MICROMECHANICAL MODELING OF THE THERMO-ELASTO-VISCOPLASTIC BEHAVIOR OF SEMI-CRYSTALLINE POLYMERS

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Abstract. A micromechanically-based constitutive model for the elasto-viscoplastic deformation and texture evolution of semi-crystalline polymers subjected to large strains is developed. The material is represented by an aggregate of two-phase layered composite inclusions. Each composite consists of a crystalline lamella, which is assumed to plastically deform by crystallographic slip, and an amorphous layer. The constitutive behavior of each phase is completed with elasticity and thermal expansion. Interface compatibility within the composite inclusion is enforced on the deformation gradients of the respective phases. Moreover, traction equilibrium is enforced across the interface. The volume-averaged deformation and stress within the inclusions are related to the macroscopic fields by either a Taylor-type or a Sachs-type interaction model. Both models are applied to initially isotropic polyethylene in uniaxial compression.
1 INTRODUCTION

Due to an increasing use of polymer materials for structural and electronic applications, there is a strong demand for improved mechanical properties of these materials. Since these macroscopic properties are strongly influenced by the microstructural morphology and deformation mechanisms, the need for a better understanding of the mechanical behavior at this microstructural level is evident. During processing, polymeric materials are often subjected to large plastic deformations, giving rise to preferential orientation of macromolecules and morphology, which may result in high anisotropy. Furthermore, in thin films, preferred orientation of the crystalline component, produced by preferential crystallization, can give rise to strong anisotropy and therefore can have a profound influence on the mechanical properties of these films. This anisotropy is a general phenomenon that plays a significant role in various thin film technologies which are widely used in, for example, the micro-electronics industry. Similar conditions, namely a thin layer of preferentially oriented material, appear in the microstructural morphology of particle-toughened materials, which are studied by finite element modeling Smit et al. [1] and Socrate and Boyce [2] for amorphous polymers and in Tzika et al. [3] for semi-crystalline polymers. In the Tzika et al. work, the important effects of initial matrix anisotropy on the micromechanics of deformation of the composite material are taken into account; however, the evolution of the anisotropic properties with deformation by mechanisms of macromolecular texturing are not considered. In the present work, a micromechanically-based constitutive model for the thermo-elasto-viscoplastic deformation of semi-crystalline polymers is developed. The model captures the effect of the evolution of texture with large deformation on mechanical properties including stiffness, strength and thermal expansion coefficients.

Viscoplastic Taylor-type models have been used by, for example, Parks and Ahzi [4] and Lee et al. [5] for the prediction of texture evolution for polycrystalline polymer materials. In these studies, the influence of the amorphous phase is neglected. However, for many semi-crystalline materials, the contribution of the amorphous phase can be rather substantial. For example the typical crystallinities of HDPE and Nylon-6 are 70 % and 40 %, respectively. To account for this amorphous phase, Lee et al. [6][7] have used a rigid/viscoplastic composite inclusion model. In this model, the material is represented by an aggregate of two-phase layered composite inclusions. Within each inclusion, the two phases are connected by interface compatibility and traction equilibrium.Because of the molecular chain inextensibility within the crystalline phase, in combination with interface compatibility, if the Taylor interaction law is used to prescribe the inclusion-averaged deformation in the rigid/viscoplastic model, the inclusion tends to lock for certain crystal orientations. Therefore, they proposed a Sachs-type interaction model to relate the volume-averaged deformation and stress within the inclusion to the macroscopic fields.

The objective of the present work is to numerically investigate the micromechanical mechanisms that influence the macroscopic material behavior. For this purpose, a micromechanically-based numerical model for the large plastic deformation and texture evolution of semi-crystalline polymers is developed. Following Lee et al. [6][7], a layered two-phase composite inclusion
model is formulated. Each composite consists of a crystalline lamella which is assumed to plastically deform by crystallographic slip and an amorphous layer for which a power law constitutive relation is used in conjunction with a back stress tensor to account for orientation-induced hardening [8]. In contrast to the rigid/viscoplastic composite inclusion model of Lee et al. [6][7], the constitutive behavior of each phase is completed with elasticity and thermal expansion. Interface compatibility within the composite inclusion is enforced on the deformation gradients of the respective phases, which are each assumed to deform homogeneously. Moreover, traction equilibrium is enforced across the interface. Both a Taylor-type and a Sachs-type interaction model are used to relate the local inclusion-averaged deformation and stress to the macroscopic fields of the aggregate. The model may be used to study the influence of microstructural morphology and deformation mechanisms on the macroscopic deformation resistances for preferentially oriented materials, and in particular the matrix deformation in particle-toughened semi-crystalline materials.

2 MATERIAL MODELS

In the present work, the mechanical behavior of semi-crystalline polymers is modeled by an aggregate of two-phase composite inclusions. Each inclusion consists of a crystalline and an amorphous phase. In this section, the kinematical framework, and the material models that are used to describe the constitutive behavior of each respective phase are discussed. In the following, let the superscripts “a” and “c” denote the amorphous layer and the crystalline lamella, respectively.

