}\overline{\widehat{\boldsymbol{\sigma}}}^{\boldsymbol{M}} + f^{I}\overline{\widehat{\boldsymbol{\sigma}}}^{\boldsymbol{I}} + f^{C}\overline{\widehat{\boldsymbol{\sigma}}}^{\boldsymbol{C}} \tag{33}$$

$$\widehat{\mathbf{E}} = f^{M}\overline{\widehat{\boldsymbol{\varepsilon}}}^{\boldsymbol{M}} + f^{I}\overline{\widehat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} + f^{C}\overline{\widehat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}} \tag{34}$$

Using the definition of volume fractions and Eqs. (31) and (34) lead to:

$$f^M \widehat{\boldsymbol{A}}^{\boldsymbol{M}} = \boldsymbol{I} - f^C \widehat{\boldsymbol{A}}^{\boldsymbol{C}} - f^I \widehat{\boldsymbol{A}}^{\boldsymbol{I}} \tag{35}$$

Finally, the expression of the symbolic effective viscoelastic moduli tensor $\widehat{\boldsymbol{C}}^{\boldsymbol{eff}}$ is obtained from Eqs. (30-35) as follows:

$$\widehat{\boldsymbol{C}}^{\boldsymbol{eff}} = \widehat{\boldsymbol{C}}^{\boldsymbol{M}} + f^I(\widehat{\boldsymbol{C}}^{\boldsymbol{I}} - \widehat{\boldsymbol{C}}^{\boldsymbol{M}}) : \widehat{\boldsymbol{A}}^{\boldsymbol{I}} + f^C(\widehat{\boldsymbol{C}}^{\boldsymbol{C}} - \widehat{\boldsymbol{C}}^{\boldsymbol{M}}) : \widehat{\boldsymbol{A}}^{\boldsymbol{C}} \tag{36}$$

Thus, the evaluation of the involved strain concentration tensors $\widehat{\boldsymbol{A}}^{\boldsymbol{I}}$ and $\widehat{\boldsymbol{A}}^{\boldsymbol{C}}$ represents the basis for the description of the effective properties in case of the two different coated inclusion homogenization schemes based on the DI or RDI models derived in section 2.3. Here, an extended Mori-Tanaka scheme for three-phase viscoelastic composites is adopted considering a low concentration of coated inclusions in the composite. Therefore, the average symbolic strain in the matrix $\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{M}}$ is equal to the reference symbolic strain $\hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}}$. Thus, the matrix phase M is chosen as the reference medium and $\hat{\boldsymbol{\varepsilon}}^{\boldsymbol{0}} = \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{M}}$ and $\widehat{\boldsymbol{C}}^{\boldsymbol{0}} = \widehat{\boldsymbol{C}}^{\boldsymbol{M}}$ are replaced in Eqs. (20) and (21). These equations are reconsidered in the context of the RVE with the introduction of the volume fractions f^I, f^C, $f^{DI} = 1 - f^M$. Thus, Eq. (20) leads to:

$$\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{C}} = \widehat{\boldsymbol{A}}^{\boldsymbol{C}} : \hat{\mathbf{E}} = \left(\widehat{\boldsymbol{A}}^{\boldsymbol{IC}}\right)^{-\mathbf{1}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} \tag{37}$$

$$\bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{I}} = \widehat{\boldsymbol{A}}^{\boldsymbol{I}} : \hat{\mathbf{E}} = \widehat{\boldsymbol{A}}^{\boldsymbol{IM}} : \bar{\hat{\boldsymbol{\varepsilon}}}^{\boldsymbol{M}}$$

where from Eq. (21):

$$\widehat{\boldsymbol{A}}^{\boldsymbol{IM}} = \left[\frac{f^I}{1-f^M}\left(\boldsymbol{I} + \widehat{\boldsymbol{T}}_{\mathbf{0}}^{\boldsymbol{DI}} : \Delta\widehat{\boldsymbol{C}}^{\boldsymbol{I/0}}\right) + \frac{f^C}{1-f^M}\left(\boldsymbol{I} + \widehat{\boldsymbol{T}}_{\mathbf{0}}^{\boldsymbol{DI}} : \Delta\widehat{\boldsymbol{C}}^{\boldsymbol{C/0}}\right) : \left(\widehat{\boldsymbol{A}}^{\boldsymbol{IC}}\right)^{-\mathbf{1}}\right]^{-\mathbf{1}} \tag{38}$$

The symbolic strain concentration tensor $\widehat{\boldsymbol{A}}^{\boldsymbol{I}}$ for the inclusion phase as defined in Eq. (31) is obtained from Eqs. (35), (37) and (38) and yields:

$$\widehat{\boldsymbol{A}}^{\boldsymbol{I,1,2}} = \left[\frac{f^I}{1-f^M}\left(\boldsymbol{I} + f^M\widehat{\boldsymbol{T}}_{\boldsymbol{M}}^{\boldsymbol{DI}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I/M}}\right) + \frac{f^C}{1-f^M}\left(\boldsymbol{I} + f^M\widehat{\boldsymbol{T}}_{\boldsymbol{M}}^{\boldsymbol{DI}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{C/M}}\right) : \left(\widehat{\boldsymbol{A}}^{\boldsymbol{IC,1,2}}\right)^{-\boldsymbol{1}}\right]^{-1} \quad (39)$$

where the strain concentration tensors $\widehat{\boldsymbol{A}}^{\boldsymbol{IC,1,2}}$ were derived in section 2.3 for both DI and RDI models, respectively. Here, both $\widehat{\boldsymbol{A}}^{\boldsymbol{IC,1,2}}$ are rewritten in the framework of the extended Mori-Tanaka homogenization scheme by introducing the matrix phase as reference medium and phase volume fractions. In the case of the DI model, from Eq. (25), $\widehat{\boldsymbol{A}}^{\boldsymbol{IC,1}}$ yields:

$$\widehat{\boldsymbol{A}}^{\boldsymbol{IC,1}} = \left[\boldsymbol{I} + \widehat{\boldsymbol{T}}_{\boldsymbol{M}}^{\boldsymbol{I}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I/M}} - \frac{f^I}{1-f^M}\left(\boldsymbol{I} + \widehat{\boldsymbol{T}}_{\boldsymbol{M}}^{\boldsymbol{DI}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I/M}}\right)\right]^{-1} \quad (40)$$

$$: \left[\frac{f^C}{1-f^M}\left(\boldsymbol{I} + \widehat{\boldsymbol{T}}_{\boldsymbol{M}}^{\boldsymbol{DI}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{C/M}}\right) - \Delta\widehat{\boldsymbol{T}}_{\boldsymbol{M}}^{\boldsymbol{I}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{C/M}}\right]$$

and in the case of the RDI model, from Eqs. (28), (29), $\widehat{\boldsymbol{A}}^{\boldsymbol{IC,2}}$ yields:

$$\widehat{\boldsymbol{A}}^{\boldsymbol{IC,2}} = \left[\boldsymbol{I} + \widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{C}} : \boldsymbol{\Delta}\widehat{\boldsymbol{C}}^{\boldsymbol{I/C}}\right]^{-\boldsymbol{1}} \quad (19)$$

where:

$$\widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{C}} = \widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{DI}} - \frac{1-f^M}{f^C}\left(\widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{DI}} - \widehat{\boldsymbol{T}}_{\boldsymbol{C}}^{\boldsymbol{I}}\right) \quad (42)$$

