

MICROMECHANICAL MODELING OF SEMICRYSTALLINE POLYMERS
WITH SPHERULITE MORPHOLOGY

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WITH SPHERULITE MORPHOLOGY**

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I hereby declare that all information in this document has been obtained and presented in accordance with academic rules and ethical conduct. I also declare that, as required by these rules and conduct, I have fully cited and referenced all material and results that are not original to this work.

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ABSTRACT

MICROMECHANICAL MODELING OF SEMICRYSTALLINE POLYMERS WITH SPHERULITE MORPHOLOGY

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In this study, a micromechanical constitutive model for semicrystalline polymers is developed. The proposed model considers interactions among the adjacent regions at the same length scale. The neighborhood information is obtained from the well characterized spherulite morphology of polyethylene. Deformation characteristics of a disk-like spherulite model that has similarities with the considered modeling approach are studied using the finite element method. The model discretizes a disk-like spherulite into amorphous and crystalline slices, separated by planar interfaces. Consistent deformation of the amorphous and crystalline phases of semircrystalline polymers at the spherulite length scale is the distinctive feature of the model. A three-dimensional spherulite is also modeled, and it is found that the employment of the crystallographic slip as the only plastic deformation mode of crystalline phase leads to plastically soft polar regions in spherulites, and therefore contradicts with the experimental observations. This simplification is employed in all semicrystalline polymer models that we are aware of in literature. In order to obtain a simplified disk-like spherulite model, a set of methods that consider the neighborhood informa-

tion is developed by extending the rank-1 laminates to support an arbitrary number of lamination directions. The equilibrium and compatibility conditions are not exactly satisfied together on the interfaces, as a consequence of the uniform strain field assumption of rank-1 laminates. The methods are consistent averaging schemes, and satisfy the Hill's energy criterion and the separation of length scales principle. According to these qualities, they could be employed in micromechanical constitutive models. Disk-like spherulite and twisted lamellar microstructure of the spherulites are modeled with the methods. Approximately transversely isotropic responses are obtained in the disk-like spherulite, and a desired tension-compression asymmetry is observed in the twisted lamellar microstructure model. Additionally, without uniform strain field assumption, another simplified disk-like spherulite model is obtained by connecting non-parallel rank-1 laminates with compatible strain fields in cylindrical coordinates, which may enable application of non-local constitutive models in rank-1 laminates. The disk-like spherulite models are considered as the analogous of the symmetric crystals with many common slip systems that are known to be good candidates for Taylor-type homogenization. A constitutive model for high density polyethylene is obtained with Taylor-type homogenization of randomly oriented realizations of the disk-like spherulites, where the elasto-viscoplastic constitutive models of the phases and their parameters are adopted from the literature. The stress-strain response and texture evolution of the model are in line with the literature. An attempt to obtain a micromechanical constitutive model for ultra high molecular weight polyethylene is also presented, and the influence of surface texture on the friction coefficient is studied using the finite element method and a constitutive model from literature.

Keywords: semicrystalline polymer, micromechanics, crystal plasticity, spherulite morphology, rank-1 laminate

ÖZ

KÜRESEL MİKRO YAPI MORFOLOJİLİ YARI KRİSTAL POLİMERLERİN MİKROMEKANİK MODELLENMESİ

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Bu çalışmada yarı kristal polimerler için mikromekaniksel bir bünye denklemi geliştirilmiştir. Modelde bölgelerin komşularıyla etkileşimi aynı ölçekte dikkate alınmaktadır. Komşuluk bilgisi polietilenin iyi karakterize edilmiş olan küresel mikro yapı morfolojisinden alınmıştır. Bünye denkleminde kullanılan yaklaşımla benzerlikleri bulunan disk biçimli bir küresel mikro yapı modelinin şekil değiştirme özellikleri sonlu elemanlar yöntemi ile incelenmiştir. Modelde, disk biçimli küresel mikro yapı düzlemsel arayüzlerle amorf ve kristal dilimlere ayrılır. Yarı kristal polimerlerin amorf ve kristal fazlarının küresel mikro yapı ölçeğinde uyumlu şekil değiştirebilmesi, modelin ayırt edici özelliğidir. Ayrıca üç boyutlu bir küresel mikro yapı modeli daha geliştirilmiştir. Kristal fazın sadece kristalografik kayma ile plastik şekil değiştirmesi yaklaşımının, deneysel gözlemlerin aksine, küresel mikro yapıların kutup bölgelerinde kolay plastik akmaya neden olduğu bulunmuştur. Bu basitleştirme bildiğimiz tüm yarı kristal polimer modellerinde kullanılmaktadır. Basitleştirilmiş disk-biçimli küresel mikro yapı modelleri elde etmek için, birinci dereceden lamelleri istenen sa-

yıda katmanlanma doğrultusuna izin verecek şekilde geliştirerek komşuluk ilişkilerini dikkate alan bir dizi yenilikçi yöntem önerilmiştir. Birinci dereceden lamelin tek tip gerinim alanı yaklaşımı değiştirilmediğinden, bu yöntemlerle ara yüzlerde denge ve kinematik uyum koşulları artık tam olarak birlikte sağlanmamaktadır. Önerilen yöntemler tutarlı ortalama yaklaşımlardır ve Hill'in enerji kriteri ile ölçeklerin ayrılması prensibini sağlar. Bu özellikleri sayesinde mikromekanik bünye denklemlerinde kullanılabilirler. Disk biçimli küresel mikro yapı ve küresel yapılarda görülen burgulu lamel yapısı bu yöntemlerle modellenmiştir. Disk biçimli modellerin davranışı yaklaşık olarak enine yönbağımsızken, burgulu lamel modelinde de istenen bir çekme-basma asimetrisi bulunmuştur. Ayrıca farklı bir basitleştirilmiş disk biçimli küresel mikro yapı modeli, tek tip gerinim alanı yaklaşımını kullanmadan paralel olmayan birinci dereceden lamellerin silindirik koordinat sisteminde uyumlu bir gerinim alanıyla birleştirilmesiye elde edilmiştir. Bu gerinim alanı lokal olmayan bünye denklemlerinin birinci dereceden lamellerle de kullanılmasını sağlayabilir. Disk biçimli küresel mikro yapı modellerinin Taylor tipi homojenleştirmeye uygun olan simetrik ve bir çok ortak kayma sistemi bulunan kristallere benzediği değerlendirilmiştir. Böylece, yüksek yoğunluklu polietilen için bir bünye denklemi rastgele yönelimli disk biçimli küresel mikro yapı modellerinin Taylor tipi homojenleştirilmesi ile elde edilmiştir. Fazların bünye denklemleri ile malzeme parametreleri ise literatürden alınmıştır. Bünye denkleminin gerilme-gerinim davranışı ve doku gelişimi literatürle uyumludur. Bünye denkleminin ultra yüksek yoğunluklu polietilene uyarlanmasına çalışılmış, yüzey dokusunun sürtünme katsayısına etkisini inceleyen sonlu elemanlar analizleri literatürden alınan bir bünye denklemiyle yapılmıştır.

Anahtar Kelimeler: yarı kristal polimerler, mikromekanik, kristal plastisitesi, küresel mikro yapı morfolojisi, birinci dereceden lamel



To My Parents H. & A. Oktay

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CHAPTER 1

INTRODUCTION

1.1 General

Polymers, being a typical constituent of plastics in industry have a wide range of application areas in modern life. Among different polymers, semicrystalline polymers (SCP) gain significant interest owing to their remarkable deformability and toughness and superior wear resistance. Notable examples of semicrystalline polymers are high-density polyethylene (HDPE), ultra high molecular weight polyethylene (UHMWPE), Nylon-6, poly(ethylene terephthalate) (PET), isotactic polypropylene (iPP). Semicrystalline polymers have been used in applications such as substrates for flexible electronic devices, plastic bags, piping systems and liquid and gas containers. Although there has been a substantial number of studies on the mechanical behavior of SCPs in the literature, a clear description of all stages of deformation is still unavailable. Micromechanical models may provide insight into the influence of the identified deformation mechanisms on the macroscopic response.

1.2 Semicrystalline Polymers

In SCPs, the segments of polymer chains orient in an orderly fashion to form crystalline structures called lamellae. The long polymer chains generally form thin lamellae by folding repetitively. As an example, the unit cell of the crystalline phase of PE is presented in Figure 1.1. In PE, the orthorhombic unit cell has the lattice parameters $a = 0.74$ nm, $b = 0.49$ nm and $c = 0.25$ nm, where c axis corresponds to the chain direction which is formed by strong covalent bonds. Considering the melt-crystal-

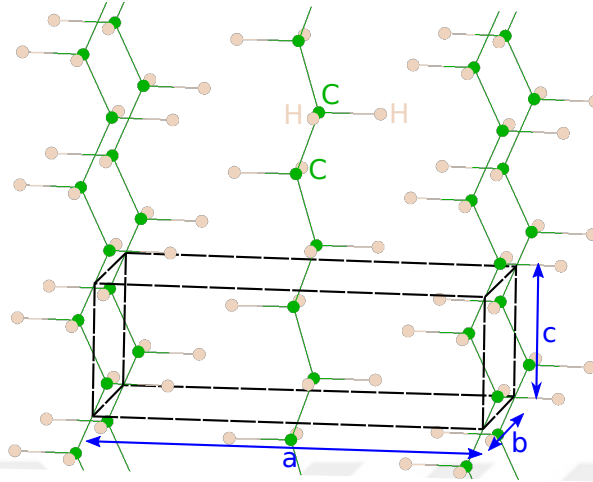


Figure 1.1: Orthorhombic unit cell of polyethylene crystal, adapted from Lin and Argon [64]

lized lamellae, the thickness of the chain folded lamellae is around 20 nm, crystal growth is along the b lattice axis and the chain folds are mostly considered to lie in the plane the normal vector of which is the lattice axis a [64]. Therefore, according to Figure 1.1, it could be imagined that the chains fold approximately towards the out-of-plane direction to form the lamellae. Between two lamellae there exist amorphous regions. Amorphous regions host tie chains that form interlamellar connections binding multiple lamellae together. As a result, amorphous and crystalline phases deform together and consistently. Crystalline lamellae could also form complex structures such as spherulites where the lamellae grow from a central nucleus in radial directions. The percentage of crystalline phase in the SCP is called crystallinity ratio. The crystallinity ratio, among many other factors, depends strongly on the thermal processing conditions, and the percentage of crystalline phase can normally vary from 10% to 80%. Deformation of SCPs is complex due to this multiphasic and hierarchical microstructure. Therefore, it is important to take into account the microstructure while developing a powerful and predictive material model. Moreover, models that consider the fundamental deformation modes, as presented in this study, may be useful to identify the shortcomings of these modes and possibly point out to overlooked deformation mechanisms.

As also expressed in the review articles [64, 80, 13], SCPs possess the deformation

mechanisms of the polymeric materials as well as the crystalline materials. Similar to metals, dislocation-based crystalline slip is the major plastic deformation mechanism of the crystalline phase of SCPs. The strong polymer chains that form the crystals remain intact during the slip, and therefore the slip planes always contain the lattice axis c presented in Figure 1.1. Slip systems with slip directions along the lattice axis c are referred to as chain slip, and the remaining slip systems the slip directions of which are perpendicular to the chain are referred to as transverse slip systems. Since the crystallographic slip does not break the chain, the polymer single crystals does not have the required number of slip systems for arbitrary plastic deformation and appear brittle when alone. However, the surrounding amorphous phase provides two additional glide systems with the interlamellar shear deformation mode [13]. The interlamellar shear plane is the interface between the crystalline lamellae and the surrounding amorphous phase. The normal vector of this plane is approximately along the chains that form the lamellae. Therefore, amorphous phase could be considered to provide the required slip systems to accommodate an arbitrary plastic deformation, and the semicrystalline polymers are ductile and quite tough. The anisotropic crystalline phase of SCPs is orders of magnitude stiffer than the isotropic amorphous phase which causes a highly inhomogeneous deformation distribution in the spherulite microstructure. The amorphous phase, on the other hand, is much easier to deform up to a locking stretch. The locking stretch corresponds to alignment of polymer chains in the direction of stretch such that the tensile strength of the strong polymer chains governs the response. In this view, the locking stretch depends on the chain length and the degree of entanglement of the chains. Due to the significant stiffness difference between these two phases and the locking stretch phenomenon, the deformation of the semicrystalline polymers is a multi-stage process.

1.3 Micromechanics

Advances in material science and composites lead to an increase in the ability to design the microstructure of many engineering materials. For instance, the volume fraction and orientations of the crystalline phase of semicrystalline polymers as well as their morphology could be adjusted with processing conditions and the molecular

weight of the chain. Moreover, the amount of cross-linking between the amorphous chains could be controlled by radiation. Depending on these microstructural properties, a wide range of engineering properties such as stiffness, toughness, yield stress, anisotropy and durability could be obtained from essentially the same material such as polyethylene. Another example frequently given in literature [41, 85, 82] is the composite laminates, which combines different materials. The materials that are used as fiber and matrix, the ordering of the layers with different fiber orientations, the number and size of the fibers within the plies are among the properties of these materials that could be tuned. While most materials are heterogeneous, they could be considered as homogeneous in most engineering designs with confidence, including the examples given. In other words, most designs are based on the experiments that characterize the materials with parameters that are representative for homogeneous materials, and are quite successful. However, a testing program for characterizing a new material is composed of numerous experiments. Therefore, alternative methods to predict the influence of the microstructure are quite valuable, which is not only to identify the promising microstructures, but also to develop intuition about the influence of the microstructural parameters on the observed response [41]. In 2008, National Research Council of the National Academies has published a report [75] that states that the development of advanced materials has become a bottleneck in the design of new products when the success of the computational engineering tools are considered. The report recommends the development of analogous computational tools for materials engineering to ultimately enable the optimization of the properties of a product by the designers. Micromechanics is the branch of continuum mechanics that analyzes the effects of the microscopic constituents and the influence of their interactions on the properties of heterogeneous materials. Constitutive behavior, size, shape, spatial distribution and volume fraction within the material are among the considered properties of the constituents. Homogenization methods of micromechanics provide a homogeneous equivalent of a heterogeneous material, generally as a constitutive model based on these properties. Considering multiple experiments are required even to characterize a linear elastic material that is anisotropic, a constitutive model is especially valuable, since it provides a remarkable amount of information.

The separation of the length scales and the statistical homogeneity conditions, that are

generally postulated by homogenization methods, are not satisfied by all materials or engineering applications. Both conditions are related to the concept of representative volume element (RVE), as follows. Consider a large body of a typical heterogeneous material with a complex and non-uniform heterogeneity distribution. The variation of the distribution throughout the body could be left out if the variation is statistically uniform. Then a small sample, called RVE, from an arbitrary position could provide a good representation of the whole. While RVE is generally taken as cubic, the size of RVE depends on the material and the application area, and could be determined according to the statistical or mechanical approaches in the literature. Since an RVE is taken from an arbitrary point in the body, by definition, the RVE is also considered to be representative for any region of the body that has the same size and the shape with the RVE. Each one of these regions could be referred to with the reference coordinate $\mathbf{X}$ of a point in the body by considering the centroid of RVE is placed on $\mathbf{X}$, as depicted in Figure 1.2 [76]. Constitutive models of homogenization methods are relationships between the volume average of the stress and strain fields over the RVE. Therefore, employing an equivalent homogeneous constitutive model in a problem is considered as an application of a moving averages procedure. In order for this approach to be applicable for a problem, clearly, the body should be much larger than the RVE, ideally up to the point that the RVE could be considered as a continuum point of the body as also presented in Figure 1.2. This condition is known as the micro-meso-macro (MMM) principle of Hashin [42]. Micro, meso and macro terms are frequently employed in literature to refer to the length scales of the heterogeneity size, RVE size and body size, respectively. The MMM principle of Hashin could also be expressed as follows. RVE should include numerous heterogeneities in order to statistically represent their complex distribution. Therefore, micro scale should be much smaller than the meso scale. On the other hand, for a constitutive model obtained from the RVE to be applicable to a macroscopic problem, mesoscopic scale should be much smaller than the macroscopic scale. The separation of length scales principle is more strict than the MMM principle of Hashin. According to this principle, the variation of the macroscopic loading should be negligible in the length scale of the RVE, in addition to the MMM principle of Hashin. Therefore, the gradients of the macroscopic stress and strain fields are negligible and are taken as zero in RVE.

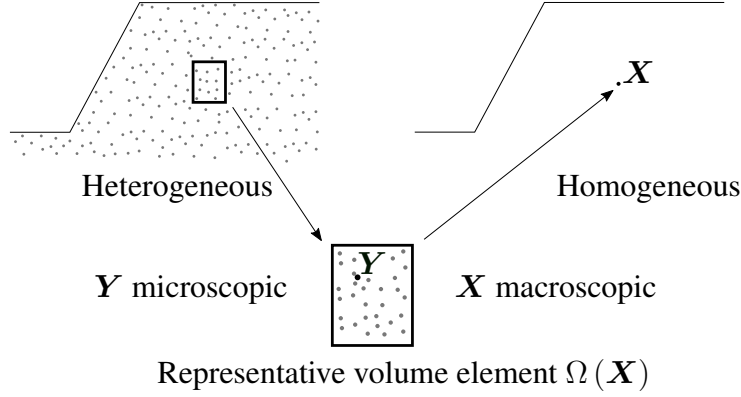


Figure 1.2: Homogeneous equivalent of a heterogeneous material with an RVE, adapted from Suquet [98]

It is known that the separation of length scales principle is not satisfied at points close to the surface of a body. This is not only due to the possible stress concentrations but also since a part of the RVE is left outside of the body for coordinates $\mathbf{X}$ close to the boundary. Nevertheless, the idea of relationship between the averages is actually borrowed from continuum mechanics [42, 69]. Since the matter is composed of atoms, continuum mechanics also applies this idea albeit at a different scale where micro scale is the atomistic scale. In continuum mechanics, the tractions and displacements on the surface of a region are considered as a major means of interaction of the region with its surroundings. In micromechanics, average stress and strain theorems with the Hill's macrohomogeneity criterion (see [76, 85, 82]) provide justification for working with the volume averages within the continuum mechanics theory.