2.1 Kinematics

For a microstructural thermo-elasto-viscoplastic constitutive description of both the crystalline and the amorphous phase, the deformation gradient tensor $F = (V_0 x)^T$ of each constituent phase, where the superscript “T” denotes the transpose, is decomposed into a plastic, a thermal and an elastic component, denoted by the subscripts “p”, “th” and “e”, respectively [9]:

$$F^{\pi} = F_e^{\pi} \cdot F_{th}^{\pi} \cdot F_p^{\pi}; \quad \pi = a, c.$$  \hspace{1cm} (1)

This multiplicative decomposition considers an intermediate stress-free state $\Omega(\theta)$, at temperature $\theta$, which results from isothermally unloading the current configuration $\Omega(\theta)$, as is schematically illustrated in figure 1. Thereafter, a second intermediate configuration $\Omega(\theta_0)$ is obtained via $F_{th}^{\tau^{-1}}$, whereby the temperature changes from $\theta$ to the reference temperature $\theta_0$. Both $F_{th}^{\tau}$ and $F_p^{\tau}$ are invariant, i.e. upon a superimposed rigid body motion

$$x \in \Omega(\theta) \mapsto x^+ = v + Q \cdot x,$$  \hspace{1cm} (2)

with $v$ a rigid body translation and $Q$ an orthogonal mapping, the respective deformation gradients are transformed to:

$$F_e^{\tau^+} = Q \cdot F_e^{\tau}; \quad F_{th}^{\tau^+} = F_{th}^{\tau} \quad \text{and} \quad F_p^{\tau^+} = F_p^{\tau}$$  \hspace{1cm} (3)
The velocity gradient tensor $\mathbf{L} = \dot{\mathbf{F}} \cdot \mathbf{F}^{-1}$ corresponding to this formulation is given by

$$\mathbf{L} = \mathbf{L}_e + \mathbf{F}_e \cdot \mathbf{L}_\text{th} \cdot \mathbf{F}_\text{th}^{-1} + \mathbf{F}_\text{th} \cdot \mathbf{L}_p \cdot \mathbf{F}_\text{th}^{-1} \cdot \mathbf{F}_e^{-1},$$

(4)

where the superscript $\pi$, denoting either the amorphous or the crystalline phase, is omitted. The elastic, the thermal and the plastic velocity gradients are defined as

$$\mathbf{L}_e = \dot{\mathbf{F}}_e \cdot \mathbf{F}_e^{-1} ; \quad \mathbf{L}_\text{th} = \dot{\mathbf{F}}_\text{th} \cdot \mathbf{F}_\text{th}^{-1} \quad \text{and} \quad \mathbf{L}_p = \dot{\mathbf{F}}_p \cdot \mathbf{F}_p^{-1},$$

(5)

respectively.

2.2 Constitutive relations

The elastic component of deformation in both the amorphous and the crystalline phase is characterized by a fourth-order elastic modulus tensor $4\mathbf{C}$ which constitutes a linear relation between the elastic Green Lagrange strain tensor $\mathbf{E}_\pi$ and the stress measure $\mathbf{\tau}$:

$$\mathbf{\tau}_\pi = 4\mathbf{C}_\pi : \mathbf{E}_\pi^\pi ; \quad \pi = a, c,$$

(6)

where the stress measure $\mathbf{\tau}$ is defined as

$$\mathbf{\tau}_\pi = J_e^{\pi} \mathbf{F}_e^{\pi-1} \cdot \mathbf{\sigma}_\pi \cdot \mathbf{F}_e^{\pi-T} ; \quad \pi = a, c,$$

(7)

with $J_e = \det(\mathbf{F}_e)$ and $\mathbf{\sigma}$ the Cauchy stress tensor. Furthermore, the Green Lagrange strain tensor is given by

$$\mathbf{E}_\pi = \frac{1}{2} \left( \mathbf{F}_e^{\pi-T} \cdot \mathbf{F}_e^{\pi} - \mathbf{I} \right) ; \quad \pi = a, c,$$

(8)

where $\mathbf{I}$ denotes the second-order identity tensor. The elastic deformation of the amorphous phase is assumed to be isotropic and to remain isotropic with deformation, i.e. characterized by the Young’s modulus $E$ and Poisson’s ratio $\nu$, whereas for the crystalline phase an anisotropic elasticity tensor is assumed.
2.2.1 Amorphous phase

The thermally-induced deformation of the amorphous phase is assumed to be isotropic and is characterized by the thermal expansion coefficient $\alpha^a$, which may be temperature dependent:

$$ F_{th}^a = \exp \left( \alpha^a [\theta - \theta_0] \right) I. $$  

Consequently, the thermally induced deformation rate tensor $D_{th}^a$ is given by

$$ D_{th}^a = L_{th}^a = C_{\alpha}^a(\theta) I \dot{\theta}, \quad \text{with} \quad C_{\alpha}^a(\theta) = \frac{d\alpha^a}{d\theta} [\theta - \theta_0] + \alpha^a. $$

The velocity gradient in the $\Omega(\theta)$ configuration, as illustrated in figure 1, can be written as

$$ L^a = \tilde{D}^a + \tilde{\Omega}^a $$

$$ = L_{th}^a + F_{th}^a \cdot L_p^a \cdot F_{th}^{-1}, $$

with $\tilde{\Omega}^a$ the skew-symmetric spin tensor. By choosing $\tilde{\Omega}^a = 0$, the deformation rate tensor $\tilde{D}^a$ in this configuration is then given by [9]

$$ \tilde{D}^a = C_{\alpha}^a(\theta) I \dot{\theta} + \tilde{D}_p^a. $$

Then, the plastic deformation rate $\tilde{D}_p^a$ remains to be constitutively prescribed.