Finally, in order to determine the viscoelastic moduli $\widehat{\boldsymbol{C}}^{\boldsymbol{eff}}$, the strain concentration tensor of the coating phase $\widehat{\boldsymbol{A}}^{\boldsymbol{C}}$ is given by for both DI and RDI models as follows:

$$\widehat{\boldsymbol{A}}^{\boldsymbol{C,1,2}} = \left(\widehat{\boldsymbol{A}}^{\boldsymbol{IC,1,2}}\right)^{-\boldsymbol{1}} : \widehat{\boldsymbol{A}}^{\boldsymbol{I,1,2}} \quad (43)$$

In the following section, the extended Mori-Tanaka homogenization scheme based on both DI and RDI models are compared with the homogenization scheme using exact solutions derived by Hervé and Zaoui (1990) in the case of isotropic properties with coated spherical inclusions embedded in a matrix material.

3 Validation and application

3.1 Validation of the RDI model for isotropic viscoelastic properties and spherical coated inclusions

In the following, the composite is constituted of three isotropic phases, where the matrix phase, inclusion and coating phases are supposed to be linear isotropic viscoelastic. Both

inclusions and coated inclusions are homothetic and form spherical composite inclusions embedded in the matrix phase. Therefore, the effective viscoelastic moduli of the composite are isotropic and $\widehat{\boldsymbol{C}}^{\boldsymbol{eff}}$ can be decomposed into the spherical and the deviatoric parts described by the effective bulk and shear modulus $\hat{k}^{eff}$ and $\hat{\mu}^{eff}$ as follows:

$$\widehat{\boldsymbol{C}}^{\boldsymbol{eff}} = 3\hat{k}^{eff}\boldsymbol{J} + 2\hat{\mu}^{eff}\boldsymbol{K} \tag{44}$$

where $\boldsymbol{J}$ and $\boldsymbol{K}$ are orthogonal projection tensors (Walpole, 1981) defined as $J_{ijkl} = \frac{1}{3}\delta_{ij}\delta_{kl}$ *and* $K_{ijkl} = \frac{1}{2}\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl}\right)$.

As a consequence of the homogenized effective behavior in Eq. (36), the effective shear and bulk moduli are described as:

$$\hat{k}^{eff} = \hat{k}^{M} + f^{I}(\hat{k}^{I} - \hat{k}^{M})\hat{A}^{I,k} + f^{C}(\hat{k}^{C} - \hat{k}^{M})\hat{A}^{C,k} \tag{45}$$

$$\hat{\mu}^{eff} = \hat{\mu}^{M} + f^{I}(\hat{\mu}^{I} - \hat{\mu}^{M})\hat{A}^{I,\mu} + f^{C}(\hat{\mu}^{C} - \hat{\mu}^{M})\hat{A}^{C,\mu} \tag{46}$$

In Eqs. (45) and (46), the $\hat{A}^{\alpha,\mu}$ and $\hat{A}^{\alpha,k}$ strain concentration factors with $\alpha = I, C$ represent the spherical and deviatoric parts of the relationships described in Eq. (39) and (43) respectively for the phases I and C. Thereby, all strain concentration tensors contain the tensor $\widehat{\boldsymbol{T}}_{\boldsymbol{\alpha}}^{\boldsymbol{I,DI}}(\widehat{\boldsymbol{C}}^{\boldsymbol{\alpha}})$ where $\alpha = M, C$. In case of spherical shapes for the inclusions and the coated inclusions, this tensor is formulated as (Eshelby, 1957; Mura, 1987):

$$\widehat{\boldsymbol{T}}_{\boldsymbol{\alpha}}^{\boldsymbol{I,DI}}(\widehat{\boldsymbol{C}}^{\boldsymbol{\alpha}}) = \frac{\boldsymbol{J}}{3\hat{k}^{\alpha} + 4\hat{\mu}^{\alpha}} + \frac{3(\hat{k}^{\alpha} + 2\hat{\mu}^{\alpha})\boldsymbol{K}}{5\hat{\mu}^{\alpha}(3\hat{k}^{\alpha} + 4\hat{\mu}^{\alpha})} \tag{47}$$

Therefore, the effective behavior can be obtained from the developed strain concentration equations of section 2.4 together with Eqs. (45)-(47), for both DI and RDI models.

For comparison and validation of the proposed models, an extended Mori-Tanaka homogenization scheme based on the analytical exact solutions obtained by Hervé and Zaoui (1990) for the spherical coated inclusion problem with isotropic elastic constituents is applied. Here, the exact formulation of Hervé and Zaoui (1990) is adapted to include the interphase properties as well as linear viscoelasticity. The corresponding extension of the analytic expressions is presented in Appendix A.1. Nevertheless, this exact formulation is only restricted to the case of spherical coated inclusions.

In the following, Figures 2 and 3 represent the symbolic normalized effective bulk modulus $\widehat{K}^{eff}$ and shear modulus $\widehat{G}^{eff}$, respectively as function of the complex LC variable p for two different coated inclusion volume fractions. These symbolic normalized effective moduli are defined as (see also Hashin, 1991; Dinzart and Sabar, 2014;):

$$\widehat{K}^{eff} = \frac{p\,\hat{k}^{eff}}{k^M} \quad with \;\; \hat{k}^M = k^M \Big/ \left(p + \frac{k^M}{k^{\upsilon M}}\right) \tag{48}$$

$$\widehat{G}^{eff} = \frac{p\,\hat{\mu}^{eff}}{\mu^M} \quad with \;\; \hat{\mu}^M = \mu^M \Big/ \left(p + \frac{\mu^M}{\eta^M}\right) \tag{49}$$