1.3.1 Volume Averaging Framework

Volume averaging framework is usually employed in the homogenization methods. Among the many tensor measures related to the strain and stress in continuum mechanics, the deformation gradient $\mathbf{F}$ and its energetically conjugate, the first Piola-Kirchhoff stress $\mathbf{P}$, are a pair of the suitable measures for volume averaging [45]. Additionally, mechanical quantities such as internal energy, dissipation, density and volume are also homogenized with volume averaging [21]. According to the separation of the length scales principle, the gradients of the stress and strain fields are negligible in the length scale of the RVE. Therefore, the RVE $\Omega(\mathbf{X})$ at $\mathbf{X}$ is ideally

considered to be under both the linear displacement

$$\mathbf{y}(\mathbf{Y})|_{\partial\Omega} = \bar{\mathbf{F}}(\mathbf{X}) \mathbf{Y} \quad \text{or} \quad \mathbf{u}(\mathbf{Y})|_{\partial\Omega} = (\bar{\mathbf{F}}(\mathbf{X}) - \mathbf{1}) \mathbf{Y} \quad (1.1)$$

and uniform traction

$$\mathbf{t}(\mathbf{Y})|_{\partial\Omega} = \bar{\mathbf{P}}(\mathbf{X}) \mathbf{N}(\mathbf{Y}) \quad (1.2)$$

boundary conditions, where $\bar{\mathbf{P}}(\mathbf{X})$ and $\bar{\mathbf{F}}(\mathbf{X})$ are the first Piola-Kirchhoff stress and the deformation gradient that are considered to be uniform in the length scale of the RVE, $\mathbf{Y}$ and $\mathbf{y}$ are the reference and current microscopic coordinates, $\mathbf{u}(\mathbf{Y})$ is the displacement vector, $\mathbf{t}(\mathbf{Y})$ is the traction vector and $\mathbf{N}(\mathbf{Y})$ is the unit normal vector of the area element of the RVE surface. Note that uniform boundary conditions do not imply the uniformity of the fields $\mathbf{F}(\mathbf{Y})$ and $\mathbf{P}(\mathbf{Y})$, which may significantly fluctuate due to the heterogeneities in the RVE. However,

$$\bar{\mathbf{F}}(\mathbf{X}) = \frac{1}{|\Omega|} \int_{\Omega(\mathbf{X})} \mathbf{F}(\mathbf{Y}) \, dV \quad (1.3)$$

whenever linear displacement boundary conditions (1.1) are applied according to the average strain theorem, and according to the average stress theorem

$$\bar{\mathbf{P}}(\mathbf{X}) = \frac{1}{|\Omega|} \int_{\Omega(\mathbf{X})} \mathbf{P}(\mathbf{Y}) \, dV \quad (1.4)$$

whenever uniform traction boundary conditions (1.2) are applied. For homogeneous regions, whenever a linear displacement boundary condition (1.1) is enforced, uniform surface tractions in (1.2) are obtained, and vice versa. However, for regions including heterogeneities, some deviation from the ideal condition is expected. Nevertheless, in homogenization methods, (1.3) and (1.4) are generally employed together. As will be revealed, this is also related to the idea of homogeneous equivalent of a heterogeneous material, the homogenization methods aim to provide. The relations (1.3) and (1.4) could be employed together such that i) for a given $\bar{\mathbf{F}}$, (1.3) is enforced and $\bar{\mathbf{P}}$ is found from (1.4), or the opposite ii) for a given $\bar{\mathbf{P}}$, $\bar{\mathbf{F}}$ could be obtained. These two approaches could be considered as analogous of the deformation- and stress-controlled experiments, respectively. If the obtained stress $\bar{\mathbf{P}}$ from the first approach, is employed in the second approach as a given, and provides a similar deformation gradient with the original $\bar{\mathbf{F}}$, then the RVE is regarded as statistically representative. Conversely, the RVE is statistically representative if $\bar{\mathbf{F}}$ obtained from the second approach provides a similar first Piola-Kirchhoff tensor in the first

approach with the original $\bar{\mathbf{P}}$. For linear elasticity at small strains, the first and second approaches correspond respectively to the statements

$$\bar{\boldsymbol{\sigma}} = \bar{\mathbb{C}} : \bar{\boldsymbol{\varepsilon}} \quad \text{and} \quad \bar{\boldsymbol{\varepsilon}} = \bar{\mathbb{M}} : \bar{\boldsymbol{\sigma}}, \quad (1.5)$$

and the corresponding statistical representativity condition is

$$\bar{\mathbb{C}} = \bar{\mathbb{M}}^{-1}, \quad (1.6)$$

where $\bar{\mathbb{C}}$ and $\bar{\mathbb{M}}$ are referred to as the effective stiffness and compliance tensors, respectively [76]. Recall that homogenization methods aim to provide an equivalent homogeneous material of a given heterogeneous material, generally as a constitutive model. Then, for linear elasticity, the desired constitutive model is (1.5), and $\bar{\mathbb{C}}$ and $\bar{\mathbb{M}}$ are the stiffness and compliance tensors of the corresponding homogeneous material. Computation of the effective properties such as $\bar{\mathbb{C}}$ and $\bar{\mathbb{M}}$, from the averages of stress and strain is referred to as the direct approach [42], when additional approximations are not introduced. In some cases, the additional approximations lead to the violation of (1.6), even when the RVE is representative. Averaging schemes that could satisfy (1.6) are distinguished by referring to them as consistent.

Alternatively, the equivalent homogeneous material could also be defined in terms of the strain energy, as follows. Returning to the general materials, according to the Hill's macrohomogeneity criterion, also known as the Hill-Mandel condition, it could be shown that the rate of change of the average strain energy in the RVE is

$$\frac{1}{|\Omega|} \int_{\Omega(\mathbf{X})} \mathbf{P}(\mathbf{Y}) : \dot{\mathbf{F}}(\mathbf{Y}) \, dV = \bar{\mathbf{P}}(\mathbf{X}) : \dot{\bar{\mathbf{F}}}(\mathbf{X}), \quad (1.7)$$

when (1.1) or (1.2) is prescribed as the boundary condition. The corresponding average rate of strain energy in the equivalent homogeneous material is

$$\frac{1}{|\Omega|} \int_{\Omega(\mathbf{X})} \mathbf{P}^u(\mathbf{X}) : \dot{\mathbf{F}}(\mathbf{X}) \, dV = \mathbf{P}^u(\mathbf{X}) : \dot{\bar{\mathbf{F}}}(\mathbf{X}), \quad (1.8)$$

where the stress field $\mathbf{P}^u$ would be uniform under the either boundary condition. Requiring the equality between the strain energy rates (1.7) and (1.8) reveals

$$\mathbf{P}^u = \bar{\mathbf{P}}.$$

Moreover, when the strain field is compatible and the stress field is in equilibrium, the rate of change of the complementary strain energy in the RVE is

$$\bar{\mathbf{F}} : \dot{\bar{\mathbf{P}}} = \frac{1}{|\Omega|} \int_{\Omega} \mathbf{F} : \dot{\mathbf{P}} \, dV, \quad (1.9)$$

when (1.7) is satisfied [76]. Then, similar to (1.1) and (1.2), it could be shown that

$$\mathbf{F}^u = \bar{\mathbf{F}}$$

where $\mathbf{F}^u$ is the uniform deformation gradient field of the homogeneous material under the either boundary condition. Requiring the equality between the free energy density function of the homogeneous equivalent materials $\bar{\Psi}(\bar{\mathbf{F}})$ and the volume average that of the phases in RVE

$$\bar{\Psi}(\bar{\mathbf{F}}) = \frac{1}{|\Omega|} \int_{\Omega} \Psi(\mathbf{F}) \, dV \quad (1.10)$$

also satisfy the equality between the rates of strain energy. Similarly the equality between the complementary strain energy density functions $\bar{\Phi}(\bar{\mathbf{P}})$ and $\Phi(\mathbf{P})$ reads

$$\bar{\Phi}(\bar{\mathbf{P}}) = \frac{1}{|\Omega|} \int_{\Omega} \Phi(\mathbf{P}) \, dV. \quad (1.11)$$

Therefore, respectively, the above first and second approaches are similar to defining a strain energy density function $\bar{\Psi}(\bar{\mathbf{F}})$ and a complementary strain energy density function $\bar{\Phi}(\bar{\mathbf{P}})$, such that

$$\bar{\mathbf{P}} = \frac{\partial \bar{\Psi}}{\partial \bar{\mathbf{F}}} \quad \text{and} \quad \bar{\mathbf{F}} = \frac{\partial \bar{\Phi}}{\partial \bar{\mathbf{P}}}.$$

Then, for a consistent averaging scheme that is employed with a statistically representative RVE, it follows that if $\frac{\partial \bar{\Psi}}{\partial \bar{\mathbf{F}}} \approx \bar{\mathbf{P}}$ then $\frac{\partial \bar{\Phi}}{\partial \bar{\mathbf{P}}} \approx \bar{\mathbf{F}}$, where

$$\bar{\Psi}(\bar{\mathbf{F}}) = \frac{1}{|\Omega|} \min_{\mathbf{y} \in \mathcal{K}(\bar{\mathbf{F}})} \int_{\Omega} \Psi \, dV \quad \text{and} \quad \bar{\Phi} = \frac{1}{|\Omega|} \min_{\mathbf{P} \in \mathcal{S}(\bar{\mathbf{P}})} \int_{\Omega} \Phi \, dV \quad (1.12)$$

according to principles of minimum potential energy and minimum complementary energy, respectively. In (1.12), $\mathcal{K}(\bar{\mathbf{F}})$ is the set of continuous deformed coordinates $\mathbf{y}$ that satisfy (1.1), and $\mathcal{S}(\bar{\mathbf{P}})$ is the set of first Piola-Kirchhoff stress fields that satisfy equilibrium and (1.2). It could be noted that, in thermodynamics, the principle of minimum potential energy is considered to be valid for equilibrium configurations with uniform temperature fields. Therefore, the appropriate strain energy density Ψ in (1.12) is the Helmholtz free energy per unit reference volume [99].

1.3.2 Homogenization Methods

Eshelby solution for the homogeneous inclusion problem has been influential in the homogenization literature. Eshelby [32] considers small strains, and imagines a uni-

form and infinite matrix D made of a linear elastic material. The stress and strain fields are uniform and zero in the matrix. Eshelby considers that an ellipsoidal region Ω of the matrix spontaneously develops a uniform eigen-strain ϵ^* . That is, if the region Ω is cut out of the matrix, its strain would be ϵ^* under zero boundary tractions. Therefore, the stress field of D is considered to become $\sigma = -\mathbb{C} : \epsilon^*$ in Ω and $\sigma = 0$ in $D - \Omega$, where $\mathbb{C}$ is the stiffness tensor, and a new equilibrium state is sought. In the equilibrium, Eshelby analytically showed that the strain and consequently the stress field within the region are uniform, and equal to

$$\epsilon = \mathbf{S} : \epsilon^* \text{ in } \Omega \quad (1.13)$$

and

$$\sigma = \mathbb{C} : (\epsilon - \epsilon^*) = \mathbb{C} : (\mathbf{S} - \mathbf{1}) : \epsilon^* \text{ in } \Omega ,$$

respectively, where $\mathbf{S}$ is the Eshelby's tensor that only depends on the aspect ratios of the ellipsoid and the stiffness matrix $\mathbb{C}$. The stress and strain fields are, of course, non-uniform outside Ω in this equilibrium. The average strain in the matrix, on the other hand, would still be very close to zero, i.e. $\frac{1}{D-\Omega} \int_{D-\Omega} \epsilon \, dV = 0$, considering D is infinite. It could be remarked that, while the uniformity is rather intuitive for a spherical region in an isotropic medium, it is also valid for arbitrary ellipsoids that may resemble pins and for anisotropic media as well. Nevertheless, not all methods that are based on the Eshelby solution support the wide aspect ratio range and anisotropic materials, given their additional assumptions. Eshelby [32] also considered a practically more important problem, where the region has a different stiffness tensor $\mathbb{C}^R$ than that of the matrix $\mathbb{C}$, and a strain $\bar{\epsilon}$ is applied to the matrix on the far-field. This problem is solved by making use of the strain field of the eigen-strain case, as follows. Consider another case, where the material with stiffness tensor $\mathbb{C}^R$ is replaced with the matrix material and an eigen-strain ϵ^* is introduced. Using the principle of linear superposition the strain field could be decomposed as $\bar{\epsilon} + \epsilon$, and ϵ is defined as the same field for the two cases. Then the stress fields would also be the same between the cases when

$$\mathbb{C}^R : (\bar{\epsilon} + \epsilon) = \mathbb{C} : (\bar{\epsilon} + \epsilon - \epsilon^*) , \quad (1.14)$$

where considering the relation (1.13), the value of the only unknown ϵ^* could be obtained. This method is known as the equivalent inclusion method. In this method,

the eigen-strain ϵ^* is a variable that also depends on the applied strain $\bar{\epsilon}$. It should be remarked that the size or the relative amount of the heterogeneity compared to the matrix is immaterial to the solution. Considering the equivalent inclusion method finds uniform stress and strain fields in the heterogeneity, following homogenization method is proposed as the Eshelby method to obtain the overall stiffness matrix of a composite composed of a matrix that surrounds particles that are far apart. In the method, the volume fraction of the heterogeneities are taken into account, where the volume fraction of heterogeneity I is defined as $\nu^I = V^I/V$, and V^I is the volume of the heterogeneity, and V is the finite volume of the RVE. Then for a given $\bar{\epsilon}$, the overall stress is computed by the volume average of stress (1.4), which becomes

$$\bar{\sigma} = \sum_{i=1}^N \nu^i \sigma^i + \left(1 - \sum_{i=1}^N \nu^i\right) \sigma^0 \quad (1.15)$$

at small strains, where each σ^I , $1 \leq I \leq N$ is obtained from the equivalent inclusion method separately and σ^0 is the average stress of the matrix taken as $\sigma^0 = \mathbb{C} : \bar{\epsilon}$. Therefore, any interaction between the inhomogeneities is disregarded. The effective modulus of the Eshelby method satisfy (1.6) only in the dilute limit, i.e. $\nu^0 \approx 1$ and $\nu^I \approx 0$ for $I > 0$. The Mori-Tanaka method aims to be more representative in the non-dilute cases, and extends the Eshelby method by replacing $\bar{\epsilon}$ in (1.14) with the average strain in the matrix ϵ^0 that is found from (1.3), which becomes

$$\bar{\epsilon} = \sum_{i=0}^N \nu^i \epsilon^i \quad (1.16)$$

for small strain kinematics, where ϵ^I for $I > 0$ is the uniform (and thus the average) strain in the heterogeneity I . Additionally the average stress of the matrix in (1.15) is taken as $\sigma^0 = \mathbb{C} : \epsilon^0$. The effective modulus of the Mori-Tanaka method satisfies (1.6) in non-dilute cases as well. In turn, the self-consistent scheme extends the Mori-Tanaka method by considering the stiffness of the matrix phase is not sufficient to represent the medium that surrounds the heterogeneities, and a better approach is to employ the effective stiffness $\bar{\mathbb{C}}$ of (1.5) instead of $\mathbb{C}$ in (1.14). Since the unknown stiffness $\bar{\mathbb{C}}$ is also employed as a variable, the resulting implicit equation needs to be solved iteratively. While the self-consistent method satisfies (1.6), it is known to provide negative or infinite bulk and shear responses when the heterogeneities are taken as voids and rigid particles respectively, for certain ranges of total volume fractions of

the heterogeneities generally 50% and above [49]. To overcome these difficulties, the generalized self-consistent method is proposed where a thin layer of matrix material is introduced between the heterogeneity and the effective medium. A comparison of these methods and their explicit equations can be found in Qu and Cherkaoui [85].

Not all composites possess a distinct matrix that surrounds the heterogeneities. Polycrystalline materials are frequently considered to be composed of anisotropic homogeneous crystals, that are identical except their orientations. The self-consistent scheme, which does not put a special emphasis on the matrix phase, is frequently employed with polycrystals such as metals. It is also employed successfully in non-linear regime, and a review could be found in Kanoute et al. [49]. Considering the wide-spread use of the method with success, there are remarks in literature that the method, while it is practically quite valuable, is in fact an approximation. In particular, the surface tractions of heterogeneities are required to be in equilibrium with the tractions computed using $\bar{\mathbb{C}}$. Hashin [42] explains that this approach violates the MMM principle since it implies that the heterogeneities, which are in the microscale, are considered to be in the macroscale with the following tree and forest analogy. A tree can only see the nearby trees, while the self-consistent scheme suggests a tree can see the forest. This analogy is also reviewed by Markov [68].

Energy-based methods are also employed for the homogenization of polycrystalline materials. One advantage of these approaches is that they could be employed to obtain bounds on the effective properties with energy principles. For the linear elastic behavior, it is known that [49], Voigt [113] and Reuss [113] estimates are the upper and lower bounds of the effective stiffness, according respectively to the principle of minimum potential energy and the principle of minimum complementary potential energy (1.12). In 1889, Voigt [113] estimated the overall stiffness with the average stress in (1.4) by assuming the strain is the same in all crystals, therefore neglected static equilibrium. Conversely, after four decades, Reuss [87] estimated the overall stiffness with the average strain in (1.3), and considered that stresses are the same in all crystals and neglected kinematical compatibility. These bounds are generic and therefore quite wide, since they only depend on the volume fractions and stiffness of the crystals, and additional information is required for narrower bounds. For example, the well known Hashin-Shtrikman bounds [43] are applicable for materials that are

statistically isotropic, and a brief review can be found in Saeb et al. [92]. In passing, it could be mentioned that, if an RVE of the material is available, it possesses much more information than the volume fractions. In that case, the response of the RVE is bounded from above by the uniform displacement boundary conditions, and from below with uniform traction boundary conditions. As explained in Section 1.3.1, both should provide the same response if the RVE is sufficiently large.