At room temperature, the amorphous phase of HDPE, with glass transition temperature $\theta_g$ near $-70^\circ C$, is in the rubbery regime. To account for the limited strain-rate sensitivity of the material under consideration, a relatively strain-rate insensitive power law relation between an effective shear strain-rate and the effective shear stress [6] is used:

$$ \dot{\gamma}_p^a = \dot{\gamma}_0^a \left( \frac{\tau^a}{\tau_0^a} \right)^{n^a}, $$

where $\dot{\gamma}_0^a$ is a reference shear rate and $n^a$ is the rate exponent. The effective shear stress $\tau^a$ is given by

$$ \tau^a = \sqrt{\frac{1}{2} S_s^a : S_s^a}, $$

where $S_s^a$ represents the deviatoric part of the driving stress tensor $\sigma_s^a$ expressed in $\Omega(\theta)$, which is defined as [9]

$$ \sigma_s^a = R_e^a \cdot \sigma^a \cdot R_e^{-1} - H^a, $$

where $R_e^a$ is the rotation tensor obtained from the right polar decomposition of $F_e^a$. Furthermore, $H^a$ is a back stress tensor which accounts for orientation induced strain hardening [8]. The plastic stretching $\tilde{D}_p^a$ is then given by

$$ \tilde{D}_p^a = \frac{\dot{\gamma}_p^a}{\tau^a} S_s^a. $$
Using the Arruda and Boyce [10] eight-chain network model of rubber elasticity, the back stress tensor is expressed as

\[ \mathbf{H}^a = \mu_R \frac{\sqrt{N}}{\lambda_{\text{ch}}} L^{-1} \left( \frac{\lambda_{\text{ch}}}{\sqrt{N}} \right) \left( \mathbf{B}_p^a - \lambda_{\text{ch}}^2 \mathbf{I} \right), \tag{17} \]

where \( N \) represents the number of rigid links between entanglements and \( \mu_R \) is proportional to the initial stiffness. Furthermore, \( \mathbf{B}_p^a = \mathbf{F}_p^a \mathbf{F}_p^a \mathbf{T} \) is the left plastic Cauchy-Green deformation tensor and \( \lambda_{\text{ch}} \equiv \sqrt{\frac{1}{3} \text{tr}(\mathbf{B}_p^a)} \) is the stretch of each chain in the model eight chain network. Finally, \( L \) is the Langevin function defined by

\[ L(\beta) = \coth(\beta) - \frac{1}{\beta} ; \quad \beta = \frac{1}{L^{-1}} \left( \frac{\lambda_{\text{ch}}}{\sqrt{N}} \right). \tag{18} \]

The symbolic inverse function \( L^{-1} \) provides a strong increase in back stress as the chain stretch approaches the limiting extensibility: \( \lambda_{\text{ch}} \rightarrow \sqrt{N} \).

### 2.2.2 Crystalline phase

The temperature dependence of the crystallographic lattice may be strongly anisotropic. For example, the \( a \)-axis in the crystalline orthorhombic unit cell of polyethylene is found to expand significantly with temperature, whereas the dimensional change in the stiff direction of the \( c \)-axis is relatively small [11]. Therefore, for the thermally-induced deformation of the crystalline phase, a three-dimensional generalization of equation (9) is used:

\[ \mathbf{F}_{c}^\text{th} = \exp \left( \alpha_c [\theta - \theta_0] \right), \tag{19} \]

with \( \alpha^c \) the symmetric second-order tensor of thermal expansion, the principal directions of which are coupled to the principal crystallographic directions in the reference configuration.

The plastic deformation of the crystalline lamella, consisting of a single crystal, is composed of the contributions of all \( N_s \) distinct slip systems and is given by

\[ \mathbf{L}_p^c = \sum_{\beta=1}^{N_s} \dot{\gamma}^\beta \mathbf{P}_0^\beta, \tag{20} \]

where the non-symmetric Schmid tensor \( \mathbf{P}_0^\beta \), associated with the \( \beta \)-th slip system, is defined as the dyadic product of the unit slip direction \( \mathbf{s}_0^\beta \) and the unit slip plane normal \( \mathbf{n}_0^\beta \), both given in the reference configuration \( \Omega_0(\theta_0) \):

\[ \mathbf{P}_0^\beta = \mathbf{s}_0^\beta \mathbf{n}_0^\beta. \tag{21} \]