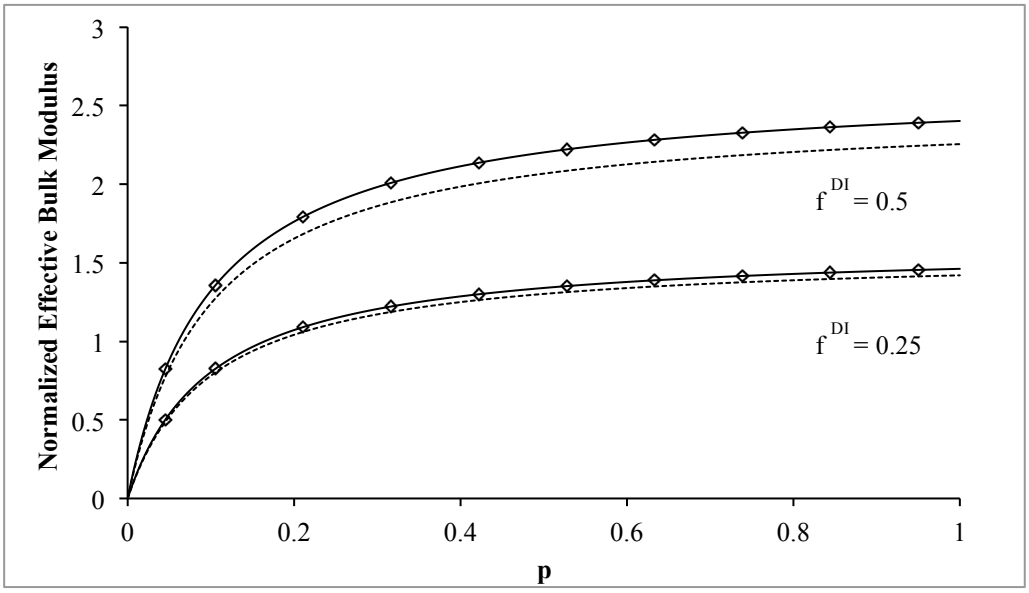


Figure 2: Symbolic normalized effective bulk modulus $\widehat{K}^{eff}$ (Eq. (48)) as a function of the LC complex variable p for coated inclusion volume fraction of 0.25 and 0.5 obtained from the extended Mori-Tanaka homogenization scheme based on coated inclusions using the exact solution from Hervé and Zaoui (1990) (solid lines), the DI model (dotted lines), the RDI model (solid lines with diamonds).

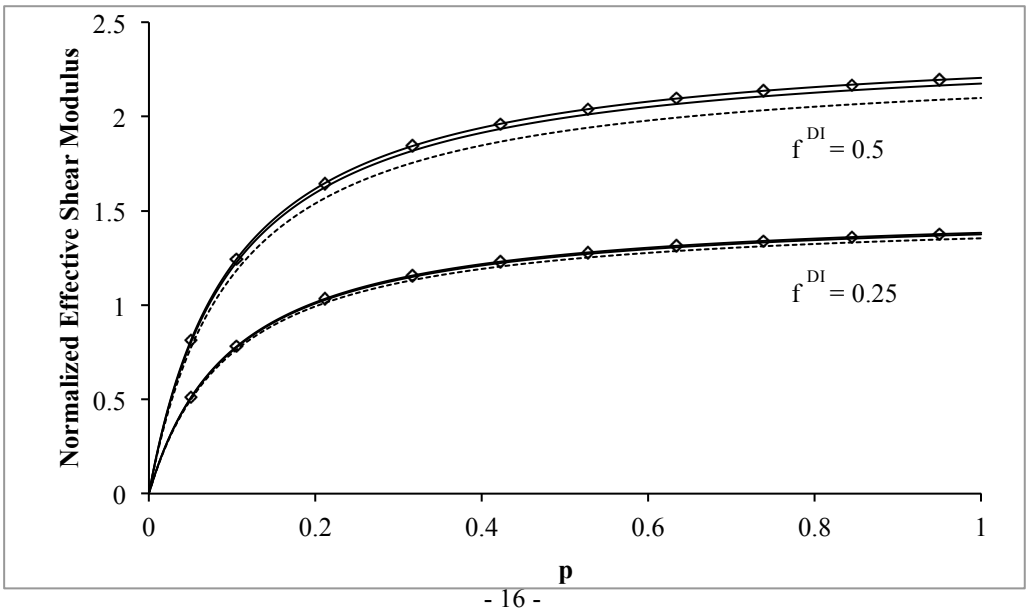


Figure 3: Symbolic normalized effective shear modulus $\hat{G}^{eff}$ (Eq. (49)) as a function of the LC complex variable p for coated inclusion volume fraction of 0.25 and 0.5 obtained from the extended Mori-Tanaka homogenization scheme based on coated inclusions using the exact solutions from Hervé and Zaoui (1990) (solid lines), the DI model (dotted lines), the RDI model (solid lines with diamonds).

The ratios of the elastic and viscous parts are set with regard to the matrix phase M to $\frac{k^M}{k^{v^M}} = \frac{\mu^M}{\eta^M} = \frac{1}{10}[s^{-1}]$. Additionally, normalized material properties are further applied for the other phases and set identically to a recent study for a four-phase self-consistent approach (Dinzart et al., 2016). Thereby, the micromechanical models are evaluated for a defined contrast between the different phases. Here, the bulk and the shear moduli contrasts between the interphase and the matrix phase are set to $\frac{k^C}{k^M} = 25$ and $\frac{\mu^C}{\mu^M} = 25$ to study the models capabilities for high moduli contrasts. The inclusion properties are defined by $\frac{k^I}{k^M} = 6$ and $\frac{\mu^I}{\mu^M} = 6$. The volume fractions of the inclusion and the coating are related by $f^C = 0.7f^I$, which corresponds to a relative thickness of the coating with respect to the inclusion radius a defined as $\frac{\Delta a}{a} = 0.2$.

Subsequently to the evaluation of the symbolic normalized effective properties for a fixed coated inclusion volume fraction, the direct effective behavior is investigated. Therefore, the coated inclusion volume fractions $f^{DI} = f^I + f^C$ are varied in a range between 0 and 0.5 for $p \to \infty$. The corresponding results for the effective bulk and shear moduli are shown in Figures 4 and 5, respectively.