There are also bounds for inelastic behavior of polycrystals. Major inelastic deformation mode of polycrystals is the dislocation based crystallographic slip, a subject of physical metallurgy. This deformation mode is modeled with crystal plasticity framework in literature, and an application of which is also presented in Section 2.2. Bishop and Hill [19] presented upper and lower bounds for the yield stress with the uniform strain and stress assumptions respectively considering rigid plasticity taking the crystallographic slip as the only deformation mode. As reviewed by Bishop and Hill [19], in uniaxial tension, Sachs [91] estimated the yield stress of face centered cubic (FCC) polycrystals as 2.2 times the slip resistance of a single crystal. The estimation implied uniform stress in all crystal similar to Reuss, since axial stress along the loading direction is taken as the same in all crystals. The result is not in good agreement with the experimental stress-strain curves. After a decade, Taylor [100] on the other hand, estimated the yield stress by considering uniform strain in all crystals. While slip amounts and therefore the strain can be calculated unambiguously for a given stress, the converse is not true for strain and there are $C_{12}^5 = 792$ possible combinations per each crystal, since an arbitrary plastic strain has 5 independent components and there are 12 slip systems in FCC crystals. Selecting a reasonable set among these is not only important for predicting the yield stress correctly but also to predict the stress-strain curve that depends on the so-called texture evolution. Crystallographic texture evolution is a result of rotation of the crystals with plastic deformation. The axis and the rate of rotation depends on the activated slip systems and slip rates. Based on experimental observations, Taylor [100] suggested to select the combination that would result in the shortest cumulative slip. Taylor additionally assumed an equal strain hardening for all slip systems. The predicted uniaxial stress-strain curve of aluminum for a rigid plastic constitutive model of the crystals was in a good agreement with the experiments. According to the curve, the yield stress of aluminum polycrystal is

approximately 3.1 times the slip resistance of its single crystal. Bishop and Hill [19], showed the assumptions of Taylor correspond to the principle of maximum work not only for uniaxial, but also for arbitrary macroscopic stresses. Taylor and Sachs approaches employ respectively the assumptions of Voigt and Reuss to crystal models that have the crystallographic slip deformation mode, and are referred to as bounds for the nonlinear material response. Success of the Taylor approach is widely appreciated, and it is considered as the first significant homogenization approach for polycrystal plasticity [119]. It is frequently used for the homogenization of materials with highly symmetric crystals that possess many common slip systems with resistances much lower than their elastic stiffness, such as FCC metals, to capture the general trends. While the uniform strain assumption of Taylor is considered as reasonable in those cases, in general the approaches based on self-consistent scheme are considered as more realistic [3]. Moreover, the self-consistent scheme is also applied to crystals that have less than five independent slip systems, since the kinematical conditions are not as strong as that of the Taylor approach [83]. There are also methods that aim to relax the Taylor approach, by weakening its uniform strain constraints or by introducing elasticity, and some of which are also reviewed in Chapter 3. Some of these studies, for example LAMEL model [111, 110], are specialized and applicable only for the specific deformation modes of metal forming, such as rolling where the grains are flat and parallel. Today, Taylor and Sachs methods are criticized for not including morphological information, as they depend only on the volume fractions of the phases in the microstructure [92]. The methods that aim to improve experimental agreement by relaxing the Taylor approach are also criticized for losing the extremal nature of the method, while not introducing the essentially missing feature, i.e. the statistical information on the distribution of the constituents in space [119]. Moreover, these methods are not considered reliable for modeling complex non-linear structures [92].

Nevertheless, approaches that are similar to the criticized studies are successfully employed in modeling semicrystalline polymers, without criticism. A notable example is Lee et al. [61], which proposed the basic structural unit of spherulites in SCPs as a laminate composed of amorphous and crystalline phases that are separated by a planar interface. As also expressed by Lee et al. [61], this approach is similar to the ones that relax the Taylor approach for the parallel and flat grains in literature.

While the crystalline lamellae of spherulites are also thin, the motivation of Lee et al. [61] is not only the shape of the lamellae but also the characteristics of the widely studied spherulite morphology. Detailed information is available in literature, such as which lattice plane is the interface that is between the crystalline and amorphous phases. Therefore, some statistical information is incorporated in the model of Lee et al. [61]. The approach of Lee et al. [61], is widely adopted by micromechanical SCP models developed ever since, including the ones that employ the self-consistent scheme [78, 2].

1.4 Objective and Outline

Aim of this study is to develop a micromechanical constitutive model for semicrystalline polymers that also takes the morphological information into account. The morphological information is based on the widely observed and well characterized spherulites of polyethylene. Therefore, in Chapter 2 a finite element method model which simplifies a disk-like spherulite as a composite that is composed of amorphous and crystalline slices separated by planar interfaces is studied and the results are compared with the important features of the spherulite deformation. Consistent deformation of the crystalline and amorphous phases within the length scale of the spherulite is the distinctive feature of the model compared to the other studies in literature. Spatial distribution characteristics of slip systems in the model are also among the investigated features of the deformation. An inconsistency with the literature on the activity of a certain slip system is further investigated with a three dimensional spherulite model. The inconsistency is associated to the widely employed approach of taking the crystallographic slip as the only plastic deformation mode of the crystalline phase.

In Chapter 3, several novel methods to take the morphological information into account are developed, in order to develop the simplified versions of the disk-like spherulite model. The methods are consistent averaging schemes, and satisfy the Hill's energy criterion and the separation of length scales principle. These methods extend the rank-1 laminate to support an arbitrary number of lamination directions. Therefore, there are multiple interfaces with different orientations at the same length scale in the proposed methods. Since the uniform stress and strain field approach of

the rank-1 laminate is preserved in the methods, equilibrium and compatibility are not satisfied together. One of these physical requirements is replaced with an energetically motivated condition that minimizes the free energy, while the other could be exactly satisfied. As the number of regions increases and the consecutive interfaces approach each other, the approximation is improved. Chapter begins with a review of the constitutive models of semicrystalline polymers. Next, their widely employed basic structural unit, the rank-1 laminate is introduced. As a preliminary approach for solving a simplified disk-like spherulite model, sequential laminates are also employed. Sequential laminates are laminates of rank-1 laminates, therefore the interfaces of the simplified spherulite model are not solved at the same length scale. Then, the rank-1 laminates are extended to support an arbitrary number of lamination directions, at the same length scale, with different sets of approximations, and the disk-like spherulite model is solved with each approximation. Depending on the employed method, models with approximately transversely isotropic responses are also obtained, where the plane of isotropy is the surface of the disk. The methods could be employed with geometries discretized into regions separated by planar interfaces where each region has at most two neighbors. In order to present an example to the different geometries that could be employed, the twisted ribbon shaped lamella is also modeled.

In Chapter 4, another simplified version of the disk-like spherulite model is presented, based on rank-1 laminates and Taylor-type homogenization, where the strain fields of the slices are compatible and non-uniform. The non-uniform strain field of the model could be employed to calculate the spatial derivatives of the strain fields, which are required when non-local constitutive models are to be employed with the rank-1 laminates.

In Chapter 5, a micromechanical constitutive model is proposed for HDPE, based on the disk like spherulite models of Chapter 3. The disk like spherulite models have approximately transversely isotropic responses, and in literature the interlamellar shear deformation mode of the amorphous phase is often commented as a slip system. Therefore, the models are considered as the analogous to the symmetric crystals with many common slip systems, that are known to be good candidates for Taylor-type homogenization. Accordingly, a micromechanical constitutive model is

obtained with Taylor-type homogenization of randomly oriented realizations of the disk-like spherulites, where the neighborhood information regarding the interactions between the lamellae and multiple amorphous regions are taken into account. In the phases, elasto-viscoplastic constitutive models in large strain framework, that are adopted from literature are employed. The stress-strain response and texture evolution of the model are presented and compared with the literature.

In Chapter 6 another semicrystalline polymer, UHMWPE, is studied. UHMWPE components are frequently employed in total joint replacement designs. Influence of surface texture on the friction coefficient is studied with FEM analyses that employ a constitutive model from literature. An attempt to adapt the HDPE constitutive model of Chapter 5 to UHMWPE is also presented. Finally, conclusions and recommendations are presented in the last chapter.



CHAPTER 2

FINITE ELEMENT MODELING OF THE SPHERULITE MICROSTRUCTURE

Bulk specimens of PE often have spherulitic morphology. In this chapter, disk-like and three-dimensional spherulite morphologies of PE are modeled with Finite Element Method (FEM). The volume fractions and spatial arrangement of the amorphous and crystalline phases at the length scale of a single spherulite are taken into account.

2.1 Introduction

One of the most common morphologies observed in the melt crystallized semicrystalline polymers is the spherulite. Reviews of the morphology and deformation of the spherulites in the context of PE, and semicrystalline polymers can be found in Lin and Argon [64] and Oleinik [80], respectively. Crist and Schultz [26] is a recent study that reviews the morphology of the spherulites. A spherulite is composed of crystalline lamellae that are embedded in a matrix of amorphous material, and grow out from a common central nucleus in radial directions. The lamellae in the spherulite are ribbon shaped, and only 3 to 20 nm thick. In many cases the lamellae have a twist along the radial directions as presented in Figure 2.1. During crystallization, growth of a lamella continues until it impinges with another spherulite. The diameter of the spherulites can range from 0.1 to 1000 μm , and in the case of PE it is in the order of 100 μm . Orientation of the crystalline phase that makes up the lamellae of the spherulite in PE could be described with the following two general conditions. Radial direction of the spherulite, i.e. growth direction of the lamellae, corresponds to the b lattice axis of the crystal [64]. $\{201\}$ plane of the crystal lattice is considered

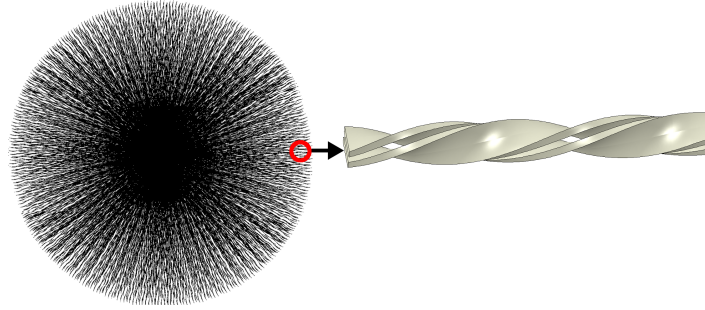


Figure 2.1: Schematic view of the spherulite composed of twisted ribbon shaped lamellae, adapted from Sedighiamiri et al. [96]

as the crystal-amorphous interface, that lies on the broad face of the ribbon shaped lamellae [35]. Considering the lamellae grow radially in all directions with a twist, these conditions also provide the observed isotropic response of the samples with spherulitic morphology.

Deformation field of a spherulite is non-uniform as a result of its multiple phases and the spatial arrangement of the lamellae described above. In the literature, deformation of the spherulites is discussed in terms of three regions presented in Figure 2.2. These are called (i) equatorial, (ii) polar and (iii) inclined regions, and are defined in terms of the orientation of the radial directions with respect to the tensile loading direction, where the origin is the nucleus of the spherulite. In equatorial and polar regions, the radial direction is almost perpendicular and parallel to the tensile loading direction, respectively. Remaining parts of the spherulite belongs to the inclined region. A more quantitative description of boundaries of equatorial, inclined and polar regions within a spherulite is given by Allan and Bevis [5] as presented in Figure 2.2, which allocates slightly less central angle to the polar region compared to the inclined and equatorial regions.

The mechanical behavior of spherulites is a complex phenomenon and it is not yet fully understood. The three-dimensional shape of spherulites increases this complexity. However, early and fundamental studies by Hay and Keller [44] and Allan and Bevis [5, 6] have been able to conduct experiments on 2D spherulites (i.e., cylinder or disk like) deliberately formed within thin film samples. Amorphous tie chains connect the lamellae to each other, and the spherulite deforms consistently as a whole. The

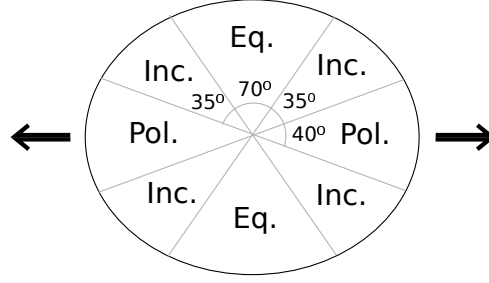


Figure 2.2: Quantitative description of equatorial (Eq.), inclined (Inc.) and polar (Pol.) regions of a spherulite with respect to the indicated straining axis, adapted from Allan and Bevis [5]

lamellae are formed from crystals, and are elastically much stiffer than the surrounding amorphous regions. Therefore, in initial deformation stages, the deformation is mainly accommodated by the amorphous phase. According to the geometrical arrangement of the lamellae, interlamellar separation and interlamellar shear deformation modes of the amorphous phase are relatively more easily activated in equatorial and inclined regions, respectively [64]. Deformation begins first in the equatorial regions, and there is high strain concentration in the nucleus [44].

It is well known that at very large tensile deformations, the chains of the polymer become parallel to the loading direction. Hay and Keller [44] and Allan and Bevis [5, 6] conducted tensile experiments on thin films with disk-like spherulite morphology. Hay and Keller [44] proposes the following deformation modes to explain the orientation change of the chain axis from that of the spherulite to the highly oriented state. In the equatorial region, crystals should rotate around the b axis in order to align the c axis with the loading direction. $(100)[001]$ chain slip and interlamellar slip provides this rotation. In the polar region, crystals should rotate around the a lattice axis, which is possible with $(010)[001]$ chain slip. In the inclined region the rotation should be on an intermediate axis between the a and b axes. Allan and Bevis [5] report crystallographic slip on (110) planes, $\{110\}$ twinning and a stress-induced martensitic transformation in the equatorial regions. Additionally, Allan and Bevis [6] propose that the following mechanisms could explain their observations: In polar region the major deformation mechanism is the $\{110\}\langle\bar{1}\bar{1}0\rangle$ transverse slip and the micro-necking initiated in the (110) planes. In the inclined region, chain slip and

the slip on the (110) planes dominate the deformation of the crystalline phase, while chain slip is not favored. Both Allan and Bevis [6] and Hay and Keller [44] considers the polar region as much more resistant to deformation than the other regions.

Lin and Argon [64] and Oleinik [80] contain the review of the early models for spherulites. One of these studies is by Wang [114], which employs the linear elasticity theory, and considers the radially aligned lamellae and the amorphous phase in between. In this model, stress and strain depend on the radius, and goes to infinity at the nucleus. Comparing the equatorial and polar regions, the strain component in the loading direction is higher in the equatorial, while the stress component is higher in the polar region. Van Dommelen et al. [108], is a more recent study, where a homogenization based elasto-viscoplastic material model [109] is employed in axisymmetric and plane strain FE models of a multi-spherulitic RVE. In addition to the stress and strain fields, they also report the activities of the deformation mechanisms of the amorphous and crystalline phases within the spherulite. Tomita and Uchida [105] proposed a three-dimensional spherulite model where a single inclusion, composed of an amorphous and a crystalline phase is employed in each integration point of the FE model. Uchida and Tada [106] assigned the response of the 3D spherulite RVE model to the integration points of a macroscopic finite element model.

These recent spherulite models do not have elements or integration points allocated to the amorphous or crystalline regions. Instead they employ the same constitutive models, which are based on the laminar composites, in all integration points. In that scheme, consistent deformation of the amorphous and crystalline phases exists at the length scale of the laminar composite, but not at the length scale of the spherulite. In this study, we propose two and three-dimensional spherulite models, which provide the consistent deformation of the crystalline and amorphous regions throughout the spherulite.

2.2 Constitutive Models

Two constitutive models, employing the finite deformation theory, are used in this study. For the amorphous phase, the 8-chain rubber elasticity model [8] is used since

the amorphous phase of the HDPE is in the rubbery state at the room temperature. The amorphous phase is governed by three material parameters; the bulk modulus κ , the shear modulus μ , and the locking stretch λ_{lock} which are taken from [109] and given in Table 2.1.

