Effective meso properties for fibre reinforced polymer curing

Christian Dammann^{1*}, Rolf Mahnken¹ and Peter Lenz¹

Micro Abstract

Our work presents volumetric effective properties in dependence on the degree of cure. They are obtained by homogenization for a representative unit cell on the heterogeneous microscale. To this end, analytical solutions for (n)- and (n + 1)-layered composite sphere models are derived. In a numerical study it is demonstrated that the effective properties lie within certain bounds. Moreover, application of the effective properties to the curing of fibre reinforced polymers is investigated.

¹Chair of Engineering Mechanics, Paderborn University, Paderborn, Germany ***Corresponding author**: Dammann@ltm.upb.de

Introduction

During resin transfer molding, a production process for fibre reinforced composites (FRP), starting with the initial uncured state the matrix of the FRP as a mixture of resin (r) and curing agent (ca) undergoes a polymerization process during curing to a solid material (sol). A simulation of such a production process requires the thermo-chemo-mechanical effective material properties (also known as *overall* or *bulk properties*, respectively) in terms of a so called degree of cure z. For this purpose two conceptions for the matrix, a *homogeneous mixture* and a *heterogeneous mixture* can be distinguished: A homogeneous mixture for all constituents, resin, curing agent and solidified material results e.g. in the compression moduli derived in [1] and [2] depending on z. These results are regarded to as (Voigt and Reuss) bounds for more advanced approaches, where a geometrical arrangement of the microscale is taken into account by a heterogeneous mixture. In this context, the (2)-layered composite sphere model originally introduced in [3] is extended to account for thermo-chemo-mechanical coupling in [4]. In this work, n spherical constituents accounting for thermo-chemo-mechanical coupling are taken into account.



Figure 1. Two idealizations of the inclusion: a) heterogeneous *n*-layered composite sphere, b) homogeneous sphere and c) 3-layered composite sphere (micro-RVE) over time: initial uncured, partially cured and fully cured state.

1 An n-layered composite sphere model for thermo-chemo-mechanical loading

The two different idealizations shown in Fig. 1.a, b have in common that a spherical inclusion is embedded in an infinite homogeneous medium denoted as matrix. Both are subjected to a

uniform thermal loading in terms of a prescribed temperature θ , a chemical loading in terms of a degree of cure z and a mechanical loading in terms of a prescribed pressure p. For a heterogeneous mixture denoted as *het* an n-layered composite sphere model according to Fig. 1.a is assumed with n spherical inclusions on the microscale. The constituents $i \in [1, n]$ with partial volumes $v^{(i)}$ and corresponding radii R_i assemble to a total volume v. In order to obtain effective properties, this is considered as a micro-RVE.

We assume a decomposition of the strain tensor according to the geometric linear theory into elastic, thermal, and chemical parts $\boldsymbol{\varepsilon}^{(i)} = \operatorname{sym}\{\nabla \boldsymbol{u}^{(i)}\} = \boldsymbol{\varepsilon}^{(i)}_{el} + \boldsymbol{\varepsilon}^{(i)}_{th} + \boldsymbol{\varepsilon}^{(i)}_{cur}$ for $i \in [1, n]$, where $\boldsymbol{u}^{(i)}$ is the displacement vector defined at any point $P^{(i)}$ within the volume v. The equilibrium condition is written at any point $P^{(i)}$ as div $\boldsymbol{\sigma}^{(i)} = \mathbf{0}$, $i \in [1, n]$. Furthermore, the following constitutive equations are employed for the individual strain contribution

1.
$$\boldsymbol{\varepsilon}_{mech}^{(i)} = \left(\mathbb{C}^{(i)}\right)^{-1} : \boldsymbol{\sigma}^{(i)}, \quad 2. \quad \boldsymbol{\varepsilon}_{th}^{(i)} = \alpha^{(i)} \Delta \theta \mathbf{1}, \quad 3. \quad \boldsymbol{\varepsilon}_{cur}^{(i)} = \beta^{(i)} \mathbf{1}, \quad i \in [1, n], \quad (1)$$

with fourth order elasticity tensor $\mathbb{C}^{(i)}$, heat- and curing-dilatation coefficients $\alpha^{(i)}$ and $\beta^{(i)}$ for phase *i* and a homogeneous temperature change $\Delta \theta = \Delta \theta^{(i)}$ for $i \in [1, n]$. We remark, that Eq. (1.1) and Eq. (1.2) are dependent on the state variables $\sigma^{(i)}$ and $\Delta \theta$, respectively, whereas Eq. (1.3) is independent of state variables. The boundary value problem (BVP) is constituted by the Navier-Lamé differential equation (resulting from the equilibrium condition in radial direction) together with the continuity conditions at the interfaces and a boundary condition where we exploit that the micro-RVE is subjected to a pressure p.