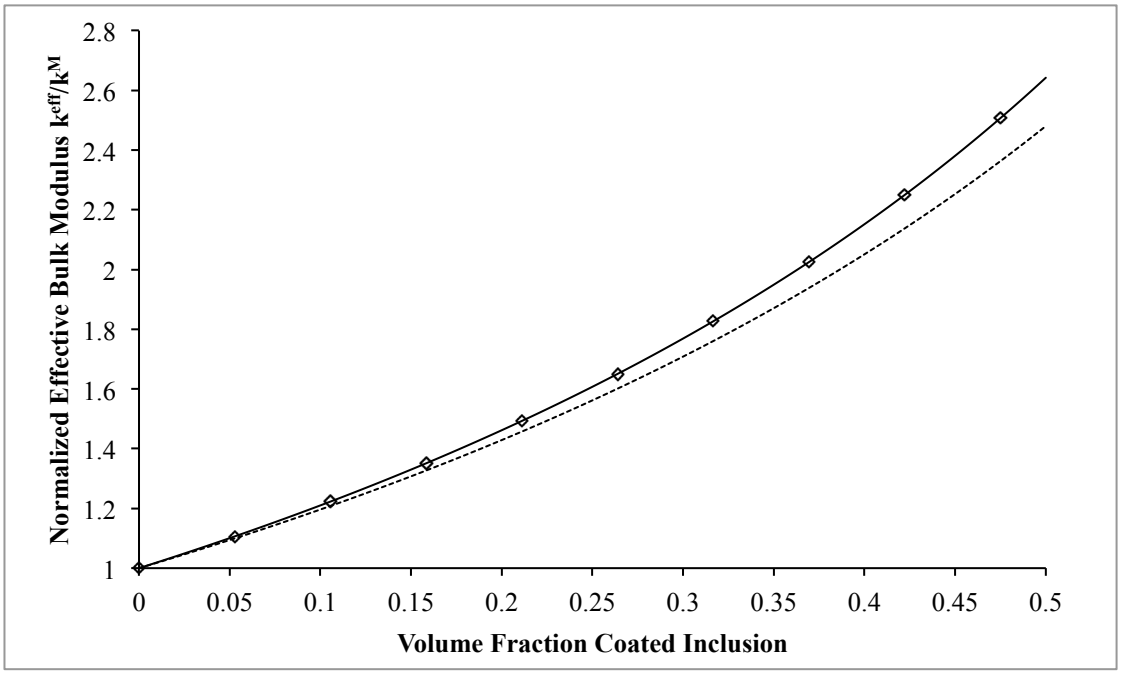


Figure 4: Normalized effective bulk modulus as a function of the coated inclusion volume fraction obtained from the extended Mori-Tanaka homogenization scheme based on coated inclusions using the exact solutions from Hervé and Zaoui (1990) (solid lines), the DI model (dotted lines), the RDI model (solid lines with diamonds).

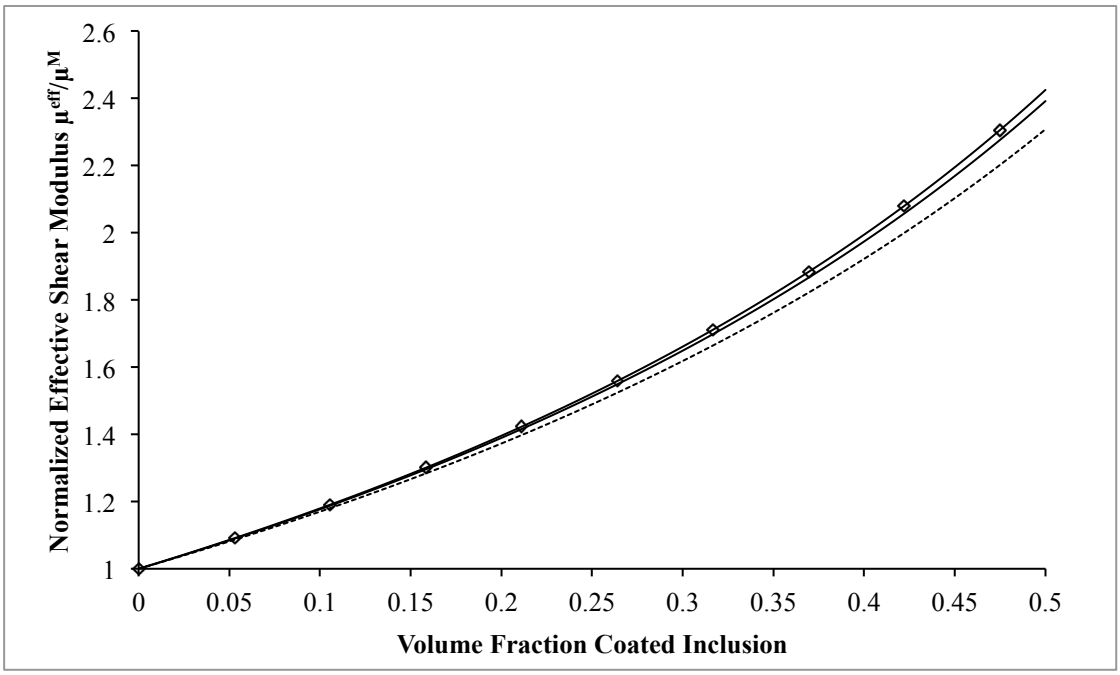


Figure 5: Normalized effective shear modulus as a function of the coated inclusion volume fraction obtained from the extended Mori-Tanaka homogenization scheme based on coated inclusions using the exact solutions from Hervé and Zaoui (1990) (solid lines), the DI model (dotted lines), the RDI model (solid lines with diamonds).

As an important result, the effective bulk modulus obtained by the homogenization scheme based on the RDI model exactly coincides analytically with the exact solution (see appendix A.1). This result is confirmed by the perfect agreement between the RDI model and the exact solution observed in Figures 2 and 4. In contrast, the DI model shows an increasing deviation from the exact solution especially at higher coated inclusion volume fractions. The results in Figure 3 and 5 indicate that effective shear moduli predicted by both models are not identical to the exact reference model. However, the RDI model provides an improved prediction with respect to the DI model concerning the convergence to the exact solution. Up to a coated inclusion volume fraction of 0.3, the prediction of the RDI model corresponds to the exact solution. For higher volume fractions, this homogenization scheme provides a slightly stiffer effective shear modulus than the one based on the exact solution from Hervé and Zaoui (1990). This is due to the fact that the extended Mori-Tanaka homogenization scheme is pushed in the present case over its established limits.

In summary, the comparison of effective bulk and shear properties validates the proposed homogenization Mori-Tanaka scheme for low concentration of coated inclusions in the case of the proposed RDI model for three-phase viscoelastic composites with interphase. In the following, only the RDI model is considered to predict the experimental DMA data in the frequency domain for a carbon-black filled styrene butadiene rubber extracted from Diani et al. (2013).

3.2 Application to the viscoelastic properties of a carbon-black filled styrene butadiene rubber and comparison with experimental DMA data

Subsequent to the validation of the homogenization scheme in last section 3.1, the extended Mori-Tanaka scheme based on the RDI model is now confronted to experimental data in the case of a carbon-black filled styrene butadiene rubber reported in Diani et al. (2013). As a first approximation, this heterogeneous material can be realistically represented by a three-phase composite with spherical coated inclusions embedded in the bulk unfilled rubber. The carbon-black fillers represent the inclusions with purely elastic behavior, and there exists a filler-matrix interphase corresponding to a rubber-layer bounded to the filler (Diani et al., 2013). Therefore, the material characterization of a carbon-black filled styrene butadiene rubber studied by Diani et al. (2013) is considered. The authors provide the necessary material parameters to describe a spherical stiff inclusion phase, surrounded by an interphase, which shows altered properties of the soft matrix. The relevant material parameters are summarized in Table 1.