Table 2.1: Material parameters for the amorphous phase

κ	μ	λ_{lock}
[GPa]	[MPa]	[-]
2.0	35.0	7

For the crystalline phase, a crystal viscoplasticity model is employed. The model utilizes the classical multiplicative decomposition of deformation gradient $\mathbf{F}$ into elastic and plastic contributions [63]

$$\mathbf{F} = \mathbf{F}^e \mathbf{F}^p. \quad (2.1)$$

In (2.1), $\mathbf{F}^e$ and $\mathbf{F}^p$ are the elastic and plastic components of the deformation gradient, respectively. In this approach it is assumed that there exists a stress-free configuration that could be reached with $\mathbf{F}^p$ from the reference configuration. Plastic flow is restricted to occur in given directions on predefined planes. Each one of these (direction, plane) pairs are referred to as a slip system. Apart from elastic deformations, it is considered that the plastic slip does not alter the orientation of the crystal lattice. The rate of plastic deformation $\mathbf{L}^p$ is the sum of the slips on all slip systems

$$\mathbf{L}^p = \dot{\mathbf{F}}^p \mathbf{F}^{p-1} = \sum_{\alpha=1}^N \dot{\gamma}^\alpha \mathbf{s}^\alpha \otimes \mathbf{m}^\alpha. \quad (2.2)$$

In (2.2), $\mathbf{s}^\alpha$ and $\mathbf{m}^\alpha$ are defined in the reference configuration and correspond to the slip direction and the slip plane normal unit vectors, respectively. Moreover, $\dot{\gamma}^\alpha$ is the slip rate in the slip system α . Evolution of the plastic component of the deformation gradient is computed from (2.2) by utilizing the exponential integration scheme, which exactly preserves the incompressibility of the plastic flow [72]. In this case the evolution equation for the plastic component of the deformation gradient can algorithmically be written as

$$\mathbf{F}_{n+1}^p = \Delta \mathbf{F}_{n+1}^p \mathbf{F}_n^p = \exp \left(\Delta t \sum_{\alpha=1}^N \dot{\gamma}_{n+1}^\alpha \mathbf{s}^\alpha \otimes \mathbf{m}^\alpha \right) \mathbf{F}_n^p. \quad (2.3)$$

In the above equation, subscripts denote the time steps and Δt is the time increment. The tensor exponential is computed numerically according to the series expansion given in [29]. Stresses are calculated from the elastic component $\mathbf{F}^e$ of the deformation gradient. For this purpose, the Saint-Venant Kirchhoff model

$$\mathbf{S}^e = \mathbb{C}^e : \mathbf{E}^e \quad (2.4)$$

is used. In (2.4) $\mathbf{S}^e$ is the lattice-based elastic second Piola-Kirchhoff stress, $\mathbb{C}^e$ is the fourth-order elastic stiffness tensor and $\mathbf{E}^e$ is the Green-Lagrange strain tensor

$$\mathbf{E}^e = \frac{1}{2} (\mathbf{F}^{eT} \mathbf{F}^e - \mathbf{I}), \quad (2.5)$$

where $\mathbf{I}$ is the second order identity tensor. The slip rate $\dot{\gamma}^\alpha$ is defined by a power law type relation

$$\dot{\gamma}^\alpha = \dot{\gamma}_0 \left(\frac{\tau^\alpha}{\tau_y^\alpha} \right) \left| \frac{\tau^\alpha}{\tau_y^\alpha} \right|^{n-1} \quad (2.6)$$

where $\dot{\gamma}_0$, τ_y^α , n are material parameters called the reference shear rate, the critical resolved shear stress of the slip system α , and the rate sensitivity parameter, respectively. In (2.6) τ^α is the Schmid shear stress. The Schmid shear stress is the slip direction component of the traction on the slip plane. The Schmid shear stress τ^α is given by [29]

$$\tau^\alpha = \mathbf{R}^{eT} \boldsymbol{\tau} \mathbf{R}^e : (\mathbf{s}^\alpha \otimes \mathbf{m}^\alpha). \quad (2.7)$$

In (2.7), $\mathbf{R}^e$ is the finite rotation tensor obtained from $\mathbf{F}^e$ according to the polar decomposition theorem, $\boldsymbol{\tau} = \mathbf{F}^e \mathbf{S}^e \mathbf{F}^{eT}$ is the lattice-based Kirchhoff stress. Elastic constants used in the model are presented in Table 2.2. These values are based on the measurements of Choy and Leung [23] on the polyethylene (PE). The PE crystal has orthotropic symmetry which requires nine independent elastic constants. Due to the lack of experimental data ($\mathbb{C}_{11}$, $\mathbb{C}_{22}$), ($\mathbb{C}_{12}$, $\mathbb{C}_{23}$) and ($\mathbb{C}_{44}$, $\mathbb{C}_{55}$, $\mathbb{C}_{66}$) components are taken as equal, respectively. This approach is previously employed in [96]. In Table 2.3 viscoplastic material parameters of the crystalline phase are presented. The slip systems of the polyethylene crystal and the critical resolved shear stresses normalized by the reference shear stress τ_0 are also given in Table 2.4. The lack of strain hardening in PE crystals is considered to be explained by the push out of the dislocations from the thin lamellae core [13]. Therefore, the values of the critical resolved shear stresses are taken as constant. On the other hand, the critical resolved shear stresses

are known to depend on the normal component of the traction vector or pressure on the slip plane. This relationship is measured and the parameters for a Coulomb-type strain hardening law are available for the chain slip systems and one transverse slip system [10]. The reason of this dependence, although not completely understood, is primarily attributed to the roughness of the slip planes due to the atomic bonds that are not in the chain direction [64]. For example in PE, as presented in Figure 1.1, the chain grows with C=C bonds, and therefore C-H bonds are not along the chain direction. The dependence of the slip resistance to the pressure is not taken into account in this study.

Table 2.2: Elastic constants for PE crystal [GPa] [96]

C_{11}	C_{22}	C_{33}	C_{12}	C_{23}	C_{13}	C_{44}	C_{55}	C_{66}
7	7	81	3.8	3.8	4.7	1.6	1.6	1.6

Table 2.3: Viscoplastic material parameters [109] of the crystalline phase

$\dot{\gamma}_0$ [s ⁻¹]	n	τ_0 [MPa]
1×10^{-3}	9	8

Table 2.4: Slip systems of polyethylene crystal [61]

	Slip System	τ_y^α / τ_0
Chain Slip	(100)[001]	1.0
	(010)[001]	2.5
	{110}[001]	2.5
Transverse Slip	(100)[010]	1.66
	(010)[100]	2.5
	{110} $\langle 1\bar{1}0 \rangle$	2.2

2.3 Disk-like Spherulite Model

In this study, we propose a new spherulite model which provides consistent deformation of both crystalline and amorphous phases throughout the spherulite. The con-

structured model is solved with FEM under different loading conditions. The elastic-viscoplastic crystal plasticity constitutive model presented in Section 2.2 is employed for the crystalline phase, while the 8-chain model [8] is employed for the amorphous phase. Crystallographic slip is considered as the only plastic deformation mechanism of the crystalline phase. Since a rubber elasticity model is used for the amorphous phase, results of this study are representative only above the glass transition temperature where the amorphous phase is rubbery [13]. It is shown that the model captures the evolution of inhomogeneous plastic deformation activity in a spherulite microstructure, as well as the important features of the spherulite deformation reported in the literature.

Hay and Keller [44] and Allan and Bevis [5] have been able to conduct experiments on 2D spherulites (i.e., cylindrical or disk like) within thin film samples. Therefore, a section of a spherulite with 100 μm diameter, is idealized as a rectangular prism with dimensions 100 μm x 100 μm x 20 μm and divided into amorphous and crystalline slices joining at the nucleus of spherulite as shown in Figure 2.3a for 50% crystallinity.

Orientations of crystallographic axes in PE are accounted for in the model with the following conditions: (i) spherulite crystal growth direction is the b lattice axis of the crystal [64], (ii) amorphous phase and crystal lamellae interface is the $\{201\}$ plane of the crystal lattice [35]. The first condition is enforced by assigning orientations with reference to a cylindrical coordinate system, origin of which is located at the nucleus of spherulite and b axis of the crystal lattice is fixed to the polar axis of the coordinate system. In this manner, crystal orientation varies in a continuous fashion for each integration point, according to integration point's location with respect to nucleus of the spherulite. The second condition is satisfied by orienting the lattice by a rotation along b axis that sets the angle between c axis and the amorphous phase interface normal to 35° . Orientation assignments are visualized in Figure 2.7 for 32 and 4 slice cases. Change of crystal orientation within a crystalline slice in this fashion may not be physically correct. However, compared to a single uniform crystal orientation per slice alternative, such continuous orientation assignment enables better idealization of the spherulite's actually very thin lamellae in our model. This is especially significant for the small number of slice cases of our model, such as the 4 slice case shown in

Figure 2.6a. As for the amorphous slices, a lack of very thin amorphous slices in our model is not consistent with a true interlamellar spacing. However, this might be justified considering that general plasticity kinetics of spherulite is controlled by the crystalline phase and amorphous phase is forced to adjust to the process that took place in crystalline phase [80, 13]. Rather thick amorphous phase slices, shown in Figure 2.3a might be considered sufficient to capture this phenomenon. Another limitation of the model is that, twist of the lamellae along the growth axis of the lamellae of the spherulite is not considered. The twisting period, which is reported as $2\text{ }\mu\text{m}$ [51], is considered to be small compared to the distance between the integration points of the employed FE meshes.

2.3.1 Loading Types

Simple shear, uniaxial compression, and simple tension loading cases are considered in this study. For all cases, loading is applied by controlling displacement. Figures 2.4a to 2.4c present the boundary conditions employed for each case; whereas Figure 2.4d presents the deformation constraints assigned along the out-of-plane direction of nucleus for simple tension case.

For simple tension case, loading is applied to the left and right surfaces of the model in opposite directions in the form of uniform nodal displacements as shown in Figure 2.4a. Additionally, as could be seen in Figure 2.4d, all nodes lying along the line passing through the nucleus in the out-of-plane direction is fixed in the y direction; while a single node that lies in the middle of that line is fixed in all directions, to prohibit rigid body translation. In uniaxial compression case, loading is applied to the side surfaces of the model as shown in Figure 2.4b while not allowing out-of-plane deformation of nodes that lie at the mid of the thickness along the surface of the model in order to hinder buckling mode of deformation. Boundary conditions at nucleus is similar to the simple tension case, except the mid-node is not fixed in the out-of-plane direction. As for the simple shear loading case, loading is applied to the top and bottom surfaces of the model in the opposite x directions, while fixing the y displacements as shown in Figure 2.4c. The boundary condition assigned to the nucleus is similar to the simple tension case, however in this case, in addition to the y

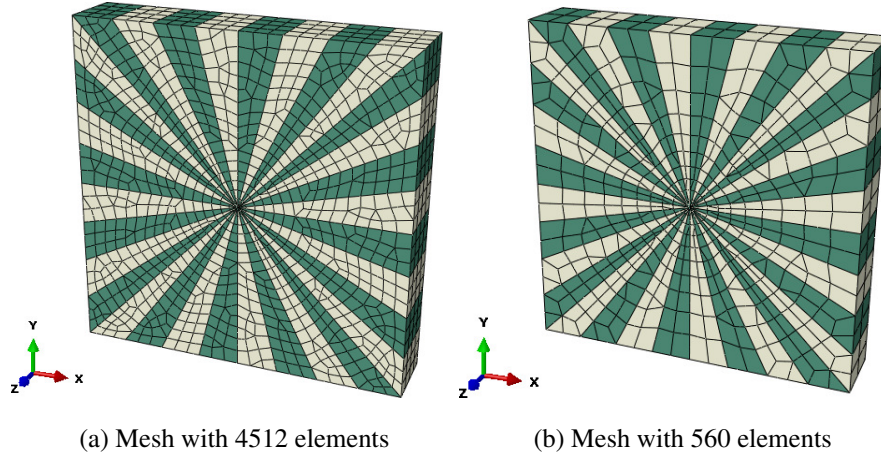


Figure 2.3: Meshes employed in the mesh convergence study

displacement constraints, x displacement is restrained for the nodes as well, in order to enhance the uniformness of shear deformation within the model. For all loading cases, loading is applied to maintain a constant engineering strain rate of $1 \times 10^{-3} \text{ s}^{-1}$.

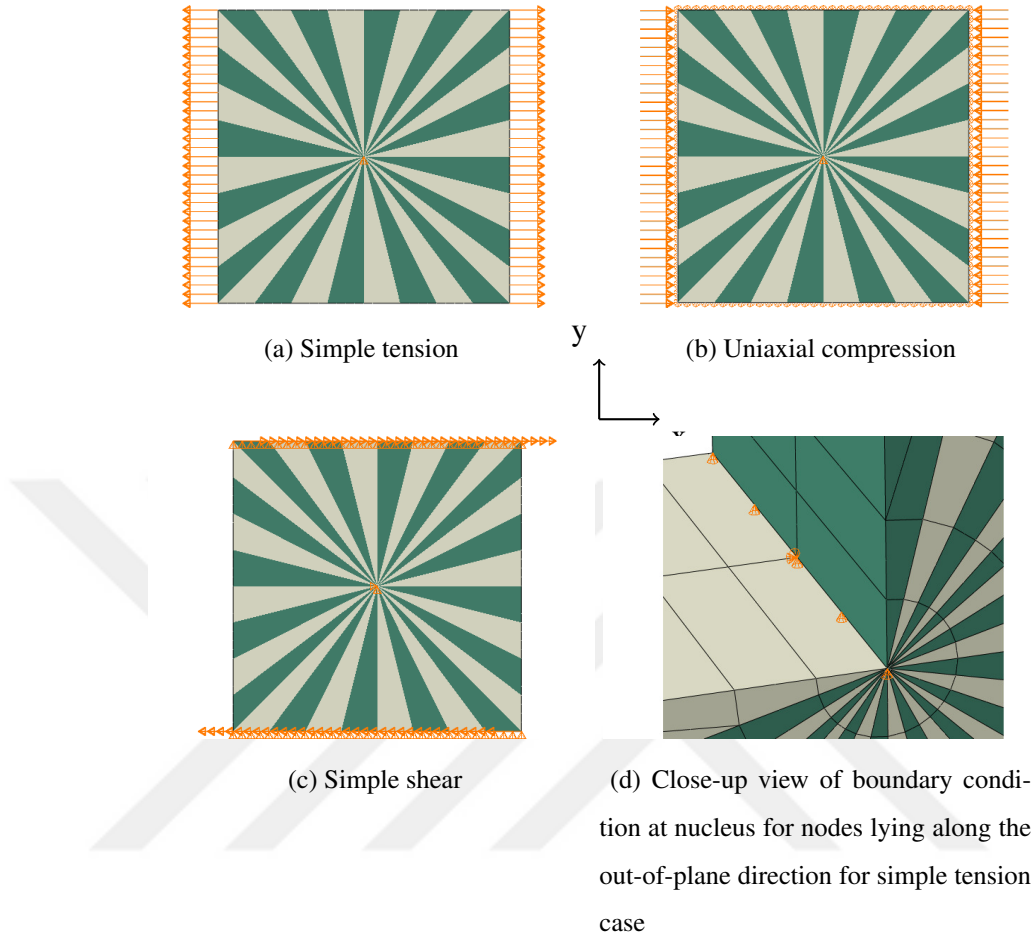


Figure 2.4: Boundary conditions employed in displacement controlled simple shear, uniaxial compression and simple tension loading cases. Dark and light colored slices correspond to amorphous and crystalline phases, respectively.

2.3.2 Mesh Convergence

Slices of the model join at the central nucleus which results in elements with small element corner angles in that region. Therefore, with mesh quality concerns, wedge shaped elements are used alongside brick shaped elements in the mesh. Quadratic reduced integration brick elements (C3D20R), and quadratic wedge elements (C3D15) that are available in Abaqus element library are employed in the analyses. A mesh convergence study is conducted on a single spherulite model composed of 32 slices. Studied meshes are given in Figure 2.3a, where slices representing amorphous phase are presented in darker color than the crystal phase slices. Corresponding stress-strain

curves obtained from finite element analyses for simple tension, uniaxial compression and simple shear loading are given in Figure 2.5. The coarse mesh consists of 560 elements; whereas the fine mesh has 4512 elements. Level of difference between curves in Figure 2.5 is considered as acceptable, and the coarser mesh shown in Figure 2.3b is used in analyses of this study.

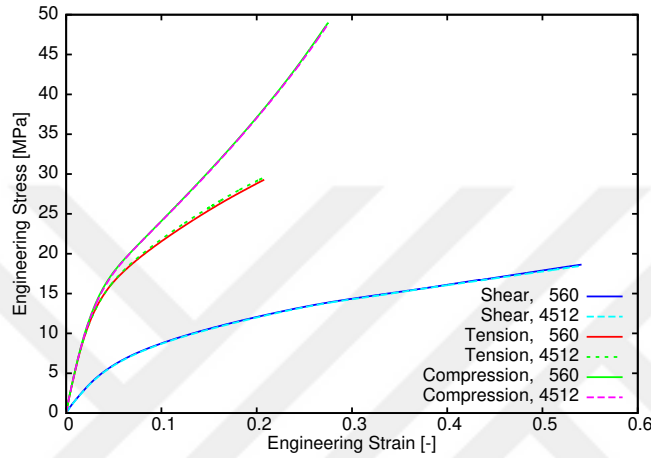


Figure 2.5: Comparison of engineering stress strain curves of different loading conditions conducted with meshes in Figures 2.3a and 2.3b

2.3.3 Effects of Slice Size

Size of the spherulite slices directly influences the minimum number of elements that could be employed in the finite element mesh. Therefore, the number of slices is one of the factors that determines the computational cost of FEM analyses. Influence of slice size on the results is investigated by dividing the single spherulite model into 4, 8, 16 and 32 number of slices as shown in Figure 2.6. In each case, 50% crystallinity by volume is considered. In order to exclude the influence of mesh effect on the solution; the same mesh, which could support the 32 slices case, is used in all models regardless of the number of slices case studied.

The constructed spherulite finite element models are subjected to simple tension, uniaxial compression and simple shear deformations and obtained engineering stress-strain curves are presented in Figure 2.8. Initial elastic stiffness of the curves in Figure 2.8 are presented in Figure 2.9. For tensile and compressive loading cases, initial

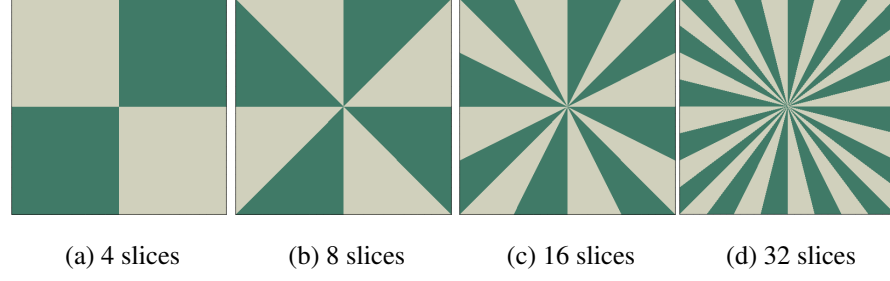


Figure 2.6: Single spherulite model of 50% crystallinity, divided into a varying number of phase slices. Dark and light colored slices correspond to amorphous and crystalline phases, respectively.

