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A micromechanical analysis of a local failure criterion for particle-reinforced composites

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Abstract

The present paper aims at predicting the ultimate mechanical properties of a particle-reinforced polymer using a micromechanical approach of a local failure criterion. The considered criterion includes both normal and shear stresses at the interface between the polymer matrix and the reinforcing material. In the case of rigid particles, a new closed-form expression of the interfacial stress concentration tensor is provided and simple analytical formula are proposed and compared for different homogenization schemes. A combination of these results with acoustic emission data allows the identification of the parameters involved in the local failure criterion. It has been shown that the predicted interfacial strength highly depends on the choice of a suitable homogenization scheme.

Key words: Particle-reinforced composites, Interfacial strength, Mechanical properties, Acoustic emission, Stress concentrations

1 Introduction

Polymer composites have been used for many years in several applications such as aircraft, space and marine structures, mass transport systems, automotive

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industries, building and construction. The experience gained in all these applications showed that the in-service behavior of a polymer composite highly depends on the properties of the reinforcing particle-matrix or endless fibre-matrix interface that contributes to stress transfer. The reinforcing particle can be a filler or a chopped fiber. This explains why the study of interfaces received much attention in order to understand phenomena and mechanisms involved there. Adhesion is one the most fundamental aspect addressed in this area because interfacial debonding is, with particle breakage, one of the basic local failure modes in particle-reinforced composites. The study of such local failure mechanism classically requires the use of a stress-based local approach. A suitable analysis can be performed by using micromechanical approaches which allow to relate the microscopic (local) stress field to the macroscopic stress tensor. The determination of the local stress field, particularly at the particle-matrix interface, depends on both the material microstructure and the choice of a suitable homogenization scheme. In this paper, we investigate the local criterion considered by Fitoussi et al. [2] and derive a new explicit formulation in the case of rigid particles, different homogenization schemes being considered and compared for this purpose. Moreover, the closed-form results are combined with acoustic emission data in order to evaluate the mechanical parameters involved in the criterion.

2 Local debonding criterion and micromechanical analysis

2.1 Local failure criterion

Let us consider a local failure criterion based on the stress field at the interface between the matrix and the reinforcing material. As previously used by Fitoussi et al. [2], an example of this type of criterion consists in a linear combination of the local normal and shear stresses and is defined as follows:

$$\sigma_n^{out} + \beta \tau_n^{out} = R_{int} \quad (1)$$

where σ_n^{out} and τ_n^{out} are the interfacial normal and shear stresses respectively; β and R_{int} are two parameters to be identified by an iterative regression between both experimental and simulated results (see [2]). The local normal and shear stresses are classically defined as:

$$\begin{cases} \sigma_n^{out} = \underline{n}^T \cdot \underline{\sigma}^{out} \cdot \underline{n} \\ \tau_n^{out} = \sqrt{\|\underline{\sigma}^{out} \cdot \underline{n}\|^2 - (\underline{n}^T \cdot \underline{\sigma}^{out} \cdot \underline{n})^2} \end{cases} \quad (2)$$

where $\underline{\sigma}^{out}$ is the interfacial stress field and $\underline{n}$ is the outward unit normal to the boundary on the interface point under consideration.

2.2 Micromechanical formulation

Requiring the displacement and the interfacial traction across the boundary to be continuous, the components of the stress tensor just outside the inhomogeneity are found to be (see for instance [1]):

$$\sigma_{ij}^{out} = \frac{1}{D(\underline{n})} \mathbb{C}_{ijkl}^1 \left[\mathbb{S}_{klab}^2 + \mathbb{N}_{kp}(\underline{n}) n_l n_q \Delta \mathbb{C}_{pqmn} \mathbb{S}_{mnab}^2 \right] \sigma_{ab}^{2in} \quad (3)$$

where σ^{out} is the stress tensor at the interface, $\mathbb{C}^1$ is the stiffness tensor of the matrix; $\mathbb{C}^2$ and $\mathbb{S}^2$ are the stiffness and compliance tensors of the inhomogeneity respectively. $\Delta \mathbb{C}$ is defined by $\Delta \mathbb{C} = \mathbb{C}^2 - \mathbb{C}^1$, σ^{2in} is the stress tensor inside the inhomogeneity, $\mathbb{N}_{kp}$ and $D(\underline{n})$ are the cofactor and determinant, respectively, of the acoustic tensor $\mathbb{K}$ associated with $\mathbb{C}^1$ and the direction $\underline{n}$:

$$\mathbb{K}_{ik} = \mathbb{C}_{ijkl}^1 n_j n_l \quad (4)$$

$$D(\underline{n}) = \epsilon_{mnl} \mathbb{K}_{m1} \mathbb{K}_{n2} \mathbb{K}_{l3} \quad (5)$$

$$\mathbb{N}_{ij} = \frac{1}{2} \epsilon_{ikl} \epsilon_{jmn} \mathbb{K}_{km} \mathbb{K}_{ln} \quad (6)$$

where ϵ_{ijk} are the components of the permutation tensor defined by:

$$\epsilon_{ijk} = \begin{cases} -1 & \text{for the even permutation of 1,2,3} \\ 1 & \text{for the odd permutation of 1,2,3} \\ 0 & \text{for other cases} \end{cases} \quad (7)$$

A starting point of the micromechanical analysis consists in relating the microscopic and macroscopic stresses. Due to the linearity of this homogenization problem, the stress tensor σ^{2in} inside the inhomogeneity is given by the stress concentration relation:

$$\sigma^{2in} = \mathbb{B}^2 : \Sigma \quad (8)$$

where $\mathbb{B}^2$ is the stress concentration tensor for the inhomogeneity embedded in the solid matrix and Σ is the macroscopic stress tensor prescribed at the boundary of the representative elementary volume.

2.3 Case of a matrix reinforced by infinitely rigid inclusions

In order to derive closed-form expressions, we consider the case of rigid reinforcements (i.e. $\mathbb{S}^2 \rightarrow 0$); we then rewrite the term $\Delta \mathbb{C}_{pqmn} \mathbb{S}_{mnab}^2$ as:

$$\Delta \mathbb{C}_{pqmn} \mathbb{S}_{mnab}^2 = (\mathbb{C}^2 - \mathbb{C}^1)_{pqmn} \mathbb{S}_{mnab}^2 = \mathbb{I}_{pqab} - (\mathbb{C}^1 : \mathbb{S}^2)_{pqab} \approx \mathbb{I}_{pqab} \quad (9)$$

where $\mathbb{I}$ is the symmetric fourth-order identity tensor defined by $\mathbb{I}_{pqab} = \frac{1}{2}(\delta_{pa}\delta_{qb} + \delta_{pb}\delta_{qa})$, δ is the Kronecker delta ($\delta_{ij} = 1$ for $i = j$ and $\delta_{ij} = 0$ for $i \neq j$). Taking into account (9), we have:

$$\mathbb{N}_{kp}(\underline{n}) n_l n_q \Delta \mathbb{C}_{pqmn} \mathbb{S}_{mnab}^2 = \mathbb{N}_{kp}(\underline{n}) n_l n_q \mathbb{I}_{pqab} \quad (10)$$

which also reads

$$\mathbb{N}_{kp}(\underline{n}) n_l n_q \Delta \mathbb{C}_{pqmn} \mathbb{S}_{mnab}^2 = \frac{1}{2} \{ \mathbb{N}_{ka}(\underline{n}) n_l n_b + \mathbb{N}_{kb}(\underline{n}) n_l n_a \} = \mathbb{N}(\underline{n}) \underline{\otimes} (\underline{n} \otimes \underline{n}) \quad (11)$$

where $(A \underline{\otimes} B)_{klab} = \frac{1}{2} [A_{ka} B_{lb} + A_{kb} B_{la}]$ for any second order tensors $\mathbf{A}$ and $\mathbf{B}$.