	First parameter	**Second parameter**
Matrix (viscoelastic)	Young's modulus: Generalized Maxwell model parameters	Bulk modulus: 3500 MPa
Inclusion (elastic)	Young's modulus: 70000 MPa	Poisson's ratio: Quasi incompressible (present case 0.499)
Coating (viscoelastic)	Young's modulus: obtained from the matrix parameters (see Eq. (51))	Bulk modulus: 3500 MPa

Table 1: Material parameters for the three homogeneous isotropic phases by (Diani et al., 2013)

The viscoelastic behavior of the matrix and the coating is described by twelve generalized Maxwell model parameter sets. These parameters describe the complex Young's modulus E^* of the matrix phase M. This complex material parameter is decomposed in a real and

imaginary part representing the storage and the loss moduli, respectively. Following the work of Park and Schapery (Park and Schapery, 1999), the real and imaginary part of the complex Young's modulus are defined in the frequency domain with the relaxation time τ_i, the relaxation strength E_i and the equilibrium modulus E_e by:

$$E_M^*(\omega) = E_M'(\omega) + iE_M''(\omega)$$
$$with\ \ E_M'(\omega) = E_e + \sum_{i=1}^{m} E_i \frac{\omega^2\tau_i^2}{\omega^2\tau_i^2 + 1} \tag{50}$$
$$and\ \ E_M''(\omega) = \sum_{i=1}^{m} E_i \frac{\omega\tau_i}{\omega^2\tau_i^2 + 1}$$

For the description of the viscoelastic rubber matrix behavior, the experimental dynamic mechanical analysis (DMA) curves of Diani and co-workers (Diani et al., 2013) are considered as shown in Figure 6. Here, to describe the viscoelastic behavior of the matrix, Eq. (50) is considered, and the same number of generalized Maxwell parameters (m=12), as well as the same values for the relaxation strengths E_i and for E_e. However, in order to well fit the experimental DMA data, the associated relaxation times were recalibrated for the present study. The corresponding Prony series parameters are detailed in Table 2.

Relaxation time τ_i [s]	**Relaxation strength E_i** [MPa]
12.038×10^{-8}	320.3
58.455×10^{-8}	293.7
28.392×10^{-7}	235.3
13.787×10^{-6}	86.88
66.950×10^{-6}	18.79
32.526×10^{-5}	4.283
15.802×10^{-4}	2.488
76.765×10^{-4}	0.891
37.271×10^{-3}	0.394
18.103×10^{-2}	0.237
87.945×10^{-2}	0.870×10^{-2}
42.705×10^{-1}	0.271×10^{-3}
	Equilibrium modulus: E_e = 2.3 MPa

Table 2: Prony series material parameters of the viscoelastic rubber matrix identied for the description of the experimental DMA data as reported in Figure 6. The relaxation strengths E_i [MPa] and the

equilibrium modulus E_e [MPa] are the same as the ones reported in Diani et al. (2013). The relaxation times τ_i [s] have been recalibrated in the present work.

To verify the implementation of the viscoelastic matrix properties compared to the original experimental data, the storage and the loss modulus of the rubber matrix are displayed in Figure 6. It is shown, that the viscoelastic behavior of the matrix can be described with the updated relaxation times using Eq. (50) and Table 2.

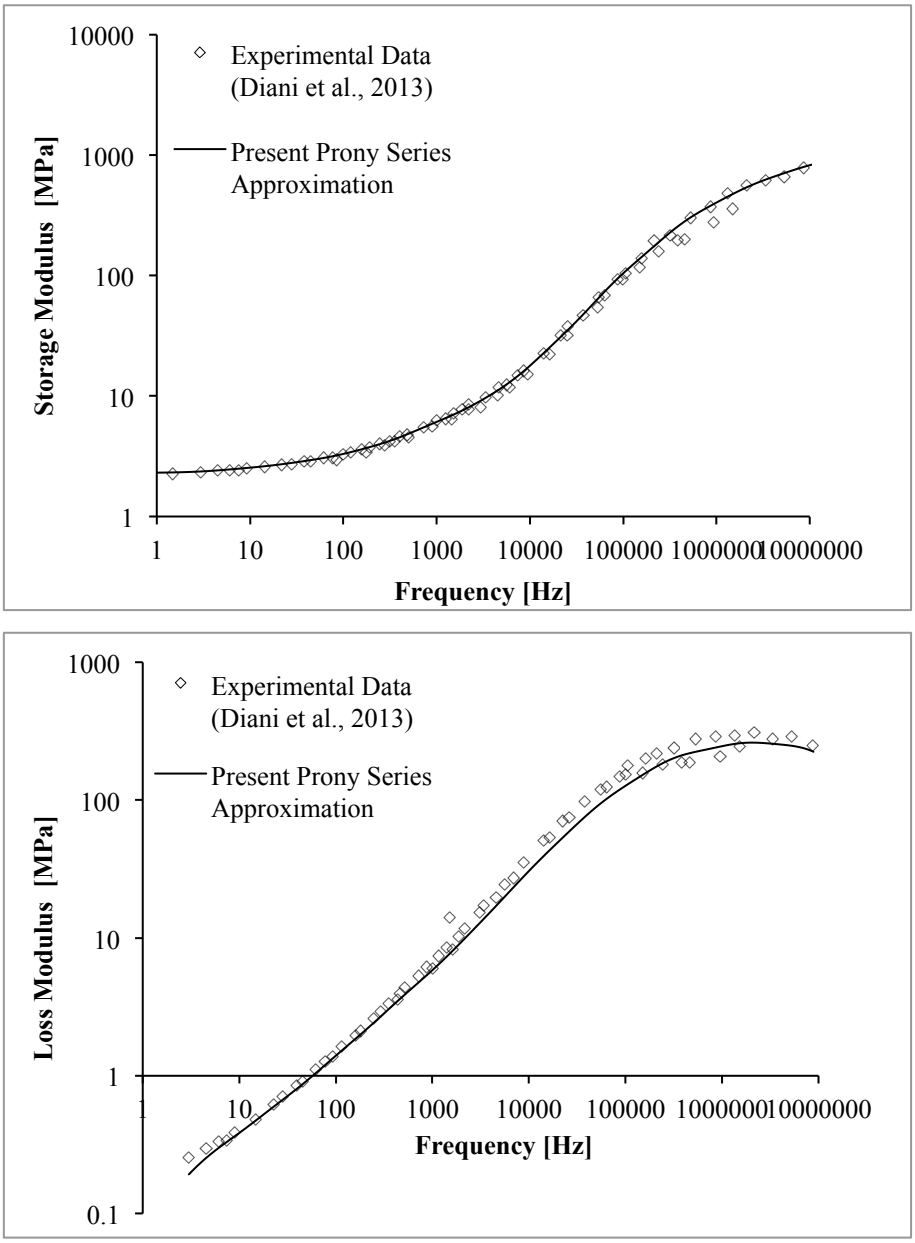


Figure 6: Comparison of the experimental matrix storage/loss moduli with the identified Prony series parameters introduced in Eq. (50) and Table 2.