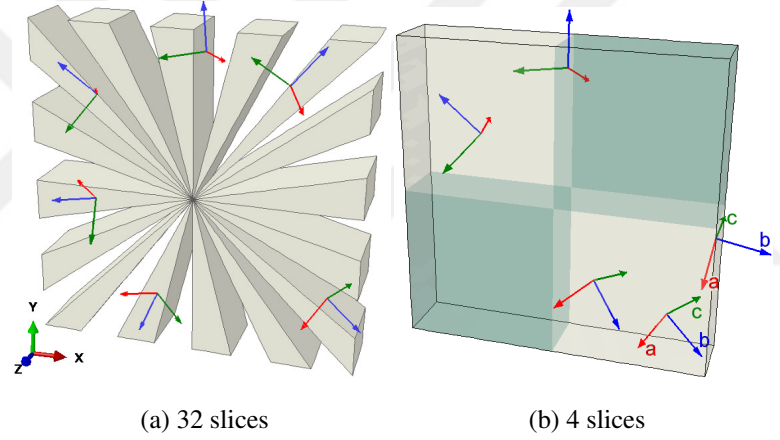


Figure 2.7: Lattice orientations assigned to crystalline phase slices, red, blue, and green bars corresponds to a , b , and c axes, respectively.

elastic stiffness of the curves increases with the increasing number of slices, whereas opposite relation exists for the elastic stiffness of simple shear loading case. Since the engineering stress-strain curves and the initial stiffnesses converge as seen in Figures 2.8 and 2.9, dividing the spherulite into 32 slices can be considered sufficient to obtain a representative model.

In Figures 2.10 and 2.11, deformed spherulite microstructure of the analyses obtained for approximately 1% and 20% engineering strain states are presented, respectively. Deformations in Figure 2.10 are magnified to a target state of 10% macroscopic strain, whereas deformation scaling is not employed for the deformed shapes in Figure 2.11.

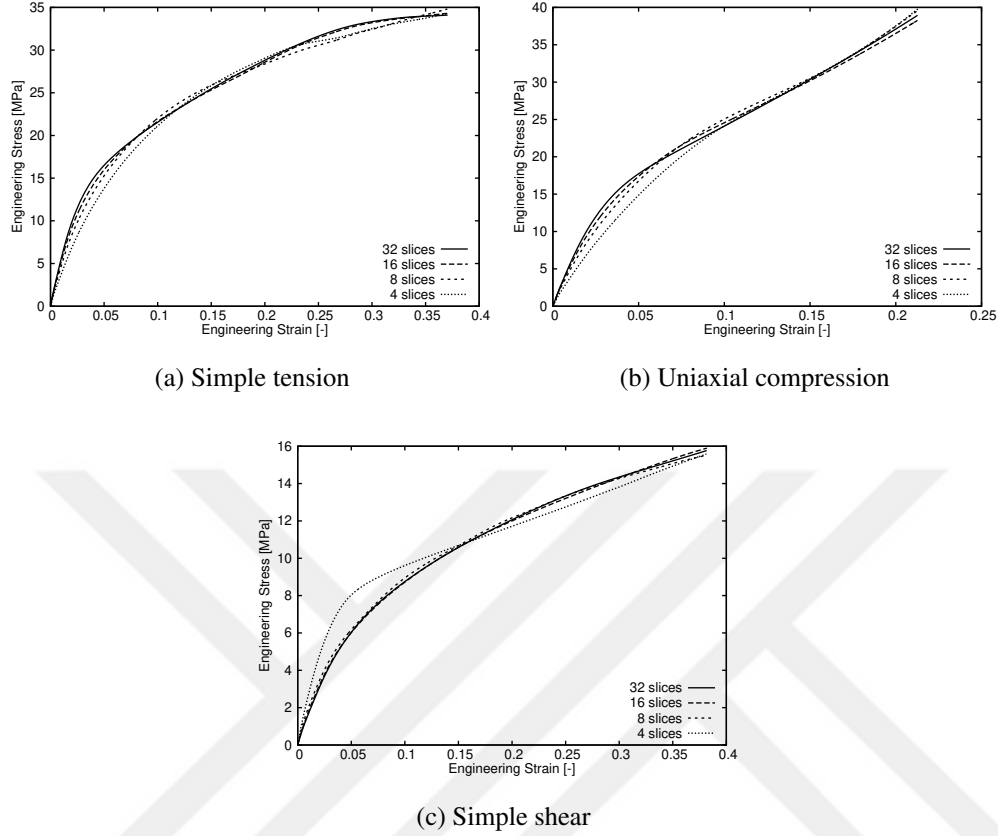


Figure 2.8: Variation of stress-strain curves of single spherulite FEM analyses with number of slices used in the spherulite model

From the deformed shapes in Figure 2.10, for the four slices case, it is seen that most of the macroscopic deformation is accompanied by the amorphous slices, whereas crystalline slices rotate and translate; but for the most part, deform only to satisfy the deformation compatibility at the nucleus. This observation is in agreement with expectations since the elastic moduli of crystalline phase slices could be two orders of magnitude greater than that of amorphous slices as presented in Section 2.2. Based on curved amorphous-crystalline slice boundaries in 32 sliced models in Figure 2.10, it is noted that as the number of slices increases so does the amount of deformation in the crystalline slices, for all loading cases studied. As the slices get smaller, surfaces of the crystalline slices that should satisfy the traction and deformation compatibility with the amorphous phase slices increases. Therefore, crystalline slices begin to deform more consistently with the amorphous phase. Considering the higher elastic stiffness of the crystalline slices, this condition explains the increasing elastic stiff-

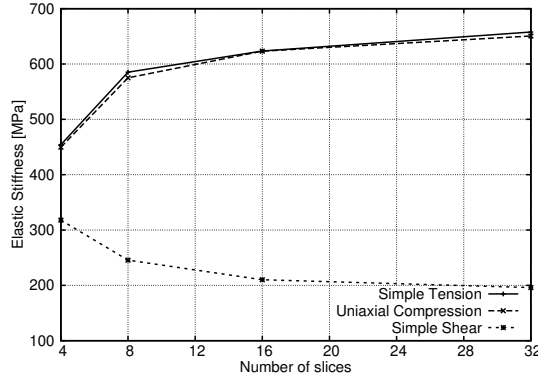


Figure 2.9: Variation of initial elastic stiffness with number of slices for different loading conditions

ness with increasing number of slices, present in Figure 2.9 for simple tension and uniaxial compression. In simple shear, however another noteworthy mechanism is in effect. This mechanism results in a decreasing elastic stiffness as the number of slices increases. For simple shear it is known that maximum tensile and maximum compressive deformations initially occur at lines oriented at +45 and -45 degree angles with the horizontal. For the four-slice model in Figure 2.11(c) it is seen that maximum compression direction passes along the middle of the two crystalline slices, forcing high compressive deformations in crystalline phases. As the number of slices increases, deformation bands that could be identified within amorphous slices relax deformation requirement on the crystalline slices. In Figure 2.11 it is seen that in inelastic stages of deformation, deformations within crystalline slices increases.

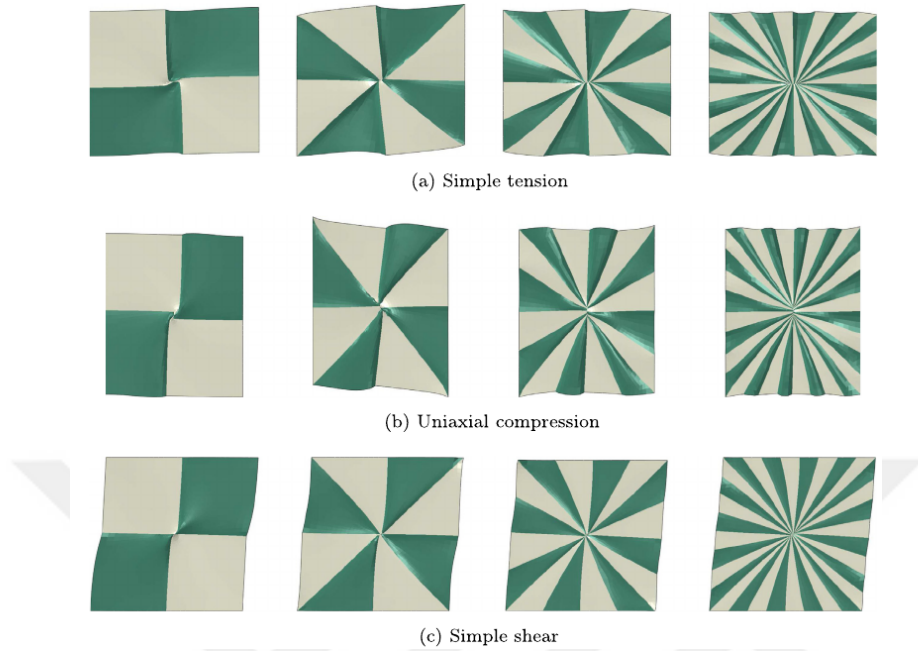


Figure 2.10: Deformed shapes of models for approximately 1% engineering strain for different loading conditions. Deformations are magnified by a factor of 10. Dark and light colored slices correspond to amorphous and crystalline phases, respectively.

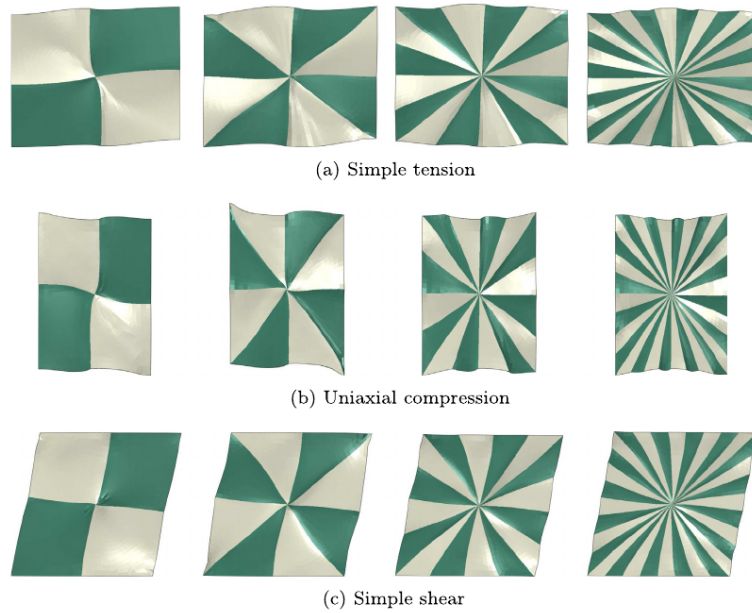


Figure 2.11: Deformed shapes of models at approximately 20% engineering strain for different loading conditions. Deformation scale factor is unity. Dark and light colored slices correspond to amorphous and crystalline phases, respectively.

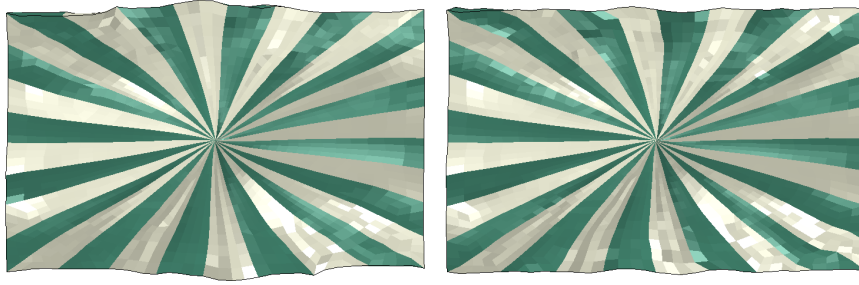
2.3.4 Results

In this section, first the effect of different boundary conditions are investigated. Afterwards, important features of spherulite deformation, and slip system activities under uniaxial tension, uniaxial compression and simple shear loading cases are presented. Finally, the influence of the degree of crystallinity is studied.

2.3.4.1 Influence of the Boundary Conditions

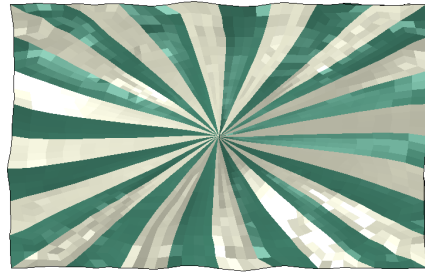
In order to investigate the influence of the boundary conditions on the response, uniaxial tension tests are conducted on a spherulite microstructure with three different boundary conditions. The resulting deformed meshes are shown in Figure 2.12. In Figure 2.12a top and bottom surfaces are free and the displacement is prescribed for the left and right surfaces. For the second boundary condition, which is shown in Figure 2.12b, top and bottom surfaces are periodic and the displacement is prescribed for the left and right surfaces. The last case consists of periodic boundary conditions for top, bottom, left and right surfaces, see Figure 2.12c. The resulting stress-strain curves are given in Figure 2.13 showing no significant effect of the boundary conditions on the macroscopic response of the microstructure.

It should be mentioned that, in general, the periodic boundary conditions provide responses between the uniform traction and uniform displacement boundary conditions, and as the size of a volume element increases, the responses obtained with these boundary conditions approach to each other. However, increasing the dimensions of the disk-like spherulite by a proportional scaling procedure would obviously not change the obtained response. The response would depend on the dimensions of the volume element, if an increase leads to incorporation of other spherulites with different relative placements. Considering a single spherulite model as an RVE and employing periodic boundary conditions would, for example, imply that the polar regions of the spherulites are connected at spherulite boundaries, which is not realistic. Saeb et al. [92] also remark this unrealistic symmetry, and states that the periodic boundary conditions should not be employed with models that discretize a single microstructure, or a part of a microstructure. It should also be stated that, while there



(a) Free Top and Bottom

(b) Periodic Top and Bottom



(c) Periodic

Figure 2.12: Deformed meshes of uniaxial tension test with different boundary conditions. Dark and light colored slices correspond to amorphous and crystalline phases, respectively.

are disk-like spherulites in thin film samples, we do not aim to provide a model for the thin films. In this section, it is found that the response of the model is not significantly influenced by the employed boundary conditions, which is promising for further studies to employ the volume averaging framework with the simplified versions of this model.

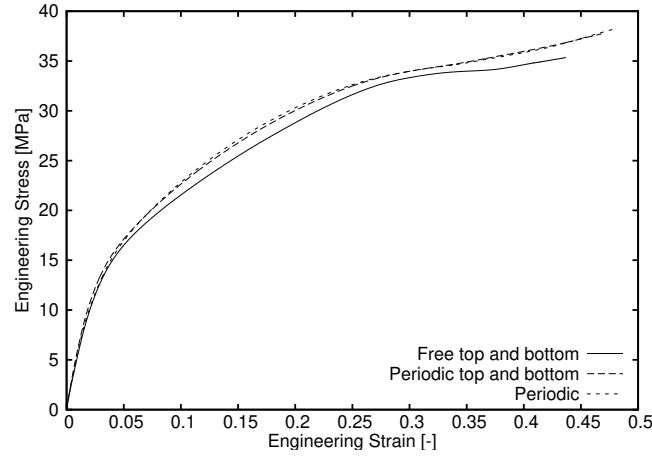


Figure 2.13: Stress-strain curves of uniaxial tension test with different boundary conditions

2.3.4.2 Single Spherulite Tension Test

Lamellae numbering convention employed in this section is presented in Figure 2.14. The load is applied along the horizontal direction. According to Figure 2.2 slices with numbers (1, 16, 8, 9) could be considered as equatorial region lamellae, (2, 3, 4, 6, 7, 10, 11, 12, 14, 15) could be considered as inclined region lamellae and finally, (5, 13) could be considered to be polar region lamellae.

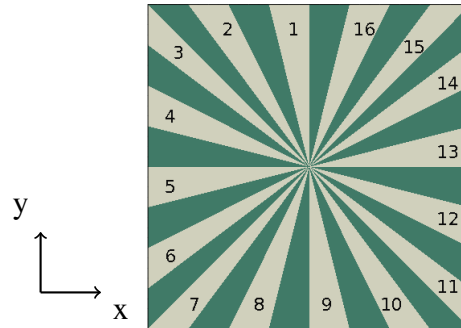


Figure 2.14: Numbering convention for crystalline lamellae slices, x is the tensile loading axis.

In Figure 2.15, variation of certain measures of stress and strain at the initial stage of deformation within a spherulite under simple tension boundary conditions is presented for comparison with important features of spherulite deformation discussed in Oleinik [80]. In Figure 2.15a, radial component of stresses according to the cylindri-

cal coordinate system origin of which is located at the nucleus is presented. It is seen that radial stresses in equatorial regions are compressive (negative). In Figure 2.15b strain component in the loading direction, which is the horizontal direction, is presented where it could be seen that strain in the tensile loading direction in equatorial region are higher than that of polar region.

As discussed in Oleinik [80] important features of spherulite deformation are (i) dependence of stress and strain on the radius, with highest values at the nucleus, (ii) strain component in loading direction is higher in equatorial regions than in polar regions, (iii) tangential and radial stresses in equatorial regions are compressive. Figure 2.15 shows that our model captures these features.

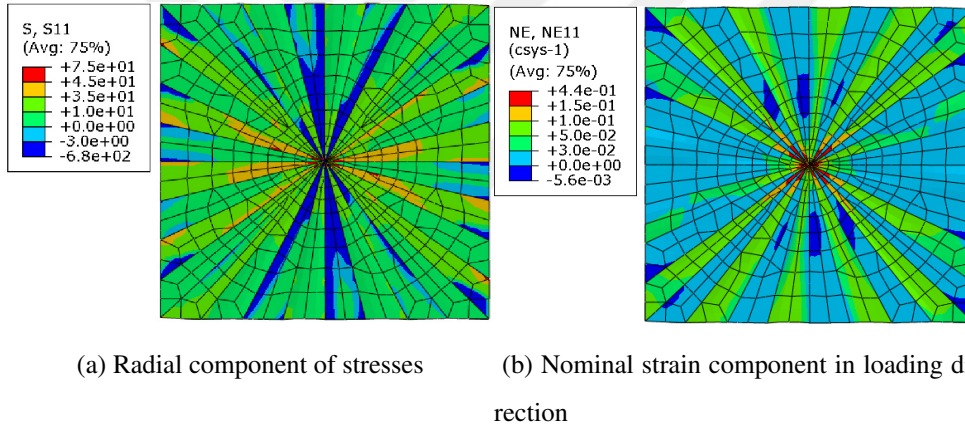


Figure 2.15: Spatial variation of radial stress and nominal strain quantities at 3% engineering strain level for simple tension loading analysis. Tensile straining is in the horizontal direction.