Then, equation (3) can be rewritten in a more readily and compact tensorial form:

$$\boldsymbol{\sigma}^{out} = \frac{1}{D(\underline{n})} \mathbb{C}^1 : [\mathbb{N}(\underline{n}) \underline{\otimes} (\underline{n} \otimes \underline{n})] : \boldsymbol{\sigma}^{2in} \quad (12)$$

Let us recall now that for different homogenization schemes, the stress concentration tensor which enters (8) is classically given for the inhomogeneity by (see for instance [8]):

$$\mathbb{B}^2 = \mathbb{B}_i^2 : \left(\sum_{k=1}^2 f^r \mathbb{B}_i^r \right)^{-1} \quad (13)$$

where f^r is the volume fraction of the phase r (the superscripts 0, 1 and 2 denote some reference medium, the matrix and the inhomogeneity respectively) and $\mathbb{B}_i^r$ is defined by:

$$\mathbb{B}_i^r = [\mathbb{I} + \mathbb{Q}^0 : (\mathbb{S}^r - \mathbb{S}^0)]^{-1} \quad (14)$$

where $\mathbb{S}^r$ is the compliance tensor of the phase r and $\mathbb{Q}^0$ is defined by :

$$\mathbb{Q}^0 = \mathbb{C}^0 - \mathbb{C}^0 : \mathbb{P}_0^2 : \mathbb{C}^0 \quad (15)$$

$\mathbb{P}_0^2$ is the Hill tensor (see [11]). Since $\mathbb{C}^0 : \mathbb{S}^0 = \mathbb{I}$ and $\mathbb{S}^2 \rightarrow 0$, one easily gets:

$$\begin{cases} \mathbb{B}_i^1 = [\mathbb{I} + \mathbb{Q}^0 : (\mathbb{S}^1 - \mathbb{S}^0)]^{-1} \\ \mathbb{B}_i^2 \approx (\mathbb{C}^0 : \mathbb{P}_0^2)^{-1} \end{cases} \quad (16)$$

Substituting (16) into (13) and recalling that $(\mathbb{A} : \mathbb{B})^{-1} = \mathbb{B}^{-1} : \mathbb{A}^{-1}$ ($\mathbb{A}$ and $\mathbb{B}$ being any symmetric fourth-order tensor, i.e. $\mathbb{A}_{ijkl} = \mathbb{A}_{jikl} = \mathbb{A}_{ijlk} = \mathbb{A}_{klij}$)

and $f^1 = 1 - f^2$, we obtain for the two phase composites:

$$\mathbb{B}^2 = \mathbb{B}_i^2 : [(1 - f^2) \mathbb{B}_i^1 + f^2 \mathbb{B}_i^2]^{-1} \approx [(1 - f^2) [\mathbb{I} + \mathbb{Q}^0 : (\mathbb{S}^1 - \mathbb{S}^0)]^{-1} : \mathbb{C}^0 : \mathbb{P}_0^2 + f^2 \mathbb{I}]^{-1} \quad (17)$$

and then, we finally define a new tensor,

$$\mathbb{B}^{out} = \frac{1}{D(\underline{n})} \mathbb{C}^1 : [\mathbb{N}(\underline{n}) \underline{\otimes} (\underline{n} \otimes \underline{n})] : [(1 - f^2) [\mathbb{I} + \mathbb{Q}^0 : (\mathbb{S}^1 - \mathbb{S}^0)]^{-1} : \mathbb{C}^0 : \mathbb{P}_0^2 + f^2 \mathbb{I}]^{-1} \quad (18)$$

such as:

$$\boldsymbol{\sigma}^{out} = \mathbb{B}^{out} : \boldsymbol{\Sigma} \quad (19)$$

$\mathbb{B}^{out}$ can be considered, for rigid inhomogeneities, as a concentration tensor for the interfacial stress field: it is readily seen that this new tensor $\mathbb{B}^{out}$ depends on both the geometry of the inhomogeneity (via the tensor $\mathbb{P}_0^2$) and the properties of the reference medium denoted by the superscript 0. Therefore, note that the expression of $\mathbb{B}^{out}$ clearly depends on the homogenization scheme (see section 2.4).

2.4 Role of the homogenization schemes

Different homogenization schemes are going to be considered in the following. For the dilute estimate (also known as the Eshelby's solution, see for instance [4]), classically devoted to non interacting inhomogeneities (at dilute concentration), the matrix is considered as the reference medium (i.e. $\mathbb{S}^0 = \mathbb{S}^1$) and is subjected to the macroscopic stress tensor. The validity of this approach corresponds to $f^2 \rightarrow 0$ and thus to, $\mathbb{B}^2 \rightarrow (\mathbb{C}^1 : \mathbb{P}_0^2)^{-1}$. We recall that in general, $\mathbb{P}_0^2 = \mathbb{S}_{ESH} : (\mathbb{C}^0)^{-1}$, where $\mathbb{S}_{ESH}$ is the Eshelby tensor which depends on both the geometry of the inhomogeneity and the reference medium (see [1]). Therefore, eqn (18) yields:

$$\mathbb{B}_{ESH}^{out} = \frac{1}{D(\underline{n})} \mathbb{C}^1 : [\mathbb{N}(\underline{n}) \underline{\otimes} (\underline{n} \otimes \underline{n})] : (\mathbb{C}^1 : \mathbb{P}_1^2)^{-1} \quad (20)$$

where obviously, $\mathbb{P}_1^2 = (\mathbb{P}_0^2)_{\text{Dilute estimate}} = \mathbb{S}_{ESH} : (\mathbb{C}^1)^{-1}$.

For the Mori-Tanaka estimate [10] (see also [5]), which allows to take into account the interactions between the inhomogeneities, the solid matrix is still considered as the reference medium, but is now subjected to its own stress. It

follows that:

$$\mathbb{B}_{MT}^{out} = \frac{1}{D(\underline{n})} \mathbb{C}^1 : [\mathbb{N}(\underline{n}) \underline{\otimes} (\underline{n} \otimes \underline{n})] : \left[(1 - f^2) \mathbb{C}^1 : \mathbb{P}_1^2 + f^2 \mathbb{I} \right]^{-1} \quad (21)$$

In the Generalized Self-Consistent scheme [6], the reference medium is the unknown effective medium denoted by SC , that is $\mathbb{S}^0 = \mathbb{S}^{SC}$. Then, eqn (18) becomes:

$$\mathbb{B}_{SC}^{out} = \frac{1}{D(\underline{n})} \mathbb{C}^1 : [\mathbb{N}(\underline{n}) \underline{\otimes} (\underline{n} \otimes \underline{n})] : \left[(1 - f^2) [\mathbb{I} + \mathbb{Q}^{SC} : (\mathbb{S}^1 - \mathbb{S}^{SC})]^{-1} : \mathbb{C}^{SC} : \mathbb{P}_{SC}^2 + f^2 \mathbb{I} \right]^{-1} \quad (22)$$

Finally, the Ponte-Castaneda and Willis estimate [7] aims at taking into account not only the interaction effect, but also the spatial distribution of the reinforcing material by means of a new tensor $\mathbb{P}_d$ (see [7] for a detailed presentation as well as the expression of $\mathbb{P}_d$). In this approach, the tensor $\mathbb{P}_1^2$ is then replaced by $\frac{1}{1-f^2} [\mathbb{P}_1^2 - f^2 \mathbb{P}_d]$ and we get:

$$\mathbb{B}_{PCW}^{out} = \frac{1}{D(\underline{n})} \mathbb{C}^1 : [\mathbb{N}(\underline{n}) \underline{\otimes} (\underline{n} \otimes \underline{n})] : \left(\mathbb{C}^1 : [\mathbb{P}_1^2 - f^2 \mathbb{P}_d] + f^2 \mathbb{I} \right)^{-1} \quad (23)$$

3 Case of a polymer-matrix composite reinforced by spherical inhomogeneities: a comparison between micromechanical schemes

3.1 Introduction

In this section, we consider an isotropic unsaturated polyester matrix reinforced by E-Glass spherical beads (volume fraction : 30 %). Our aim here is to determine the sensitivities of both the parameter R_{int} and the local stress field to the homogenization scheme. The spherical particles have been burnt five hours at 520° C to remove sizing in order to promote interfacial debonding instead of matrix cracking. Fig. (1) shows a SEM inspection of the composite material. The lack of polymer matrix at the particle surface, i.e. the particle surface is smooth, gives the evidence that interfacial debonding is the damage mechanism.

The values of the elastic moduli and Poisson's ratios for each phase (the superscripts 1 and 2 correspond to the matrix and the inhomogeneity respectively)

Figure 1. SEM micrograph of the glass beads-filled unsaturated polyester resin

are:

$$\begin{cases} E^1 = 2.7 [GPa] \\ \nu^1 = 0.37 \\ E^2 = 73 [GPa] \\ \nu^2 = 0.22 \end{cases} \quad (24)$$

The consideration of these data has basically motivated the assumption of rigid particles.