The shear rate \( \dot{\gamma}^\beta \) of each slip system is assumed to be related to the resolved shear stress \( \tau^\beta \) via the viscoplastic power law relation [12][13]

\[ \dot{\gamma}^\beta = \dot{\gamma}_0^c \left| \frac{\tau^\beta}{g^\beta} \right|^{n-1}, \tag{22} \]
where $g^\beta$ is the shear strength associated with the $\beta$th slip system, $\dot{\gamma}_0^c$ is a reference shear rate and $n^c$ is the rate exponent. The resolved shear stress is defined such that $\sum_{\beta=1}^{N_s} \tau^\beta \dot{\gamma}^\beta$ represents the rate of plastic work per volume in the $\bar{\Omega}(\theta)$ configuration:

$$\tau^\beta = \tau^c \cdot C^c \cdot P^\beta,$$

with

$$C^c = F^c \cdot T^c \cdot F^c \quad \text{and} \quad P^\beta = F^\beta_{th} \cdot P^\beta_{0} \cdot F^{c^{-1}}_{th}.$$ (24)

It is noted that a crystal can possess a maximum of five independent slip systems. Since polymer crystals are generally lacking a set of five independent slip systems, due to the plastic inextensibility of the molecular chain, the plastic deformation of these crystals is restricted to the deformation modes provided by the $N_s$ slip systems. Therefore, the plastic deformation of the crystalline phase must satisfy extra kinematic constraints besides incompressibility.

3 MICROMECHANICAL MODEL

When slowly cooled from the melt, many polymers, such as for example polyethylene, possess a polycrystalline structure. These semi-crystalline materials often show a spherulitic morphology [14]–[16]. Each spherulite consists of a radial assembly of thin crystalline lamellae which are separated by amorphous layers. The morphology of this initially isotropic material is further discussed in section 5. Furthermore, in thin films crystallized on a substrate, a sheaf-like morphology with preferentially-oriented crystalline lamellae is found [17]. The mechanical behavior of this microscopically heterogeneous material is modeled by an aggregate of layered two-phase composite inclusions as was proposed by Lee et al. [6][7] for rigid/viscoplastic behavior.

3.1 Composite inclusion

Each composite consists of a crystalline lamella which is mechanically coupled to its corresponding amorphous layer, as is represented in figure 2. The stress and deformation fields within each phase are assumed to be piecewise homogeneous; however, they may differ between the two coupled phases. It is assumed that the crystalline and amorphous components remain fully mechanically coupled. Interface compatibility within the composite inclusion is enforced on the deformation gradients of the respective phases. Moreover, traction equilibrium is enforced across the interface.

The volume fractions of the respective phases are given by the relative thicknesses $f^a$ and $f^c = 1 - f^a$, which, because of the elasticity and the thermal expansion, evolve as

$$f^\pi = \frac{J^\pi f^\pi_0}{J^a f^a_0 + J^c (1 - f^a_0)} ; \quad \pi = a, c.$$ (25)
with $J^\pi = \det(F^\pi)$ the volume ratio of each respective phase and $f_0^\pi$, the initial volume fraction of the considered phase. The inclusion-averaged deformation gradient $F^I$, where the superscript “I” denotes the composite inclusion, and the Cauchy stress $\sigma^I$ are given by the volume-weighted average of the respective phases:

$$F^I = f_0^a F^a + (1 - f_0^a) F^c,$$  \hfill (26)

and

$$\sigma^I = f_0^a \sigma^a + (1 - f_0^a) \sigma^c.$$  \hfill (27)

On the interface between the crystalline lamella and the amorphous layer, mechanical coupling is maintained by enforcing compatibility on the deformation gradients. Let $n^I_0 = e^I_03$ denote the unit normal vector of the interface in the reference configuration $\Omega_0(\theta_0)$, with $e^I_{0i}$ a local orthonormal vector basis. Furthermore, let $x^I_0$ denote an arbitrary vector in the plane of the interface $X^I_{01}$, spanned by $e^I_{01}$ and $e^I_{02}$. Then, the interface compatibility condition, in combination with volume averaging, can be written as

$$F^c \cdot x^I_0 = F^a \cdot x^I_0 = F^I \cdot x^I_0 \quad \forall x^I_0 \in X^I_{01}. \hfill (28)$$

Moreover, traction equilibrium on the interface is given by

$$\sigma^c \cdot n^I = \sigma^a \cdot n^I = \sigma^I \cdot n^I,$$  \hfill (29)

with $n^I$ the unit normal of the interface in the current configuration $\Omega(\theta)$.

### 3.2 Inclusion interaction law

To relate the volume-averaged mechanical behavior of each composite inclusion to the macroscopically imposed boundary conditions, a local-global interaction law is formulated. First, the Taylor-type interaction model [12][13], which is the most widely used interaction law in polycrystal plasticity, is described. This model however, may lead to unreasonably high stresses
since the crystalline phase of the polymer material under consideration is strongly anisotropic. Thereafter, a Sachs-type interaction model is discussed, in which micromechanical equilibrium is addressed in a stronger sense, in combination with a weakened measure of compatibility.