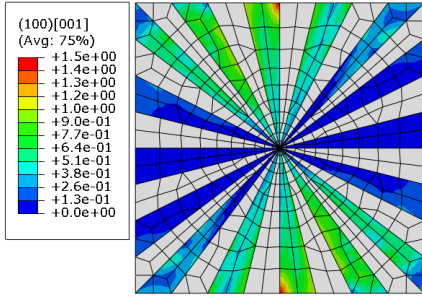
In the following paragraphs activities of crystallographic slip systems; in polar, equatorial and inclined regions of the spherulite are presented for the simple tension loading experiment conducted for 50% crystallinity case. The engineering stress-strain curve of this analysis is presented with the 32 slices label in Figure 2.8a. Spatial variation of the absolute value of the slip $\int \dot{\gamma}^\alpha dt$ at the end of the analysis is given separately for each slip system α in Figure 2.16, where $\dot{\gamma}^\alpha$ is in (2.6).

From Figure 2.16a it is seen that in the equatorial region lamellae (2, 1, 16, 8, 9, 10) there is a large amount of plastic slip in (100)[001] system. As for the lamel-

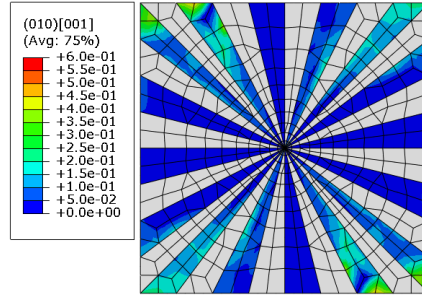
lae belonging to inclined region (3, 4, 6, 7, 11, 12, 14, 15) the amount of slip in (100)[001] system is observed to decrease as the lamellae's location approaches to the polar region. For the polar region lamellae, no slip in (100)[001] system is observed. Additionally, comparison of the maximums of legends of Figure 2.16 reveals that (100)[001] is the most active slip system.

Figure 2.16b shows that (010)[001] slip system is mostly active on lamellae lying in diagonal regions. Very little slip is observed in equatorial region lamellae in this slip system, and no slip is observed in lamellae lying in polar region.

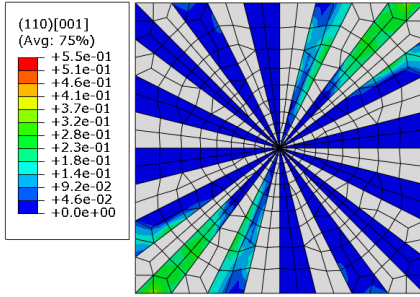
According to Figures 2.16c and 2.16d {110}[001] systems are found active in all of the inclined region lamellae. Slight slip activity is observed in equatorial lamellae close to the inclined region, whereas no slip is observed in polar lamellae.



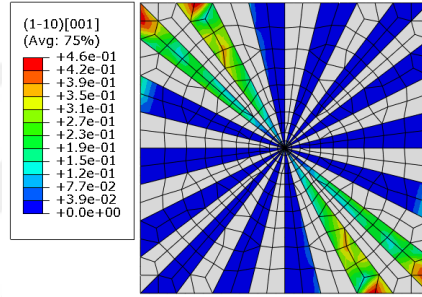
(a) Chain Slip (100)[001]



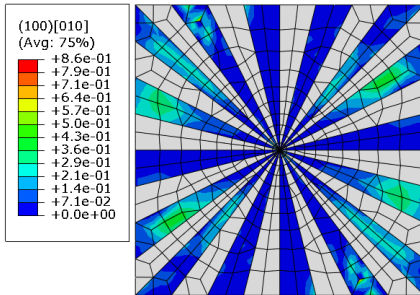
(b) Chain Slip (010)[001]



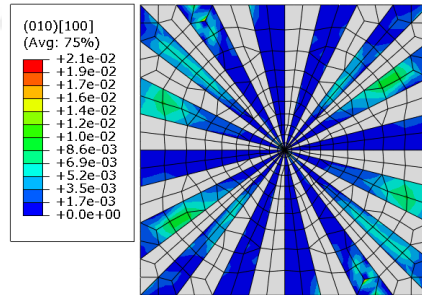
(c) Chain Slip (110)[001]



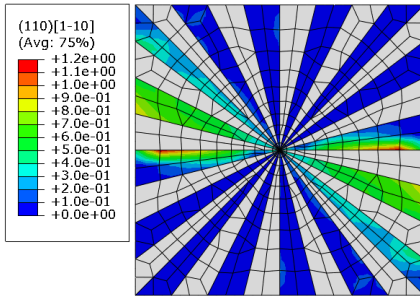
(d) Chain Slip ($\bar{1}10$)[001]



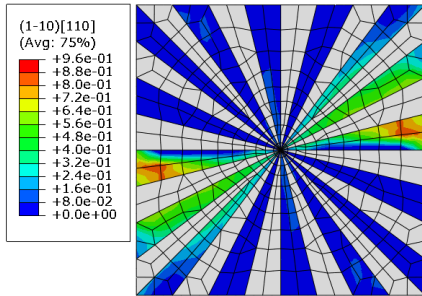
(e) Transverse Slip (100)[010]



(f) Transverse Slip (010)[100]



(g) Transverse Slip (110)[$\bar{1}10$]



(h) Transverse Slip ($\bar{1}10$)[110]

Figure 2.16: Distribution of crystallographic slip magnitudes for simple tension analysis. Loading direction is the horizontal axis.

Comparison of Figures 2.16e and 2.16f reveals that the spatial variation of (100)[010] and (010)[100] transverse slip systems' activity is very similar, with only the amount of slip different. Amount of slips are different due to the difference between the relative slip resistances of these slip systems, where higher magnitudes of slip is observed in (100)[010] system. Both slip systems are found to be active in inclined region lamellae, and be inactive in polar region lamellae. A slight amount of activity in these slip systems is observed in equatorial regions as well. Activity of these systems in polar regions is negligible.

$\{110\}\langle 1\bar{1}0\rangle$ slip systems are found as the major slip systems that are active in polar regions. Additionally, these slip systems are found to be active in all lamellae of the spherulite. Among the lamellae of the spherulite, the highest activity of $\{110\}\langle 1\bar{1}0\rangle$ is in the polar regions.

The results presented in this section for the spatial variation of slip magnitudes are in good agreement with the review presented in [64]. This will be elaborated here. It should be noted that our model considers two main aspects of spherulite microstructure (i) the crystallographic slip systems of polyethylene and (ii) geometrical effects originating from the orientation of lamellae. The high activity found in (100)[001] system in equatorial regions is in agreement with expectations of Hay and Keller [44] based on their well known experimental study on thin film polyethylene specimens. Activity in this system results in rotation around the b lattice axis, and in the case of lamellae lying in equatorial regions it provides the mechanism for chain axis alignment with the tensile loading direction, which is the same as the final configuration of deformation. The decrease of activity in (100)[001] system in inclined region from lamellae with orientation similar to equatorial regions to the lamellae with orientation similar to polar regions are also in agreement with the cited study. Some activity of (010)[001] system in inclined regions is also expected by Hay and Keller [44] as it would provide a mechanism of rotation about a lattice axis to facilitate the alignment of chain direction with the tensile loading direction. It could be noted that critical resolved shear stress of (010)[001] is 2.5 times higher than that of (100)[001] as presented in Table 2.4 and this may be the reason why comparison of Figure 2.16a with Figure 2.16b shows that slip magnitudes of (100)[001] is somewhat higher than that of (010)[001] in inclined region lamellae.

Activity of $\{110\}(001)$ and $\{110\}\langle 1\bar{1}0 \rangle$ in almost all lamellae regions is in agreement with Allan and Bevis [6], where it is stated that slip in these systems is usually favored in the majority of lamellae. Inactivity of all chain slip systems in polar region lamellae and among transverse slip systems $\{110\}\langle 1\bar{1}0 \rangle$ being the major slip system active in polar regions findings are in agreement with Allan and Bevis [6] as well, where it is indicated that chain slip is not favored in polar regions, and one major deformation mechanism in these regions is the $(110)[1\bar{1}0]$ transverse slip.

A similar activity observed for the transverse slip systems $(100)[010]$ and $(010)[100]$, with the same location pattern but different slip magnitudes is expected as explained by Lin and Argon [64]. Since these two slip systems are orthogonal, resolved shear stresses will be the same, and the slip magnitude difference is the result of difference between their critical resolved stresses presented in Table 2.4.

Response of a single spherulite for different loading rates, i.e., $\dot{\epsilon} = 10^{-2}, 10^{-3}, 10^{-4} \text{ s}^{-1}$ and for a cyclic loading at $\dot{\epsilon} = 10^{-3}$ are given in Figure 2.17. The rate dependency of the spherulite microstructure can be seen from Figure 2.17a. Furthermore, the loading-unloading curve in Figure 2.17b clearly shows the dissipative response of the model.

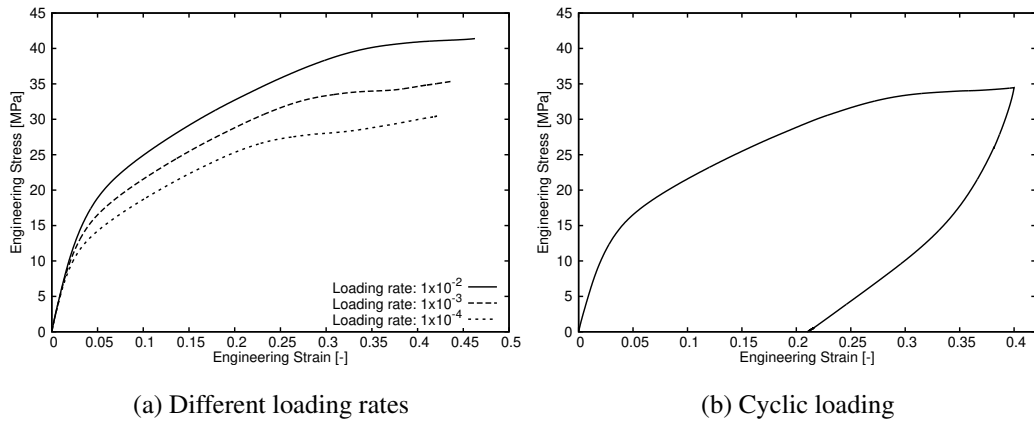


Figure 2.17: Stress-strain curves for different loading rates and a cyclic loading test in simple tension.

2.3.4.3 Single Spherulite Compression Test

In this section, the spatial variation of activities of crystallographic slip systems within the spherulite model is presented for the uniaxial compression loading analysis conducted for 50% crystallinity case. The engineering stress-strain curve of this analysis is presented with the 32 slices label in Figure 2.8b. Spatial variation of absolute magnitude of slip amounts at the end of the analysis is given separately for each slip system in Figure 2.18.

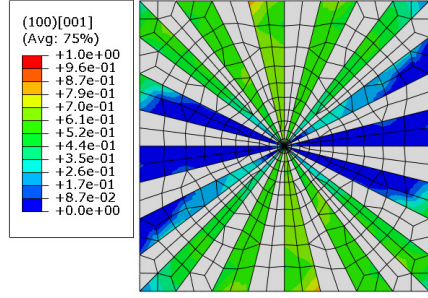
From Figure 2.18 it is seen that (100)[001] chain slip, which has the smallest critical resolved shear stress according to Table 2.4 is by far the most active slip system. Next most active slip system is $\{110\}\langle 1\bar{1}0\rangle$ where activities of which could be seen in Figures 2.18g and 2.18h. Following most active slip systems are (100)[010] and (010)[001] as shown in Figures 2.18b and 2.18e. As expected due to the reason given in Section 2.3.4.2, spatial distribution patterns of (100)[010] and (010)[100] are almost identical.

Bartczak et al. [12] conducted an experimental study on texture evolution of HDPE samples with degree of crystallinity exceeding 70% under uniaxial compression. They have deduced that (100)[001] and (100)[010] slip systems act together and are the dominant slip systems. They have found no evidence of activation of $\{110\}[001]$ slip system. They cannot find much evidence on activation of slip on (010) and (110) planes.

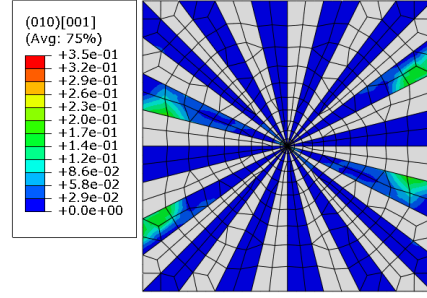
Therefore, our finding of (100)[001] slip system as the dominant system active on most lamellae is in agreement with Bartczak et al. [12]. Next, (100)[010] is also found as a considerably active slip system in our model as well, however it is not active in all lamellae like (100)[001] slip system. Hence, we could not confirm that these slip systems acts together. In agreement with the cited study, our model also indicates low activity in $\{110\}[001]$ slip system. However, the major difference between Bartczak et al. [12] and our study is that our model indicates significant activity in $\{110\}\langle 1\bar{1}0\rangle$ slip system whereas the cited study cannot find much evidence on their activation.

A considerable amount of activity in $\{110\}\langle 1\bar{1}0\rangle$ systems was predicted by our model under simple tension loading as well, as described in Section 2.3.4.2; although in that

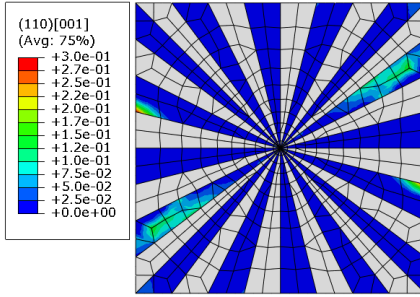
case this activity was in agreement with the experimental study of Allan and Bevis [6]. Study of Allan and Bevis [5, 6] is conducted on thin films where it is stated that the films are composed of portions of "two-dimensional spherulites" and hence are more similar to our proposed model and employed boundary conditions. On the other hand, uniaxial compressive loading experiments of Bartczak et al. [12] were conducted on samples obtained from HDPE plates of 20 mm thickness where the spherulites are reported as fully developed with sizes in the range of 50 μm at the core of the plate. Therefore, reason behind the discrepancy in activity of $\{110\}\langle 1\bar{1}0\rangle$ slip systems with our model and Bartczak et al. [12] may be attributed to the fact that our model is essentially a two-dimensional idealization of the three-dimensional spherulite. Therefore, a three-dimensional spherulite model is studied in Section 2.4.



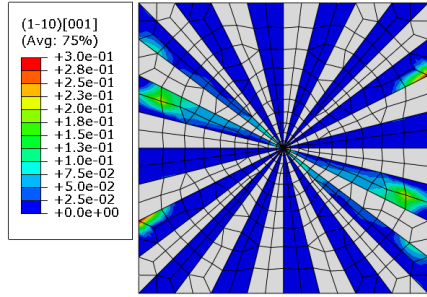
(a) Chain Slip (100)[001]



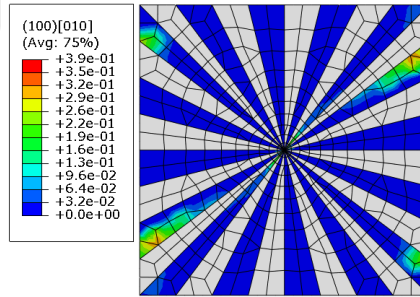
(b) Chain Slip (010)[001]



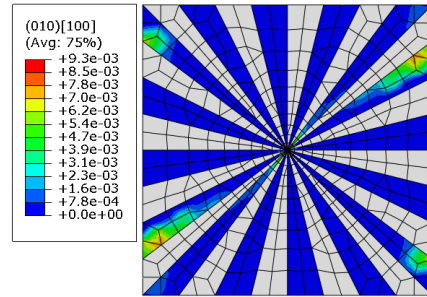
(c) Chain Slip (110)[001]



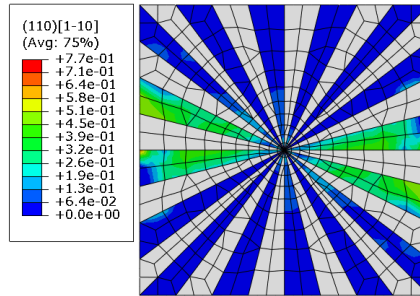
(d) Chain Slip (1 $\bar{1}$ 0)[001]



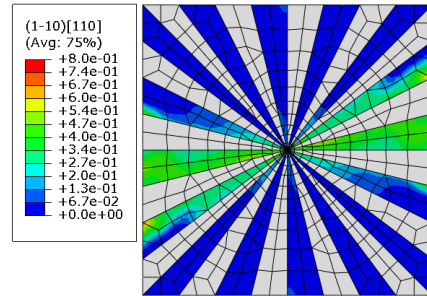
(e) Transverse Slip (100)[010]



(f) Transverse Slip (010)[100]



(g) Transverse Slip (110)[1 $\bar{1}$ 0]



(h) Transverse Slip (1 $\bar{1}$ 0)[110]

Figure 2.18: Distribution of crystallographic slip magnitudes for uniaxial compression analysis. Loading direction is the horizontal axis.

2.3.4.4 Single Spherulite Shear Test

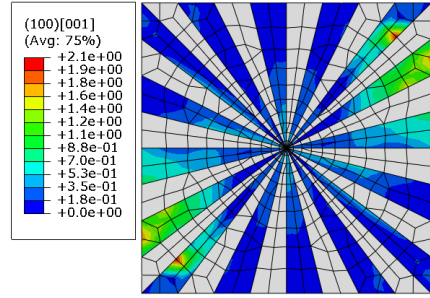
In this section, the spatial variation of activities of crystallographic slip systems within the spherulite model is presented for the simple shear loading analysis conducted for 50% crystallinity case. The engineering stress-strain curve of this analysis is presented with the 32 slices label in Figure 2.8c. Spatial variation of absolute magnitude of slip amounts at the end of the analysis is given separately for each slip system in Figure 2.19.

From Figure 2.19 it could be seen that slip systems could be sorted from most active to least active as $(100)[001]$, $\{110\}\langle 1\bar{1}0 \rangle$, $(100)[010]$, $(010)[001]$, $\{110\}[001]$. As expected due to the reason given in Section 2.3.4.2, spatial distribution patterns of $(100)[010]$ and $(010)[100]$ are almost identical.