For spherical inhomogeneities, the Hill tensor $\mathbb{P}_1^2$ is given by (see [11] or [1]):

$$\mathbb{P}_1^2 = \frac{\alpha^1}{3k^1} \mathbb{J} + \frac{\beta^1}{2\mu^1} \mathbb{K} \quad (25)$$

with

$$\alpha^1 = \frac{3k^1}{3k^1 + 4\mu^1}; \quad \beta^1 = \frac{6(k^1 + 2\mu^1)}{5(3k^1 + 4\mu^1)} \quad (26)$$

k^1 and μ^1 are the bulk and shear moduli of the matrix respectively. $\mathbb{J}$ and $\mathbb{K}$ are the classical symmetric fourth-order tensors defined by $\mathbb{J} = \frac{1}{3} \mathbf{i} \otimes \mathbf{i}$ and $\mathbb{K} = \mathbb{I} - \mathbb{J}$, $\mathbf{i}$ is the second-order symmetric identity tensor : $i_{ij} = \delta_{ij}$. Due to the isotropy of the constitutive phases, the stiffness tensor for the phase r can be written as:

$$\mathbb{C}^r = 3k^r \mathbb{J} + 2\mu^r \mathbb{K} \quad (27)$$

and then, the compliance tensor reads:

$$\mathbb{S}^r = [\mathbb{C}^r]^{-1} = \frac{1}{3k^r} \mathbb{J} + \frac{1}{2\mu^r} \mathbb{K} \quad (28)$$

In the case of the SC scheme, the Hill tensor $\mathbb{P}_{SC}^2$ is given by:

$$\mathbb{P}_{SC}^2 = \mathbb{S}_{ESH} : (\mathbb{C}^{SC})^{-1} = \frac{\alpha^{SC}}{3k^{SC}} \mathbb{J} + \frac{\beta^{SC}}{2\mu^{SC}} \mathbb{K} \quad (29)$$

for which we recall that k^{SC} and μ^{SC} are the effective bulk and shear moduli of the composite respectively. α^{SC} and β^{SC} are obtained from (26) in which k^1 and μ^1 must be replaced by k^{SC} and μ^{SC} respectively. Moreover, experimental results in the case of a tensile test suggest that the debonding does appear at the pole of the spherical particle, as illustrated on Fig. (2).

Then, we note that:

Figure 2. Interfacial debonding during a tensile test

$$\sigma_{n, \text{pole}}^{\text{out}} = \text{Max} \left(\sigma_n^{\text{out}} \right), \tau_{n, \text{pole}}^{\text{out}} = 0 \quad (30)$$

and recalling eqn (1), we get:

$$R_{int} = \sigma_{n, \text{pole}}^{\text{out}} \quad (31)$$

It follows that R_{int} can be determined by a pure macroscopic uniaxial tensile test coupled with an acoustic emission measure, as the debonding macroscopic force F_{deb} (and so, the interfacial failure) can be determined by comparing the acoustic emission (AE) results (that are the counts and the amplitude - in dB - vs. time - in seconds -) with the tensile test results (the force - in N - vs. time); see section (3.3). We recall here that AE is widely used for non destructive control as well as to identify and to monitor damage mechanisms and evolution (see for instance [9] or [3]). In fact, each failure mode can be characterized by a range of amplitude; the debonding phenomena basically corresponds to the range [58...68] dB. Thus, we define the macroscopic tensile stress tensor Σ (which is used for the micromechanical analysis) by :

$$\Sigma = \text{Diag} [0, 0, \Sigma_{33}] \quad (32)$$

3.2 Sensitivity of the local stress field

Due to the geometrical and material symmetries of the reinforcing inhomogeneities, the local normal and shear stresses are characterized by using a parametrization of the unit normal $\underline{n}$, that is (see Fig. (3)):

$$\underline{n} = [\cos(\alpha), 0, \sin(\alpha)]^T, \alpha \in \left(0, \frac{\pi}{2}\right) \quad (33)$$

Figure 3. Parametrization of the unit normal

Without any loss of generality, we consider an unit macroscopic tensile stress tensor $\underline{\Sigma} = \text{Diag}[0, 0, 1]$ and the local normal and shear stress fields are computed, for the different homogenization schemes, by using eqns (20), (21), (22) or (23) combined with eqn (19). The results are shown on Fig. (4) and (5).