3.2.1 Taylor-inclusion model

The Taylor interaction model is based on the assumption that the local inclusion-averaged deformation equals the macroscopically imposed deformation of the aggregate of inclusions:

\[ \mathbf{F}^I_i = \bar{\mathbf{F}} ; \quad i = 1, \ldots, N_I, \]

(30)

where \( \bar{\mathbf{F}} \) represents the macroscopic deformation of the aggregate. This assumption assures local compatibility. Additionally, the macroscopic Cauchy stress tensor is assumed to be given by the volume averaged Cauchy stress of all inclusions in the aggregate:

\[ \sum_{i=1}^{N_I} f^I_i \sigma^I_i = \bar{\sigma} \quad \text{with} \quad f^I_i = \frac{1}{N_I}, \]

(31)

where \( f^I_i \) is the constant volume-fraction of the \( i \)th inclusion. Consequently, global equilibrium is provided by the global volume average, whereas local equilibrium of the inclusions is not addressed.

Since the basic element of the Taylor model, which is a layered two-phase composite inclusion, includes a crystalline phase with limited modes of plastic deformation, the Taylor assumption may lead to unreasonably high stresses, and the model becomes troublesome for some inclusions [6]. To overcome this difficulty, in the next section, an interaction law is discussed that provides a weakened measure of compatibility.

3.2.2 Sachs-inclusion model

In the Sachs interaction model, the volume-averaged Cauchy stress of each inclusion is assumed to equal the macroscopically homogeneous Cauchy stress \( \bar{\sigma} \) of the aggregate of composite inclusions:

\[ \sigma^I_i = \bar{\sigma} \quad ; \quad i = 1, \ldots, N_I, \]

(32)

which provides local equilibrium between the composite inclusions. Global compatibility is provided by the assumption that the macroscopically imposed deformation \( \bar{\mathbf{F}} \) equals the volume-average of all inclusion-averaged deformations, scaled by the volume-averaged volume-ratio \( \bar{J} \):

\[ \left( \frac{\bar{J}}{J_\Sigma} \right)^{\frac{1}{3}} \sum_{i=1}^{N_I} f^0_i \mathbf{F}^I_i = \bar{\mathbf{F}}, \quad \text{with} \quad \bar{J} = \sum_{i=1}^{N_I} f^I_i J^I_i \quad \text{and} \quad J_\Sigma = \det \left( \sum_{i=1}^{N_I} f^I_i \mathbf{F}^I_i \right) \],

(33)
with \( f_0^I = 1/N_I \) the initial volume fraction of the \( i \)th inclusion and \( J^I = \det(F^I) \). To complete the interaction model, the rotation of each inclusion, in the polar decomposition sense, is assumed to equal the macroscopically imposed rotation:

\[
\mathbf{R}^I = \bar{\mathbf{R}}; \quad i = 1, \ldots, N_I, \tag{34}
\]

where \( \mathbf{R}^I \) and \( \bar{\mathbf{R}} \) are the rotation tensors obtained from the right polar decomposition of the deformation gradients \( F^I \) and \( \bar{F} \), respectively. Then, equation (33) can be rewritten in terms of the corresponding right stretch tensors:

\[
\left( \frac{\bar{J}}{J_{\Sigma}} \right) \sum_{i=1}^{N_I} f_0^I U^I = \bar{U}. \tag{35}
\]

4 SOLUTION PROCEDURE

In the previous section, a two-phase layered composite inclusion model is presented, which is used as a representative microstructural element for the prediction of the large strain mechanical behavior and texture evolution of semi-crystalline polymers. Moreover, in section 2, the constitutive behavior of the local amorphous and crystalline phases is given. In this section, some numerical aspects concerning the solution procedure for the composite inclusion model are discussed. For this purpose, the time domain is subdivided into discrete time steps. First, the time-integration of the viscoplastic deformation of the respective phases is discussed. Then, for each discrete time level, a set of non-linear equations, comprising the interaction law, the local interface conditions and the volume averaging equations, is solved. Thereafter, the interface normals are explicitly updated.

4.1 Time-integration of plastic flow

The viscoplastic behavior of both the amorphous and the crystalline phase is given by a rate-dependent formulation, equation (16) and (20), respectively, whereas the composite inclusion model and interaction laws presented in section 3 are based on the local deformation gradients within each inclusion. Therefore, the plastic deformation gradient of each phase is obtained from time-integration of the plastic flow rule \( \hat{\mathbf{F}}_p^\pi = \mathbf{L}_p^\pi \cdot \mathbf{F}_p^\pi \), with \( \pi = a, c \), denoting either the amorphous phase or the crystalline phase. If \( \mathbf{L}_p^\pi \) is assumed to be constant during an increment with length \( \Delta t \), the plastic deformation at the end of this increment, at time level \( \tau = t + \Delta t \), is given by

\[
\mathbf{F}_p(\tau) = \exp[\Delta t \mathbf{L}_p(\tau)] \cdot \mathbf{F}_p(t), \tag{36}
\]

where the superscript \( \pi \) is omitted. The tensor exponential is numerically evaluated by the Padé approximation [18], so that the incremental plastic deformation gradient is given by

\[
\mathbf{F}_{\text{inc}} = \exp[\Delta t \mathbf{L}_p(\tau)] = \left[ \mathbf{I} - \frac{\Delta t}{2} \mathbf{L}_p(\tau) \right]^{-1} \cdot \left[ \mathbf{I} + \frac{\Delta t}{2} \mathbf{L}_p(\tau) \right]. \tag{37}
\]
For finite increments, the determinant of the approximated exponential term may deviate slightly from unity, which is corrected by a straightforward normalization:

\[ F_p(\tau) = J_{p_{inc}}^{-1/3} F_{p_{inc}} \cdot F_p(t) \quad \text{with} \quad J_{p_{inc}} = \det(F_{p_{inc}}). \tag{38} \]