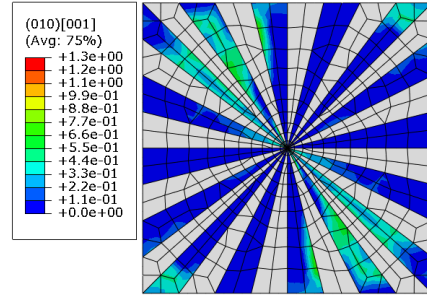
Bartczak et al. [11] has studied texture evolution of high density polyethylene samples with a degree of crystallinity exceeding 70% under simple shear deformation. They have found that most active slip system is the $(100)[001]$ chain slip which is supported by the $(100)[010]$ transverse slip which is followed by the $(010)[001]$ chain slip in activity. Finally, they have observed only trace amounts of $\{110\}[001]$ chain slip.

Our study is in agreement with Bartczak et al. [11] on the most active slip system being $(100)[001]$, and the order of the activities of other slip systems except the $\{110\}\langle 1\bar{1}0 \rangle$ transverse slip. While our model indicates that $\{110\}\langle 1\bar{1}0 \rangle$ system is the second most active system, see Figures 2.19g and 2.19h, Bartczak et al. [11] does not mention activity of this slip system. Additionally only trace amounts of activity is observed in $(110)[001]$ slip system by Bartczak et al. [11], similar to our study.

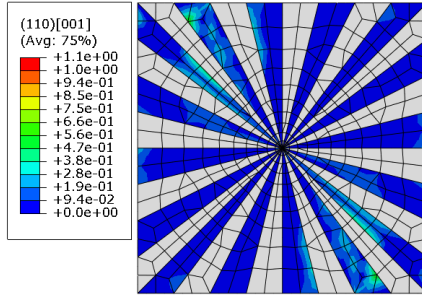
Higher activity in $\{110\}\langle 1\bar{1}0 \rangle$ transverse slip systems predicted by our model compared to Bartczak et al. [11] may be attributed to the essentially 2D idealization of spherulite in our model. Since the discussion presented in Section 2.3.4.3 in activity of $\{110\}\langle 1\bar{1}0 \rangle$ for uniaxial compression loading experiment of Bartczak et al. [12] is valid for this case as well, considering materials and the sample preparation of [11] and [12] are very similar. Therefore, a three-dimensional spherulite model is studied in Section 2.4.



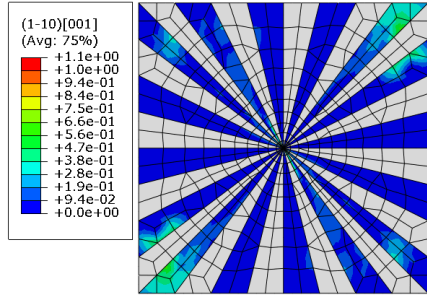
(a) Chain Slip (100)[001]



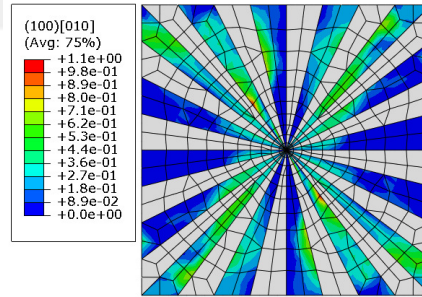
(b) Chain Slip (010)[001]



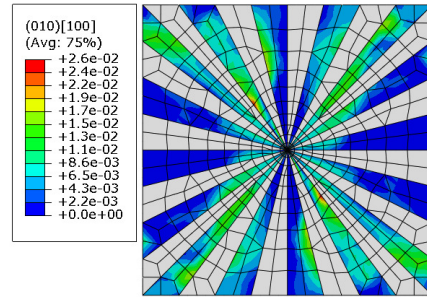
(c) Chain Slip (110)[001]



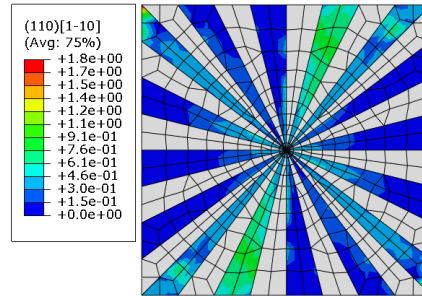
(d) Chain Slip ($\bar{1}10$)[001]



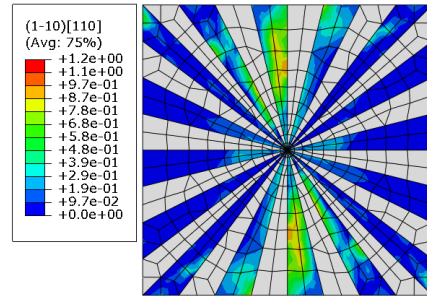
(e) Transverse Slip (100)[010]



(f) Transverse Slip (010)[100]



(g) Transverse Slip (110)[$\bar{1}10$]



(h) Transverse Slip ($\bar{1}10$)[110]

Figure 2.19: Distribution of crystallographic slip magnitudes for simple shear analysis.

2.3.5 Effect of the Degree of Crystallinity

Crystalline and amorphous phase material models are assigned to the slices of the model with 32 slices in order to study the influence of the degree of crystallinity, see Figure 2.20. In this manner the models with volumetric crystallinity degrees of 62.5%, 75% and 87.5% in addition to the studied 50% crystallinity case are obtained. All models employ the same FEM mesh that is presented in Figure 2.3b.

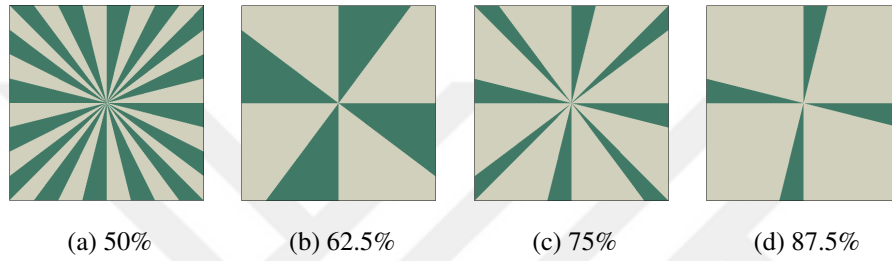


Figure 2.20: Amorphous (dark) and crystalline (light) slice distribution of a single spherulite model for different degrees of crystallinities.

Simple shear, simple tension and uniaxial compression analyses have been conducted. The engineering stress-strain curves are presented in Figure 2.21. From Figure 2.21, it is seen that for all loading conditions, both initial slopes and yield stresses of the curves increase with increasing degree of crystallinity.

In order to qualitatively compare the influence of crystallinity ratio on the yield stress and the initial modulus, the simple tension experiment results of Crist et al. [25] are employed. Figure 2.22 presents a comparison of the yield stress and the Young's modulus obtained from the curves in Figure 2.21a according to the method employed in Crist et al. [25] for the engineering stress-strain curves of simple tension tests. Young's modulus is the slope of the initial tangent to the engineering stress-strain curve, while the yield stress is the stress corresponding to the point of intersection of the initial tangent and the tangent to the curve at approximately 20% strain. It is seen that the yield stress and the Young's modulus are in harmony with the experimental findings of Crist et al. [25]. It should be noted that two of the data points of Crist et al. [25], corresponding to rapidly quenched samples, are not plotted in Figure 2.22 on the grounds that they would not provide the required time for formation of proper

spherulites.

As explained by Oleinik [80], the yield stress of PE is in reality a function of the lamellae thickness and not the degree of crystallinity. A close experimental relationship between the yield stress and crystallinity is due to the direct correlation between the crystallinity and the lamellae thickness. Monolithic nucleation of screw dislocations is considered to explain the increase of yield stress with lamellae thickness [13], and Argon et al. [7] proposed two additional nucleation modes that are independent of lamellae thickness to explain the constant plastic resistance beyond a lamellae thickness of 40 nm. Since our FEM model does not take the lamellae thickness into account in the employed crystal phase material model, it might be deduced that construction of a structure composed of slices as shown in Figure 2.20 is representative for the influence of the lamellae thickness on the yield stress. However, this could simply be related to the increase of the volume fraction of the crystalline phase, since the constitutive models of semicrystalline polymers [109, 77] that employ quite similar crystal phase material models with this study also show a similar increase of the yield stress with increasing volume fraction of the crystalline phase.

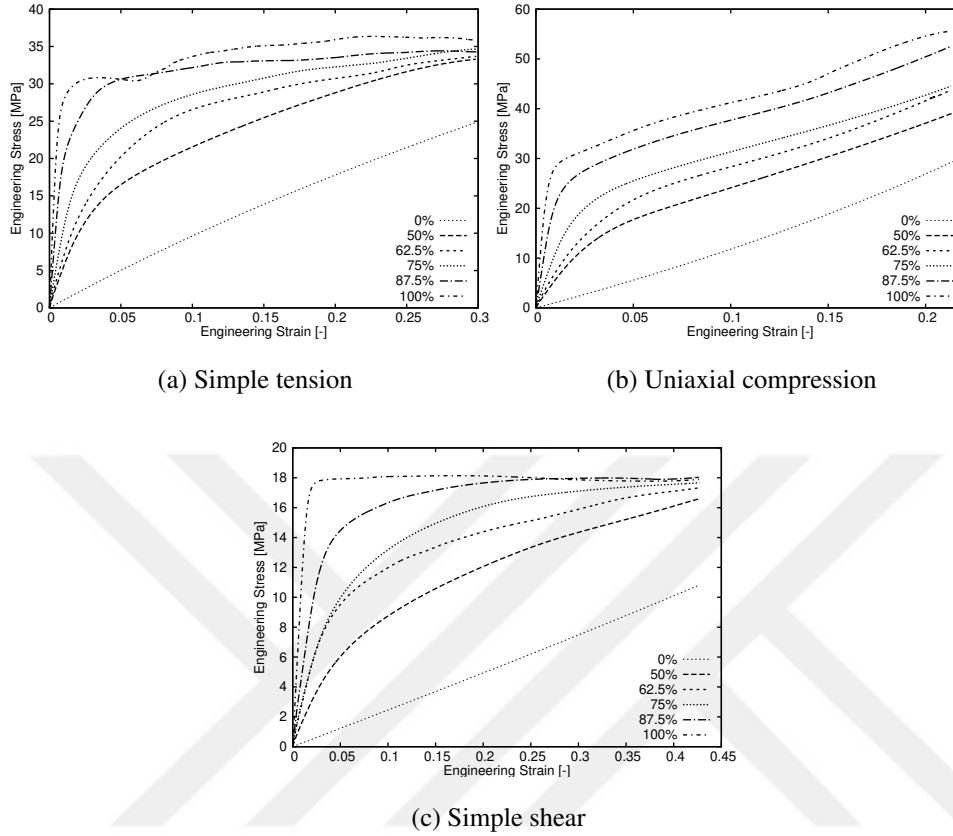


Figure 2.21: Variation of engineering stress - strain curves of single spherulite FEM analyses with volumetric degree of crystallinity used in the spherulite model

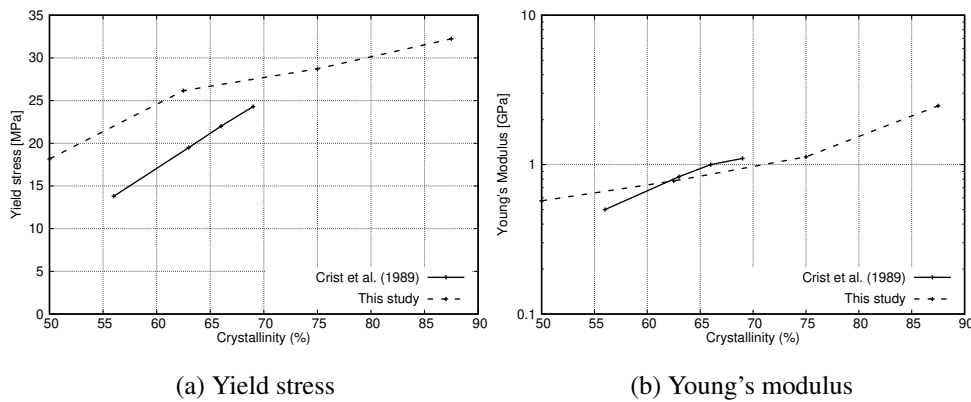


Figure 2.22: Comparison of variation of yield stress and Young's modulus obtained from simple tension analyses with degree of crystallinity to findings of Crist et al. [25]

2.3.6 Discussion

In this study, a FEM based model of a spherulite microstructure is proposed for semicrystalline polymers. Simple tension, uniaxial compression, and simple shear loading analyses are conducted with the model for varying number of slices and degree of crystallinity. The ability of the model to capture important spherulite deformation mechanisms is validated via stress and strain distribution characteristics and relative activities of crystallographic slip systems with the studies in the literature. The limited number of slices employed in this study results in many orders of magnitudes thicker crystalline and amorphous phase layers compared to the actual spherulite. Nevertheless, it is found that for the case with 32 slices, the proposed model is a reasonable idealization since the response of the model does not significantly change with increasing number of slices. The rate sensitivity and the dissipative nature of the model are shown through simulations with different loading rates and a cyclic loading. The correlation between the yield stress and the initial modulus with crystallinity is similar to the experimental results of Crist et al. [25].

Although the spatial distribution of the crystallographic slip system activities of this study are in general agreement with the experimental studies in all deformation modes studied, the high activity of $\{110\}\langle 1\bar{1}0\rangle$ system in simple compression and shear is not in good agreement with the experimental studies. Similar level of $\{110\}\langle 1\bar{1}0\rangle$ activity is observed in this study in simple tension and compression. On the other hand, while the significant activity of $\{110\}\langle 1\bar{1}0\rangle$ is expected for disk-like spherulites by Allan and Bevis [6] in simple tension, the activity of $\{110\}\langle 1\bar{1}0\rangle$ are not reported in experimental studies on bulk specimens composed of three-dimensional spherulites in simple compression [12] and shear [11] deformation modes. A three-dimensional spherulite model is studied in Section 2.4 in order to assess the effect of the disk-like spherulite idealization on the activity of $\{110\}\langle 1\bar{1}0\rangle$ system.

2.4 Three-dimensional Spherulite Model

A three-dimensional finite element model of a spherulite is constructed by dividing a cube of 100 micron size into amorphous and crystalline regions. Regions of the

crystalline phase, referred to as lamellae from now on, of the model are presented in Figure 2.23. All lamellae originate from the nucleus, which is at the geometric center of the cube. There are 26 lamellae in the model, and these could be grouped into three according to their shapes, as shown in Figure 2.23. There are 6, 8 and 12 lamellae in the lamella group FC, C and ME, respectively. Lamellae of FC, C and ME are directed towards the face centers, corners and mid points of the edges of the cube, respectively. Lamellae shapes of each group are selected to enable modeling of arbitrary crystallinity ratios without overlap of any lamellae. Lamella of type FC, C and ME are composed of 1, 3 and 2 square based pyramids respectively. Regardless of the lamellae group they belong, edge lengths of the pyramid bases are equal. Therefore, if a lamella that belongs to the group FC has a volume V , then each lamella of the group C and ME has volumes $3V$ and $2V$, respectively.

One distinct feature of the model is the overly thick crystalline lamellae (about 10-20 μm), compared to the thin crystalline lamellae of actual PE spherulites (about 10-20 nm). Therefore, a single lamella in the model is considered as the average of many lamellae of a real spherulite of semicrystalline PE.

Quadratic tetrahedral finite elements C3D10 and C3D10H of the Abaqus element library are used for the crystalline and the amorphous regions, respectively. In the amorphous regions, hybrid elements (C3D10H) are used for their better performance in almost incompressible material models. The 8-chain elasticity and the crystal plasticity models and material parameters presented in Section 2.2 are employed in the amorphous and crystalline regions, respectively.

2.4.1 Orientation of the Crystal Lattice

Since the polyethylene (PE) spherulite is considered, the orientation of crystallographic axes is selected to satisfy the following criteria: (i) the crystal lamella growth is in the b lattice axis direction [64], (ii) the interface between the amorphous phase and the lamellae is the $\{201\}$ crystallographic plane [35]. For the first condition, a spherical coordinate system, origin of which is located at the nucleus is employed. Then, b lattice axis is oriented in the direction of the radial basis vector e_r of the coordinate system. For the second condition, it should be noted that any lamella of the

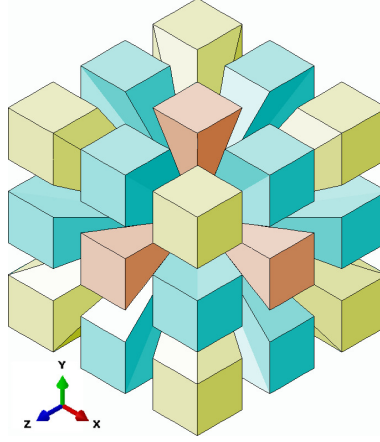


Figure 2.23: Lamellae of spherulite model categorized according to the lamella geometry. Red: lamella FC, Yellow: lamella C, Blue: lamella ME

model is bounded by multiple planar surfaces that form interfaces with the amorphous phase, see Figure 2.23. In order to impose (ii) for all of the interfaces of a lamella, each lamella is divided into several regions, and orientations are defined separately with basis vectors of different spherical coordinate systems. Lamellae belonging to groups FC, C and ME (shown in different colors in Figure 2.24) are divided into 2, 6 and 5 regions, respectively. The basis vector e_θ of the spherical coordinate system is selected to define the orientation of the chain. For each lamella region, orientation of the spherical coordinate system is selected such that, on the interface plane, the interface plane normal vector and the e_θ basis vectors are aligned. Then, to set the desired angle between the interface normal and the chain direction, the coordinate system is rotated 35° around the b lattice axis, i.e. the e_r basis. In this case, the interface normal is aligned with the normal of the $\{201\}$ plane of the lattice.