The following remarks can be done:

- For both normal and shear stresses: the different homogenization schemes provide the same profiles but different amplitudes; moreover, we note that the local normal and shear stresses are not maxima at the same interfacial point, which means that the failure can occur at points located between the two points defining the maximum of the normal and shear stresses respectively. The biggest difference between the estimates is observed at the point of maximal stress, that is for instance $\alpha = \frac{\pi}{2}$ for the normal component.
- For the local normal stress: as expected, the component is minimal and null at the equator of the spherical inhomogeneity, while reaching its maximum at the pole. As α increases from 0 to $\frac{\pi}{2}$, the local normal stress starts to decrease from 0 to its minimal value, and thus defines a local compression; the component then starts increasing from its minimal to its maximal positive value obtained for $\alpha = \frac{\pi}{2}$ (change from a local compression to a local traction).
- For the local shear stress: the shear component is positive and null at both the equator and the pole of the reinforcing inhomogeneity.

The maximum relative errors ϵ [%] between the estimates are also computed. Note that these errors for the local normal and shear stresses are exactly the same, as the use of different homogenization schemes basically results

Figure 4. Distribution of the local normal stress

in different predictions for the interfacial stress field which is used for the determination of both local normal and shear stresses. The results are reported in table (1).

	Dilute	Mori-Tanaka	SC	PCW $w_d = 0.8$	PCW $w_d = 1.2$
Dilute	0 [%]	22.6 [%]	6.9 [%]	20.1 [%]	24.6 [%]
Mori-Tanaka		0 [%]	16.8 [%]	3.1 [%]	2.6 [%]
SC			0 [%]	14.2 [%]	19 [%]
PCW $w_d := 0.8$				0 [%]	5.6 [%]
PCW $w_d := 1.2$					0 [%]

Table 1

Maximum relative errors for the different homogenization schemes

3.3 Sensitivity of the parameter R_{int}

For application purposes, the tensile test has been carried out on a Instron tensile machine and coupled with an acoustic emission system (threshold: 30 dB). Eight tensile tests were carried out (see Fig. (6)) and a mean value for

Figure 5. Distribution of the local shear stress

F_{deb} was obtained:

$$F_{\text{deb}} \approx 39.42 \text{ [daN]} \quad (34)$$

Given a mean section of $S = 37.84 \text{ [mm}^2\text{]}$, we find the uniaxial debonding macroscopic stress as:

$$\Sigma_{\text{deb}} = \frac{F_{\text{deb}}}{S} \approx 10.42 \text{ [MPa]} \quad (35)$$

Substituting $\Sigma_{33} = \Sigma_{\text{deb}}$ in eqns (20), (21), (22) and (23) and using eqn (19), we finally derive the estimates of R_{int} for the different homogenization schemes under consideration (see table (2)).

Homogenization Scheme	Estimate of R_{int}
Dilute	20.06 [MPa]
Mori-Tanaka	15.53 [MPa]
SC	18.67 [MPa]
PCW $w_d = 0.8$	16.03 [MPa]
PCW $w_d = 1.2$	15.13 [MPa]

Table 2
Estimates of R_{int}

Figure 6. AE spectra (amplitudes and counts vs. time) used for the identification of the debonding macroscopic force

It is seen that the predictions for the mechanical parameter R_{int} are very different depending on the micromechanical formulation under consideration: a maximum relative error of 24.6[%] is for instance observed between the Ponte-Castaneda and Willis ($w_d = 1.2$) estimate and the Eshelby's solution which seems to overestimate the interfacial strength.

4 Concluding remarks

The role of the homogenization scheme in the analysis of a local failure criterion has been investigated. In particular, a new closed-form expression of the interfacial stress concentration tensor has been proposed in the case of rigid particle-reinforced composites. The proposed methodology can be easily applied to a more general class of materials (for instance thermoplastics reinforced with short or long fibres). The coupling of such a micromechanical analysis with acoustic emission data allowed us to determine the interfacial strength whose sensitivity to the homogenization scheme is emphasized. Thus, it is observed that the predicted value of the interfacial strength (and in gen-

eral, of the effective mechanical properties) strongly depends on the homogenization scheme which in practice has to be carefully chosen according to the real morphology of the material.

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