The Young's modulus of the interphase is formulated based on the presented matrix behavior. Following the description of Diani et al. (2013), the mechanical properties of the interphase (coating phase C) depend on the complex Young's modulus of unfilled bulk rubber matrix ${E_M}^*(\omega)$ as well as its constant value in the glassy state denoted E_{MG}^* (purely elastic state) where $E_{MG}^* = E_{MG}'$ since $E_{MG}'' = 0$ for elastic materials (Diani et al., 2013):

$$E_C^*(\omega) = E_{MG}^* + x(E_M^*(\omega) - E_{MG}^*) \tag{51}$$

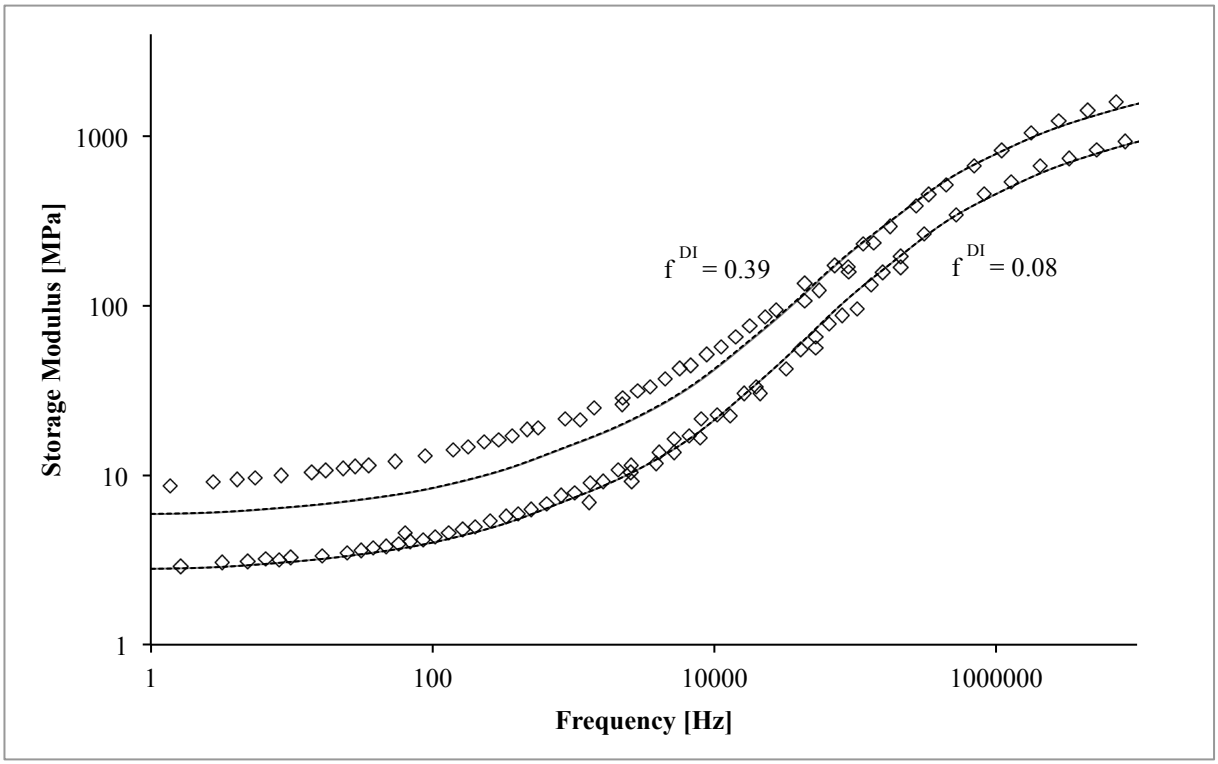
In accordance to the work of Diani et al. (2013), the factor $x \in [0,1]$ is set to a value of 0.95. Thus, all material parameters displayed in Table 1 can be specified and are subsequently applied to represent the mechanical behavior of each phase of the composite. These material parameters are incorporated in the extended Mori-Tanaka homogenization based on the exact solutions of Hervé and Zaoui (1990) and based on the proposed RDI model to derive the effective storage and loss moduli as functions frequency and of coated inclusion volume fractions. As a consequence, the complex moduli of the three-phase composite can be developed using the proposed modeling approaches for f^{DI} not exceeding 40%. The work presented in Diani et al. (Diani et al., 2013) provides experimental DMA results for composites with different volume fractions of coated inclusions. Thus, these experimental data can be applied for further validation of the proposed RDI model for the determination of the effective viscoelastic behavior of this three-phase composite as compared to section 3.1. In the present study, two composite material datasets are selected from the experimental DMA results. A summary of the corresponding volume fraction of each phase is given in Table 3 for two composite materials denoted M5 ($f^{DI} = 0.08$) and M30 ($f^{DI} = 0.39$), respectively not to exceed 40% of coated inclusion volume fraction.

Material Name	**Volume Fraction Inclusion** f^I	**Volume Fraction Coating** f^C	**Volume Fraction Coated Inclusion** f^{DI}
M5	0.05	0.03	0.08
M30	0.28	0.11	0.39

Table 3: Selected composite materials with different coated volume fractions based on the materials data given in (Diani et al., 2013) for comparison with the extended Mori-Tanaka homogenization based on the exact solutions of Hervé and Zaoui (1990) and based on the proposed RDI model.

For the subsequent comparison between the experimental results and the micromechanical models, the extended homogenization schemes based on the exact solution and the RDI model

are both investigated. Figure 7 represents the effective complex modulus of the composite, which splits into the real and complex parts, the storage and loss moduli, respectively. Thereby, both Materials M5 and M30 are implemented with their corresponding volume fractions. The material properties described in the previous section are identical among the different materials in order to investigate the influence of the composite volume fraction on the models prediction capabilities.