### 4.2 Well-posed boundary value problem

For an aggregate of composite inclusions, a consistent set of boundary conditions is prescribed for the macroscopic deformation and stress fields. Consider the right polar decomposition \( \mathbf{F} = \mathbf{R} \cdot \mathbf{U} \) of the macroscopic deformation gradient. Then, certain components of \( \mathbf{R}, \mathbf{U} \) and \( \mathbf{\sigma} \) are prescribed according to a well-posed boundary value problem. For example, for uniaxial compression or tension in \( e_3 \)-direction, the following set of boundary conditions are prescribed with respect to a macroscopic orthonormal vector basis \( e_i \):

\[ \mathbf{\bar{R}} = \mathbf{I} \quad ; \quad \bar{U}_{33} = \lambda, \tag{39} \]
\[ \bar{\sigma}_{11} = \bar{\sigma}_{22} = \bar{\sigma}_{12} = \bar{\sigma}_{13} = \bar{\sigma}_{23} = 0. \tag{40} \]

Given these boundary conditions, a set of \( 18 \cdot N_I \) equations can be formed from the equations for interface compatibility (28) and traction equilibrium (29) in combination with either the Taylor interaction law, equations (30) and (31), or the Sachs interaction law, equations (32), (34) and (35), in which the equations for volume averaging within the composite inclusion, (26) and (27), are substituted. With the multiplicative decomposition given by equation (1) and the material models discussed in section 2, all \( 18 \cdot N_I \) equations can be expressed in terms of the elastic deformation gradients \( F^\pi_i \) of each phase \( \pi \) of all \( N_I \) inclusions \( i \), which form a set of \( 18 \cdot N_I \) unknowns. For each increment, this set of non-linear equations for either the Taylor or the Sachs model are solved by a Newton-Raphson iteration procedure.

### 4.3 Morphological texture update

The evolution of morphological texture, i.e. the orientation of the amorphous-crystalline interfaces, is accounted for explicitly. During an increment, the orientations of the normals \( n^i \) remain fixed. After each increment, at time-level \( t \), the inclusion normals are updated, based on convected material vectors within the planar interface [6]. For this purpose, let \( \delta x_{01}^1 \) and \( \delta x_{02}^1 \) be two independent vectors at time \( t = 0 \) in the crystalline-amorphous interface \( \mathcal{X}_0^i \) of the composite. At time \( t \), these vectors are convected to

\[ \delta x_{1}^1 = \mathbf{F} \cdot \delta x_{01}^1 \quad \text{and} \quad \delta x_{2}^1 = \mathbf{F} \cdot \delta x_{02}^1, \tag{41} \]

where \( \mathbf{F} \) can be either \( F^a, F^c \) or \( F^l \). The interface unit normal \( n^l \) is then obtained by

\[ n^l = \frac{\mathbf{F}^l \cdot \delta x_{01}^1 \times \mathbf{F}^l \cdot \delta x_{02}^1}{\| \mathbf{F}^l \cdot \delta x_{01}^1 \times \mathbf{F}^l \cdot \delta x_{02}^1 \|}. \tag{42} \]
Since $F^I \cdot \delta x^I_{01} \times F^I \cdot \delta x^I_{02} = \det(F^I)F^{I-T} \cdot n^I_0$, equation (42) can be rewritten as

$$n^I = \frac{F^{I-T} \cdot n^I_0}{\|F^{I-T} \cdot n^I_0\|}.$$  \hspace{1cm} (43)

5 APPLICATION

In this section, the presented elasto-viscoplastic Taylor-inclusion and Sachs-inclusion models are used to simulate the mechanical behavior of initially isotropic HDPE. The spherulitic structure of melt-crystallized HDPE is represented by an aggregate of 100 inclusions with randomly generated initial orientations of the crystallographic phases. The HDPE crystal lattice is orthorhombic, with the $c$-axis corresponding to the molecular chain direction, and with lattice parameters $a = 7.4$ Å, $b = 4.9$ Å and $c = 2.5$ Å [14]. The initially random distribution of orientations of the principal lattice directions is represented in figure 3(a)–(c). Experimental studies of melt-crystallized polyethylene show that lamellar surfaces are of the $\{h0l\}$ type, where the angle between the chain direction $c$ and the lamellar normal $n^I_0$ varies between $20^\circ$–

![Figure 3: Pole figures representing the initially random orientation distribution of the principal crystallographic lattice directions, (a)–(c), and the interface normals, (d). The projection direction corresponds to the axial loading direction.](image-url)
and 40° \[19][15\]. Here, the initial angle between \(c\) and \(n_0\) is set at 35°, corresponding to the low-index \(\{201\}\) planes. The initial orientations of these lamellar normals are shown in figure 3(d).