Next, an equivalent homogeneous spherical inclusion is considered with a loading as shown in Fig. 1.b. The basic relations for the *n*-layered inclusion are applied to this inclusion by setting n = 1. Analogously to Eq. (1), the constitutive equations

1.
$$\boldsymbol{\varepsilon}_{mech} = \mathbb{C}^{-1} : \boldsymbol{\sigma}, \qquad 2. \quad \boldsymbol{\varepsilon}_{th} = \alpha \Delta \theta \mathbf{1}, \qquad 3. \quad \boldsymbol{\varepsilon}_{cur} = \beta z \mathbf{1},$$
 (2)

are employed. In Eq. (2) we have the fourth order elasticity tensor \mathbb{C} , the heat- and the curingdilatation coefficients α and β , the temperature change $\Delta\theta$ and the degree of cure z, respectively. Upon comparing Eq. (2) to Eq. (1), we observe the same structures for Eq. (2.1) and Eq. (1.1) as well as for Eq. (2.2) and Eq. (1.2), since they are dependent on the state variables p and $\Delta\theta$, respectively. Contrary to the temperature θ , the degree of cure z cannot be assumed homogeneous within the heterogeneous mixture. Therefore, different structures are defined for Eq. (2.3) and Eq. (1.3). Setting n = 1 the Navier-Lamé differential equation together with a pressure boundary condition constitutes the BVP for the equivalent homogeneous sphere.

In order to obtain the effective properties, as a homogenization condition, the displacement at the outer boundary of the equivalent homogeneous sphere is equated to the one of the composite sphere. This yields the effective compression modulus K, as well as the effective heat-dilatation coefficient α and the effective curing-dilatation coefficient β , see [5] for more details.

2 Application to curing

In this section the results for the special case with three phases for a homogeneous mixture (hom) and the heterogeneous mixture (het) are applied to curing. Related to hom, an equally distributed mixture is assumed for n phases on the microscale according to Fig. 1.b. For this, Voigt and Reuss bounds derived in [5] are applied. In particular we are interested in the effective compression moduli, K_V and K_R , and the effective heat-dilatation coefficient, α_V and α_R , each in terms of the degree of cure z. In addition, the effective curing-dilatation coefficient β_{est} from [5] is used which is not depending on z.

Fig. 1.c illustrates the micro-RVE with three partial volumes $v^{(i)}$, i = sol, ca, r over time t. Fig. 2.a shows the effective compression modulus K for the heterogeneous matrix versus the degree

of cure z. The bounds K_R and K_V obtained from the homogeneous matrix are comparatively plotted. Fig. 2.b shows the effective heat-dilatation coefficient α versus z. The bounds for the effective heat-dilatation coefficient α_R and α_V are plotted in addition. Fig. 2.c shows the effective curing-dilatation coefficient β and the estimate for the effective curing-dilatation coefficient β_{est} versus z. As a further result, Fig. 2.d shows the chemical part of the volumetric strain. For the heterogeneous matrix, the trace of ε_{cur} in Eq. (2.3) is combined with β . The result is denoted as e_{cur} in Fig. 2.d, where β_{est} is used in e_{cur} for the homogeneous matrix.



Figure 2. Effective properties and bounds versus the degree of cure: a) compression moduli, b) heat-dilatation coefficients, c) curing-dilatation coefficients and d) volumetric curing strains.

Conclusions

We conclude, that the effective elastic and thermal properties lie within the Voigt and Reuss bounds, whilst for the chemical part of the model, an analogous result is obtained for the effective strains. This principal difference is a consequence of the assumptions for a homogeneous distribution of the temperature change and an inhomogeneous distribution of curing in the constitutive equations on the microscale.

Acknowledgements

This work is based on investigations of the "SPP 1712 - Intrinsische Hybridverbunde für Leichtbautragstrukturen", which is kindly supported by the Deutsche Forschungsgemeinschaft (DFG).

References

- [1] M.H.H. Meuwissen et al., Microelectro. Relia. 44, 1985–1994 (2004).
- [2] R. Mahnken, Int. J. Solids Struct. 50, 2003–2021 (2013).
- [3] R.M. Christensen, Mechanics of composite materials (John Wiley and Sons, New York, 1979).
- [4] R. Mahnken, C. Dammann, Int. J. Solids Struct. 100–101, 341-355 (2016).
- [5] R. Mahnken, C. Dammann, P. Lenz, Int. J. Mult. Com. Eng., accepted (2017).
- [6] E. Herve, A. Zaoui, Int. J. Eng. Sc. **31** 1–10 (1993).