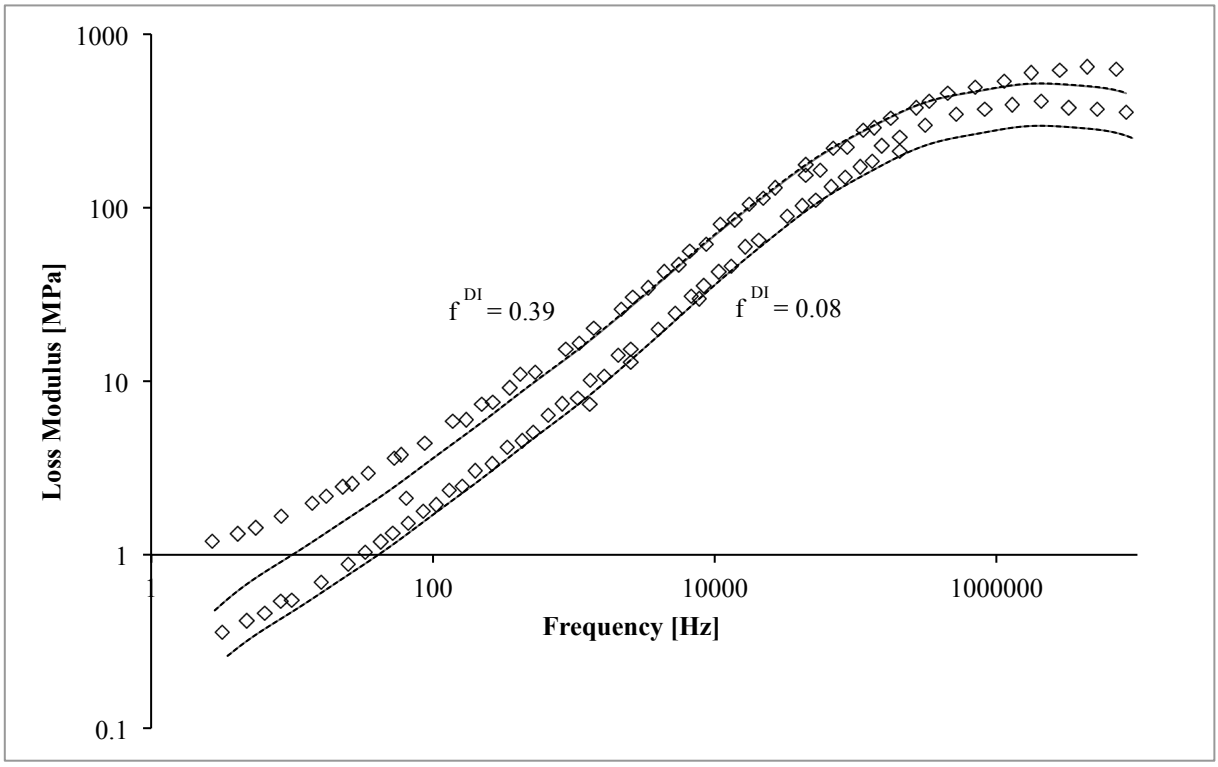


Figure 7: Storage and loss moduli of two composites M5 (f^{DI}=0.08) and M30 (f^{DI}=0.39): comparison between experimental DMA data extracted from Diani et al. (2013) (diamonds) and predictions obtained from the extended Mori-Tanaka homogenization scheme based on coated inclusions using the exact solutions from Hervé and Zaoui (1990) (solid lines) and the proposed RDI model detailed in section 2 (dotted lines).

It is observed from Figure 7, that the effective behavior described with the exact analytical solution and the RDI model are superimposed independently for the different volume fractions of coated inclusions and for the broad range of frequency. This result is seen as a confirmation of the results presented in Figures 2 to 5. Nevertheless, the prediction of the experimental results shows a distinct influence of the respective volume fraction. In case of the M5 material containing a small amount of coated inclusions with $f^{DI} = 0.08$, the storage modulus is predicted with high accuracy for all frequencies. In addition, the loss modulus is correctly approximated, especially in the frequency range between 10^1 Hz and 10^6 Hz. The deviations at lower and higher frequencies are principally based on the initial Prony parameters of the matrix material, which show the same diverging tendency for the loss modulus in Figure 6. Furthermore, the M30 material with a volume fraction of $f^{DI} = 0.39$ represents the limit of the present extended Mori-Tanaka scheme. Especially at low frequencies, an increased deviation from the experimental data is found. However, starting from frequencies greater than 10^3 Hz, both storage and loss modulus converge closer to the experimental data. Thus, the storage and loss modulus are predicted with the same performance as the M5 material for higher frequencies.

In summary, increased coating inclusion volume fractions are leading to an underestimation of the complex modulus at lower frequencies. Nevertheless, at these volume fractions the experimental and numerical datasets are still well converging at frequencies above 10^3 Hz. For this reason, the proposed micromechanical approach is also applicable for composites with volume fractions similar to the M30 material. Therefore, the material parameter f^C and x should be reconsidered. In the reference study by Diani et al. (Diani et al., 2013), these two parameters were introduced as fitting parameters and are set in order to calibrate a four-phase self-consistent scheme (Maurer, 1990). Thus, the identically applied values of f^C and x in this study could possibly not describe optimal choices of these parameters in the framework of the RDI model together with an extended Mori-Tanaka scheme. With regard to the definition of the complex modulus of the interphase in Eq. (51), the parameter x is notably seen as the

determining factor to raise the interphase properties especially in the range of low frequencies. In the present study, these fitting parameters are considered identical to the reference work of Diani et al. (Diani et al., 2013) to investigate the relevance of the present micromechanical model. Thus, it is shown that an appropriate prediction of the viscoelastic behavior can be given already without a recalibration of the fitting parameters. Consequently, the presented RDI model is also efficient in the framework of the present homogenization scheme for the description of linear viscoelastic experimental data of three-phase composites like carbon-black filled styrene butadiene rubber.

4 Conclusion

In this study, the reconsidered DI model (abbreviated RDI model) first introduced by Aboutajeddine and Neale (2005) was adopted to the case of three-phase viscoelastic composites with coated inclusions and with different mechanical properties in an extended Mori-Tanaka homogenization scheme. The entire micromechanical framework was formulated in the frequency domain to include linear viscoelasticity by using the correspondence principle and Laplace-Carson transforms. The classic DI model originally proposed by Hori and Nemat-Nasser (1993) was also considered following the same Mori-Tanaka homogenization scheme but including different strain concentration tensors for three phase composites in order to enable some comparisons with the RDI model. Both models were applied to three-phase composites with inclusions and interphases constituted of isotropic viscoelastic phases embedded in a viscoelastic matrix. To validate the proposition of the RDI model based homogenization scheme, the application to spherical coated inclusions and the comparison with the exact solution by Hervé and Zaoui (Hervé and Zaoui, 1990) were presented. It was shown that the RDI model provides better predictions than the DI model. Herein, it was emphasized, that the proposed Mori-Tanaka extension in the RDI model predicts exactly the same effective bulk modulus as the exact reference model. Additionally, the shear moduli were seen to be very close to the reference solution in the case of the RDI model, which was not the case for the DI model. Based on this satisfying validation, the homogenization schemes based on the RDI model and on the exact solutions were applied to predict the effective viscoelastic behavior of real three-phase composites with interphase. The models predictions were confronted to experimental data in the case of a carbon-black filled styrene butadiene rubber reported in Diani et al. (2013). As a first approximation, this