The crystalline phase in HDPE employs eight physically distinct slip systems, which are summarized in table 1. This set of slip systems comprises four linearly independent slip systems. The molecular chain axis \(c\) is a constrained direction and therefore in this direction no plastic deformation is possible. The most easily activated slip system is the \((100)[001]\) chain slip system. In table 1, also the resistances at room temperature of all slip systems are given, normalized to \(\tau_0 \equiv g^{(100)[001]} = 8.0 \text{ MPa} \[6][20\]. Strain hardening in the crystalline lamella is neglected.

<table>
<thead>
<tr>
<th>Slip system</th>
<th>(g^\beta/\tau_0)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chain slip</td>
<td></td>
</tr>
<tr>
<td>((100)[001])</td>
<td>1.0</td>
</tr>
<tr>
<td>((010)[001])</td>
<td>2.5</td>
</tr>
<tr>
<td>({110}[001])</td>
<td>2.5</td>
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<tr>
<td>Transverse slip</td>
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<tr>
<td>((100)[010])</td>
<td>1.66</td>
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<td>((010)[100])</td>
<td>2.5</td>
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<td>({110}[1\bar{1}0])</td>
<td>2.2</td>
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Table 1: Slip systems of HDPE and the normalized resistances \[6][20\].

The elastic behavior of the crystalline phase is anisotropic, with a relatively high stiffness in the molecular chain direction. The elastic moduli of the HDPE crystal \[21\] are given in table 2. Following Lee et al. \[6\], the rate exponents of the amorphous and the crystalline phase are set to an equal value of \(n = 9\). Moreover, also the reference shear rate of the amorphous phase is set equal to that of the crystalline phase. The reference initial shear strength of the amorphous phase is written as by \(\tau_0^a = a\tau_0\), with \(a\) the initial deformation resistance of the amorphous phase, relative to the easiest slip system of the crystalline phase. The material parameters of the amorphous phase are summarized in table 3, where the viscoplastic and hardening behavior is taken from Lee et al. \[6\]. Since the typical crystallinity of HDPE is 70 %, the initial volume fraction of the amorphous phase in each composite is set to \(f_0^a = 0.3\).
Elastic \( E \) [MPa] \( \nu \) Viscoplastic [6] \( \dot{\gamma}_0 \) [s\(^{-1}\)] \( n \) \( a \) Hardening [6] \( \mu_R \) [MPa] \( N \)

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<td>0.4</td>
<td>( 10^{-3} )</td>
<td>9</td>
<td>1.2</td>
<td>1.6</td>
<td>49</td>
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Table 3: Material properties of the amorphous phase [6].

In this application, the initially isotropic HDPE material is subjected to constant strain-rate uniaxial compression with \( \lambda = \exp(-\dot{\gamma}_0 t) \). The deformation is considered to be isothermal at temperature \( \theta = \theta_0 \), implying there is no thermal expansion. Figure 4(a) shows the equivalent macroscopic stress \( \bar{\sigma}^{eq} = \sqrt{\frac{2}{3} \bar{\mathbf{S}} : \bar{\mathbf{S}}} \), with \( \bar{\mathbf{S}} = \bar{\sigma} - \frac{1}{3} \text{tr}(\bar{\sigma}) \mathbf{I} \), as a function of the equivalent macroscopic strain \( \bar{\varepsilon}^{eq} = \sqrt{\frac{2}{3} \bar{\varepsilon} : \bar{\varepsilon}} \), with \( \bar{\varepsilon} = \ln(\bar{\mathbf{U}}) \) the macroscopic logarithmic strain tensor, of both the Taylor-inclusion model and the Sachs-inclusion model. Moreover, in figure 4(b), equivalent stress measures of the phase-volume-averaged Cauchy stress tensors are represented for both interaction models. The difference in stress-strain behavior between the two models is due mainly to the behavior of the anisotropic crystalline phases. In the Taylor-inclusion model, all inclusions are enforced to deform by the same inclusion-averaged deformation, whereas in the Sachs-inclusion model, the macroscopic deformation is accommodated mainly by relatively weak inclusions. Therefore, the Taylor-inclusion model shows a significantly higher initial yield strength than the Sachs-inclusion model. In the rigid/viscoplastic Sachs-inclusion model of Lee et al. [6], using the same viscoplastic and hardening parameters,
an initial yield stress of $\bar{\sigma}^{eq}/\tau_0 = 2.9$ is found. For relatively low strains, in the present elasto-viscoplastic Sachs-inclusion model, slightly lower flow stresses are found. At equivalent macroscopic strain $\bar{\varepsilon}^{eq} = 0.5$, a flow stress similar to the Lee et al. model is found. The macroscopic Young’s modulus of the semi-crystalline aggregate is $E_T = 1.9$ GPa for the Taylor-inclusion model and $E_S = 0.65$ GPa for the Sachs-inclusion model. In figure 5, the crystallographic textures are given for both models at $\bar{\varepsilon}^{eq} = 0.5$. To enrich the information given in the plots, each pole is reflected with respect to the central point of the pole plot. The Sachs-inclusion model exhibits a more distinct evolution of crystallographic texturing than the Taylor-inclusion model and consequently, the stress-strain behavior of the Sachs-inclusion model shows more textural strain hardening in the early stage of deformation, which is approximately linear within this strain range. The Taylor-inclusion model however, shows less strain hardening up to $\bar{\varepsilon}^{eq} = 0.3$, whereas for higher equivalent macroscopic strains, increased textural strain hardening is observed. In the inclusions that accommodate most deformation, i.e. that are “most active”, in the Sachs-inclusion model, the crystalline phase is found to initially deform mainly by the two easiest slip systems, the (100)[001] chain slip system and the (100)[010] transverse slip sys-
tem, which is in agreement with observations by Bartczak et al. [22]. In the Taylor-inclusion model however, all inclusions are forced to be active. This is accomplished by deformation on other slip systems than mentioned above for relatively stiff inclusions. For both models, most (100) poles, which represent the crystallographic slip planes of the two easiest slip systems, are moving towards the compression direction, with, for the Sachs-inclusion model, a maximum intensity of poles at an altitude angle of 20° with the loading direction, which is in agreement with experimental results of Bartczak et al. [22]. In the Taylor-inclusion model however, a more diffuse pattern is formed, with a maximum intensity of poles with an altitude angle in the range 0°–40°. In figure 6, the path of each pole is shown. The Sachs-inclusion model exhibits a more structured pattern for the crystallographic plane trajectories. However it shows more variation in the length of the trajectories since only relatively weak inclusions are active, in contrast to the Taylor-inclusion model, where all inclusions are forced to be active. Since, in the latter model, slip occurs on planes other than the principal slip plane (100), the direction of the pole trajectories is more diverse. For the Sachs-inclusion model, the (100) poles approaching the compression direction are found to change direction when attaining an angle of approximately 15°–20°. Consequently, the initial goal, which is the compression direction, is not reached. This alteration of the direction of the trajectories corresponds to the activation of less easy slip systems, mainly the {110}<110> slip systems. Some (100) planes, with their normals close to the radial direction, move away from the compression direction. Most (001) poles in the Sachs-inclusion model are moving away from the compression direction, forming a ring with a maximum intensity at an altitude angle of 78°. The trajectories of the (001) poles in the Taylor-inclusion model are more diverse, resulting in a more spread out ring with a maximum intensity at approximately 60°–70°.