heterogeneous material was realistically represented by a three-phase composite with spherical coated inclusions embedded in the bulk unfilled rubber. The effective viscoelastic behavior was estimated with a good accuracy in terms of the storage and loss moduli for different volume fractions of coated inclusions up to 40%. Furthermore, the RDI model coincides with the exact solution independently of the coated inclusion volume fraction. In case of high volume fractions of coated inclusions ($f^{DI} = 0.39$) and high frequencies superior to 10^3 Hz, the storage and loss moduli of the composite were still well represented. However, the complex moduli at low frequencies were significantly lower than the ones given by experimental results. This effect could be explained with the choice of the mechanical interphase properties at lower frequencies, which were identical to the work of Diani et al. (Diani et al., 2013). Currently, the experimental characterization of the interphase (micro-geometry, mechanical behavior) in polymer-based composites remains a challenging issue (Schöneich et al., 2015). The present work provides an efficient micromechanical model for viscoelastic composites based on the coated-inclusion problem solved with the RDI model. The present approach can be applied to further applications in an extended Mori-Tanaka scheme or in a four-phase self-consistent scheme (see Dinzart et al., 2016) without a restriction to spherical or cylindrical inclusions in contrast with the exact solutions. Thus, anisotropic viscoelastic behavior and non-homothetic inclusions and coated inclusions could be considered in further studies with the present model.

Appendix A

A.1. Exact strain concentration solution for spherical coating inclusion with isotropic properties

The exact solution by Hervé and Zaoui (Hervé and Zaoui, 1990) is extended to the application to three phases to include the coating (interphase) C, which surrounds the spherical inclusion I. The coated inclusion (*DI*=*I*+*C*) is embedded in the matrix phase M. Therefore, the Mori-Tanaka scheme is also applied as well as the further extension to viscoelasticity using the correspondence principle and the involved Laplace-Carson transformation. Thus, the fundamental exact strain concentration equations of the dilatational and deviatoric part denoted as $\hat{\theta}_{C,I}$ and $\hat{\bar{e}}_{C,I}$ adopted from the original work are still valid and implemented as:

$$\hat{\theta}_C = \frac{\left(3\hat{k}^M + 4\hat{\mu}^M\right)\left(3\hat{k}^I + 4\hat{\mu}^C\right)}{\left(3\hat{k}^C + 4\hat{\mu}^M\right)\left(3\hat{k}^I + 4\hat{\mu}^C\right) + 12c(\hat{\mu}^M - \hat{\mu}^C)\left(\hat{k}^C - \hat{k}^I\right)}\hat{\theta}_M$$

$$\hat{\theta}_I = \frac{\left(3\hat{k}^M + 4\hat{\mu}^M\right)\left(3\hat{k}^C + 4\hat{\mu}^C\right)}{\left(3\hat{k}^C + 4\hat{\mu}^M\right)\left(3\hat{k}^I + 4\hat{\mu}^C\right) + 12c(\hat{\mu}^M - \hat{\mu}^C)\left(\hat{k}^C - \hat{k}^I\right)}\hat{\theta}_M \tag{A.1}$$

$$\hat{\bar{e}}_C = \frac{15(1-\upsilon^M)X_0}{(1-c)} \times \frac{(X_0 - 1)\left(A + 60c(1-\upsilon^C)\left(\eta_1 c^{7/3} - \eta_2(7 - 10\upsilon^C)\right)\right) + 35(1-\upsilon^C)\eta_2\eta_3(1-c)}{\Delta}\hat{e}_M \tag{A.2}$$

$$\hat{\bar{e}}_I = 225(1-\upsilon^M)(1-\upsilon^C)X_0 \times \frac{-4(X_0 - 1)\left(\eta_1 c^{7/3} - \eta_2(7 - 10\upsilon^C)\right) + 35\eta_2(1-\upsilon^C)}{\Delta}\hat{e}_M$$

In the present contribution, no changes regarding the involved parameters $c, X_0, A, \eta_1, \eta_2, \eta_3$ and Δ have been made. Thus, these parameters are defined with reference to the original work of Hervé and Zaoui (Hervé and Zaoui, 1990) and the corresponding Poisson's ratio $\upsilon^\alpha, \alpha = I, C, M$ as:

$$
\begin{aligned}
&c = f^I / f^{DI} \\
&\Delta = [2(4-5\upsilon^M)C + (7-5\upsilon^M)AX_0](X_0 - 1) + 525\eta_2(1-\upsilon^C) \\
&\qquad \times[2\alpha(\upsilon^C - \upsilon^M)c + (1-\upsilon^M)\eta_3]X_0 \\
&\eta_1 = (49 - 50\upsilon^C\upsilon^I)\alpha + 35(1+\alpha)(\upsilon^I - 2\upsilon^C) + 35(2\upsilon^I - \upsilon^C) \\
&\eta_2 = (7+5\upsilon^I)(1+\alpha) + 4(7-10\upsilon^I) \\
&\eta_3 = 2(1+\alpha)(4-5\upsilon^C) + 7 - 5\upsilon^C \\
&\alpha = (\hat{\mu}^I / \hat{\mu}^C) - 1 \\
&X_0 = \hat{\mu}^M / \hat{\mu}^C \\
&A = -4[\eta_3 - 2\alpha(4-5\upsilon^C)c]\left[\eta_1 c^{7/3} - \eta_2(7-10\upsilon^C)\right] \\
&\qquad -126\alpha\,\eta_2 c\left(1 - c^{2/3}\right)^2 \\
&C = -[\eta_3 + \alpha(7-5\upsilon^C)c]\left[4\eta_1 c^{7/3} + \eta_2(7+5\upsilon^C)\right] \\
&\qquad -126\alpha\,\eta_2 c\left(1 - c^{2/3}\right)^2
\end{aligned}
\tag{A.3}
$$

In the homogenization step, the strain concentration factors of the inclusion and the coating following the exact solution by Hervé and Zaoui are emerging directly from Eqs. (A.1) (A.2) and (A.3) and from the homogenization procedure described in Eqs. (44-46). At this point, it can be remarked that the effective solution for the dilatational part of the exact approach is identical to the solution for the effective bulk modulus from the homogenization scheme based on the RDI model. In contrast, the classic DI model cannot retrieve this result (see Figures 2 and 4).

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