In figure 7, the morphological texture, i.e. the orientation of the lamellar normals, is shown for both the Taylor and the Sachs-inclusion model. Additionally, in figure 8, the trajectory of each pole is shown. In both models, the normals of the inclusion interfaces are migrating towards the compression direction, with a maximum intensity of normals in the center of the pole figure. The Sachs-inclusion model exhibits a slightly sharper texture than the Taylor-inclusion model. Although, the final textures show a large resemblance, the trajectory plots clearly demonstrate the difference between the Taylor-inclusion model and the Sachs-inclusion model. Since in equation (43), the local deformation gradient tensor can be replaced by the macroscopic deformation gradient $\bar{F}$, the lamellar normal trajectories show a homogeneous pattern. The Sachs-inclusion model, however, imposes a much weaker constraint on the local deformations, allowing for variations in the local deformation. Therefore, the interface normals in the Sachs-inclusion model show a more heterogeneous pattern.

6 CONCLUSIONS

A thermo-elasto-viscoplastic model for the constitutive behavior of semi-crystalline polymers is developed. The model is based on the micromechanically observed morphology and deformation mechanisms of this material. As a representative microstructural element, a two-phase layered composite inclusion is used, based on the lamellar structure that is commonly
observed in semi-crystalline polymers. Both the crystalline and the amorphous phase are represented in the composite inclusion model and are mechanically coupled by enforcing compatibility and traction equilibrium on the interface between the crystalline lamella and the corresponding amorphous layer. For both phases, a micromechanically-based constitutive model is formulated within the framework of an elasto-thermo-viscoplastic decomposition of the deformation gradient. To relate the local inclusion-averaged deformation and stress to the macroscopic fields of the aggregate, both a Taylor-type and a Sachs-type interaction model are used.

By addition of elastic and thermal deformation components to the local constitutive models of the respective phases, the model captures the effects of macroscopic stiffness and thermal expansion. For HDPE in uniaxial compression, application of the Taylor-inclusion model leads to an unreasonably strong increase of stresses in relatively stiff inclusions for strains larger...
than $\bar{\varepsilon}^{\text{eq}} = 0.3$. The Sachs-inclusion model however, provides a weakened measure of compatibility, and thus circumvents the difficulty of relatively stiff inclusions. The crystallographic textures obtained in this model seem to be in agreement with experimental observations.

The presented composite inclusion model can be used to study the influence of microstructural morphology and deformation mechanisms on the macroscopic deformation resistances for preferentially oriented semi-crystalline polymers. In particular, the model will be used to study the matrix deformation in particle-toughened semi-crystalline materials. For this purpose, an aggregate of composite inclusions will be used as a representative microstructural unit at each integration point of a finite element formulation.

REFERENCES


Figure 8: The trajectory of each pole of the interface normals as predicted by the Taylor-inclusion model, (a), and the Sachs-inclusion model, (d), for HDPE subjected to uniaxial compression at equivalent macroscopic strain $\bar{\varepsilon} = 0.5$. The dots represent the initial position of the poles.