The explained orientation assignment procedure employs different coordinate systems in each region and unavoidably results in discontinuous orientation jumps between regions. However, the orientation of the b axis is continuous throughout the lamellae, and the discontinuity at region boundaries is only in the a and c axes. In actual PE spherulites, orientations of the a and c change along the lamella growth direction, and span 360° domain, since the crystal growth includes a twist of the lamellae along the b axis of the ribbon shaped lamellae. Considering the distance between the integration points of the model is large compared to the twist period, the

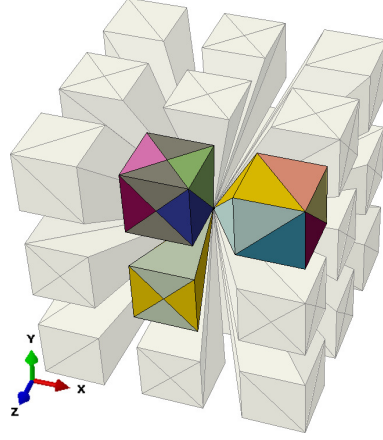


Figure 2.24: Regions of lamellae. Regions that use the same spherical coordinate system for the orientation assignment is shown in same colors. For the sake of clarity only regions of the three lamellae are colored.

changes in the a and c lattice axes orientations of the lamellae, admittedly discontinuous, are considered as representative of the actual crystalline texture of the PE spherulite morphology.

2.4.2 Boundary Conditions

The response of the model is investigated under simple tension and compression. Boundary conditions employed in these loadings cases are presented in Figure 2.25. In Figure 2.25, 1/8th of the model is hidden in order to present the boundary conditions at the nucleus and inside the spherulite. The nucleus is fixed in all directions. In order to prohibit any rigid body rotation, displacements in the y -direction are constrained on the axis that passes through the nucleus and parallel to the z -direction, see Figure 2.25. A uniform displacement along the x -direction is assigned to the faces of the cube having normals parallel to the x -direction. The loading is applied with a deformation control at a constant engineering strain rate of $1 \times 10^{-3} \text{ s}^{-1}$. It is aimed to model a single spherulite, and not the spherulitic microstructure in a bulk specimen. Therefore, instead of the boundary conditions more suitable for homogenization, such as the periodic boundary conditions, boundary conditions similar to that of the disk-like spherulite model in Section 2.3.1 are employed.

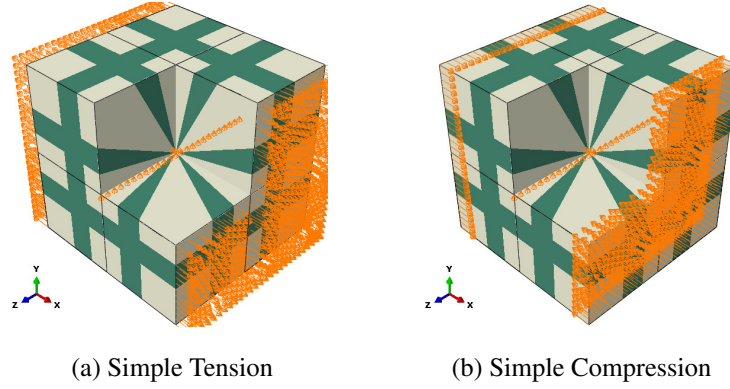


Figure 2.25: Boundary conditions for the simple tension and the simple compression load cases. Dark and light regions correspond to the amorphous and crystalline phases, respectively. 1/8th of the model is hidden.

2.4.3 Results

In this section, the conformity of the model with the important features of the spherulite deformation is investigated. To this end, stress, strain and slip system activity distribution within the spherulite model are reported and compared with the studies in the literature for simple tension and simple compression. Then, influences of the degree of crystallinity on the initial Young's modulus and the yield strength are investigated and the predictions of the model are compared with the experimental results.

2.4.3.1 Tensile Loading of a Single Spherulite

As discussed in Section 2.1, the strain distribution within a spherulite depends strongly on the orientation of lamellae with respect to the loading direction. In the literature, a spherulite is divided into three regions according to the lamella orientation with respect to the tensile loading direction [64] as presented in two dimensions in Figure 2.2. These regions are referred to as the equatorial, the polar and the inclined regions. They could also be defined as follows: In the polar regions, the lamella growth direction, i.e., the b lattice axis, is nearly parallel to the loading direction. In the equatorial regions, on the other hand, the lamellae growth direction is nearly perpendicular to the loading direction. Regions that neither belong to the equatorial nor

to the polar regions belong to the inclined regions. Figure 2.26 presents crystalline regions of the proposed model categorized into the equatorial, polar and inclined regions, considering that the loading is applied along the x-axis in this study.

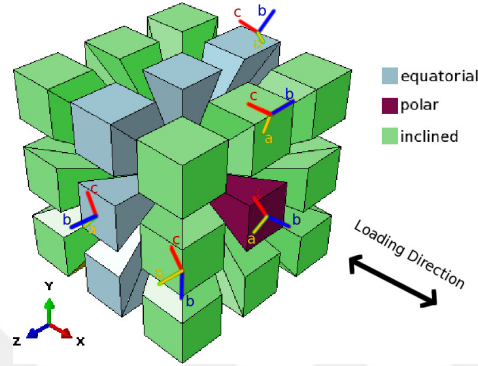


Figure 2.26: Equatorial, polar, and inclined lamellae of the spherulite model for the tensile loading along x-axis, and the representative lattice directions in the lamellae

Figure 2.27 presents the spatial variation of the stress and the strain components within the spherulite at an initial stage of deformation. All strain plots of this study present the components of the nominal strain tensor $\sqrt{\mathbf{F}^T \mathbf{F}} - \mathbf{I}$ defined in the Abaqus documentation [28], where $\mathbf{F}$ and $\mathbf{I}$ are the deformation gradient and the identity tensor, respectively. In all stress plots, the components of the Cauchy stress are presented. A view of the amorphous and the crystalline slices is available in Figure 2.27a. The model has 50% volumetric crystallinity ratio. In Figure 2.27 it is seen that the stress and the strain depend on the distance from the nucleus, while the maximum values are at the nucleus. Strain component in the loading direction is high in the amorphous phase of the equatorial regions. These observations indicate that the deformation begins at the nucleus and spreads through the equatorial regions along the radius. Finally, compressive radial stresses in the equatorial regions are seen in Figure 2.27c. These observations are in agreement with the important features of the spherulite deformation presented in the review article [80].

Figure 2.28 presents the deformed views of the model at an engineering strain of 0.16. Fourteen lamellae are numbered in this figure for future reference in this section. According to Figure 2.26, lamellae 1, 5, 9, 12 are in the equatorial region; 3, 7 are in the polar region and the remaining 2, 4, 6, 8, 10, 11, 13, 14 are in the inclined region. In Figure 2.29, deformation of a selected set of lamellae is presented. Under

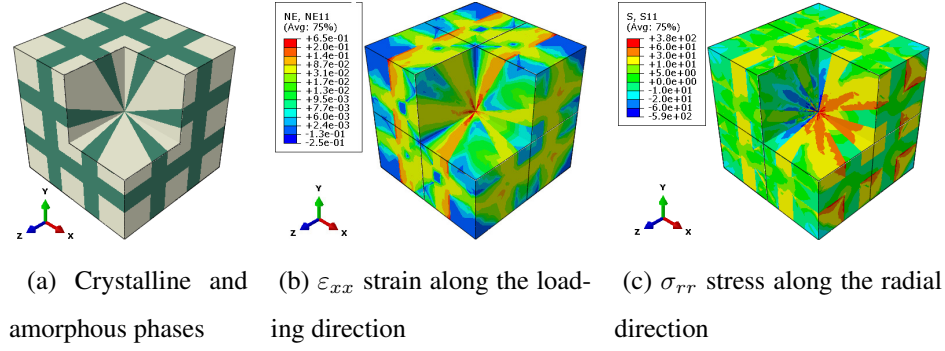


Figure 2.27: Stress and the strain distribution within an undeformed view of the spherulite at 3% engineering strain. In (a) dark and light colored regions are amorphous and crystalline, respectively. In (c), σ_{rr} is with respect to a spherical coordinate system origin of which is at the nucleus.

simple tension, the spherulite model should extend in the loading direction, and contract in the transverse directions. Contraction of the equatorial lamellae, extension of the polar lamellae, and rotation of the inclined lamellae around the nucleus are the noticeable features of the lamellae deformation in Figure 2.29. The polar and equatorial lamellae, composed of the kinematically constrained and strongly anisotropic PE crystals that are oriented according to a relatively complex orientation field, can accommodate significant axial deformation. Significant rotation of the inclined region lamellae (2,4,6,8,10,11,13,14), could be considered as the lamellae-stack rotation phenomenon. Stack rotation is considered as the result of the combination of the interlamellar shear and interlamellar separation deformation modes, which are discussed in the next paragraph, and the chain slip in the crystalline phase [64].

In Figure 2.30, polar rr , $\theta\theta$, $r\theta$ components of the strain field on the two planes in the model are presented. Both planes divide the model into two halves, while one of them is vertical and the other is inclined. The strain components are with respect to cylindrical coordinate systems, the e_z bases of which are aligned with the normal vectors of these planes. Then, the e_r and e_θ bases of the coordinate systems, which rotate with the polar coordinate θ , span these planes. According to Figure 2.26, for the vertical cut plane, the e_θ basis vector is the surface normal of the lamellae in Figure 2.30, while for the inclined cut plane the lamellae normals

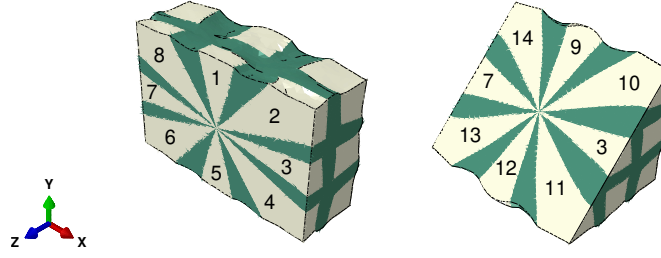


Figure 2.28: Deformed view at 0.16 engineering strain. Deformations are scaled by a factor of 2. Dark and light colored regions are the amorphous and the crystalline phases, respectively. The first view is a vertical cut through the nucleus. The second view is an inclined cut with a normal vector of $(0,1,1)$.

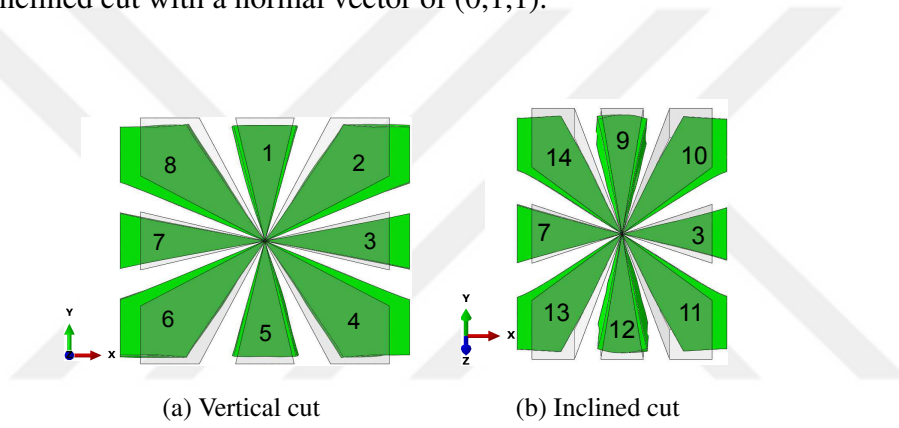


Figure 2.29: Undeformed (translucent) and deformed (solid) views of the selected lamellae of the model seen on the cuts passing through the nucleus. Viewing axes are aligned with the normal vector of the cross sections. Deformations are in true scale. In (b) cut plane normal is $(0,1,1)$.

may have components in the e_z basis as well. Nevertheless, for both the vertical and the inclined cuts, the projection of the lamellae normals to the cut-planes are the e_θ bases. Therefore, considering the amorphous region, the $\theta\theta$ and $r\theta$ components of the strain fields in Figure 2.30 indicate interlamellar separation and interlamellar shear deformation modes, respectively. Interlamellar separation is observed in the equatorial region in Figure 2.30(a), while the interlamellar shear is dominant in the inclined region in Figure 2.30(b). Then, two of the three well-known deformation modes of the amorphous phase [64] are clearly observed in the model. Moreover, the locations of these deformation modes within the spherulites are in agreement with the literature [80]. The third deformation mode of the amorphous phase, the stack

rotation, is present in the model when the lamellae of the model are considered as the average of many thin lamellae of the actual spherulite.

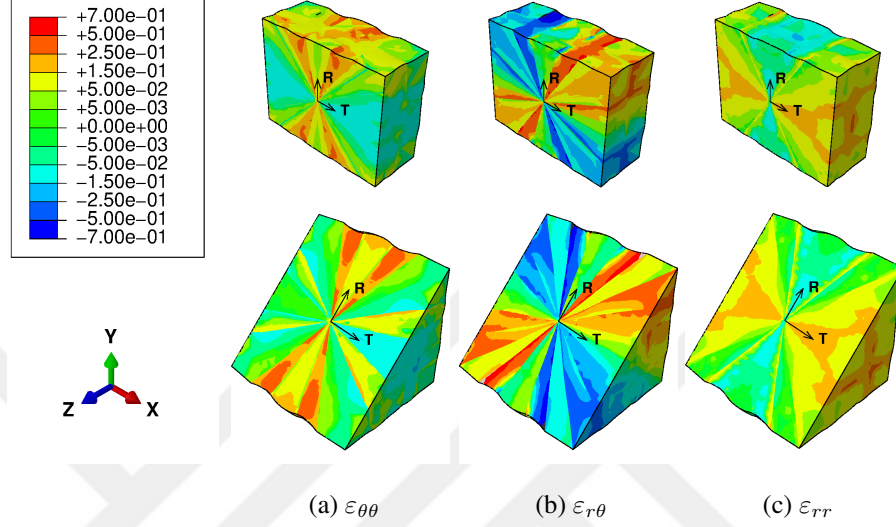


Figure 2.30: Distribution of the polar strain components at 0.16 engineering strain. Strain components are in cylindrical coordinate systems, where origins are at the nucleus and z directions are along the normals of the cut planes. Normal vector of the inclined cuts are (0,1,1) in the Cartesian coordinate system shown.

Figure 2.31 presents the evolution of the strain within the model along the loading direction at three strain levels. Initially, the highest strains are in the amorphous phase in the equatorial region. The strain accommodated in the crystalline phase, which is mostly due to the plastic deformation, approaches to that of the amorphous phase as the deformation increases. Including the maximum deformation stage, the amorphous phase of the model accommodates higher strains than the crystalline phase. In all strain levels, strains are greater in the polar lamellae than in the equatorial lamellae. Additionally, in the final deformation stage, in Figure 2.31(c), highest strains are in the polar region lamellae (3 and 7) among all the lamellae presented. This is the result of the displacement boundary conditions of our model (i.e., the fixed boundary condition at the nucleus and the applied displacements at the free surface of the polar region), since the polar region lamellae have to fully accommodate the macroscopic strain component in the loading direction. Figure 2.32 presents distribution of the

Cauchy stress component in the loading direction at uniaxial engineering strain equals to 0.16. Among lamellae, the stresses are highest in the equatorial region and lowest in the polar region. Therefore, since the highest strains are in the polar lamellae as well, the softest lamellae of the model are in the polar region. This observation is not in good agreement with the general view in literature, where the polar regions are considered to possess the highest deformation resistance [64, 80].

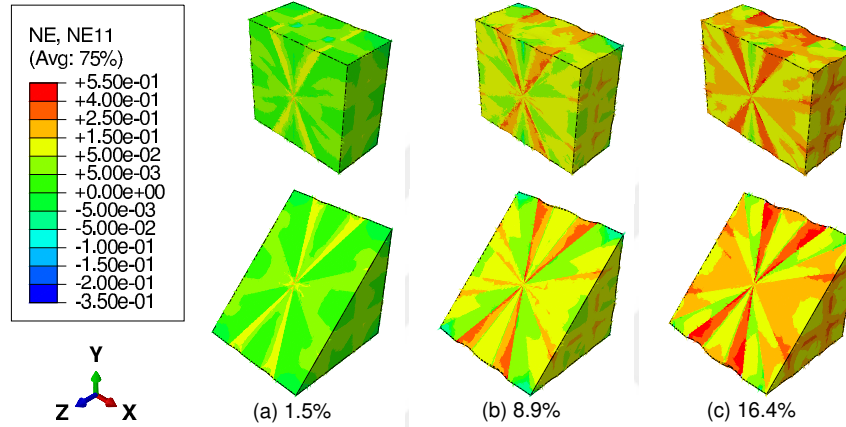


Figure 2.31: Distribution of the nominal strain component along the loading direction ϵ_{xx} , for the simple tension analysis at different strain levels.

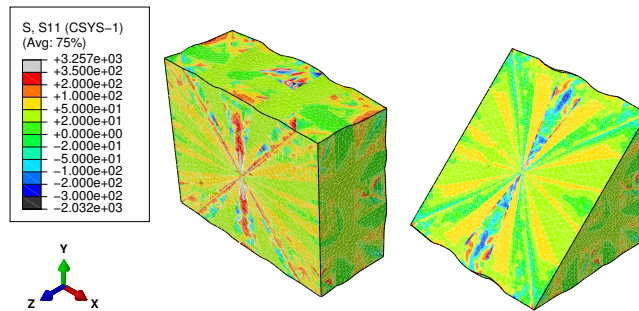


Figure 2.32: Distribution of the Cauchy stress component in the loading direction for the simple tension analysis. Loading is in the x direction. The first view is a vertical cut through the nucleus. The second view is an inclined cut, with a normal vector of $(0,1,1)$, through the nucleus.

In crystal plasticity, plastic slip depends on the stress tensor and the orientation of the slip systems, according to the viscoplastic flow rule, given in (2.6) and (2.7). Equation (2.7) is a linear relationship between the Schmid shear stresses, and the $\tau_{\alpha\beta}$

components of the Kirchhoff stress tensor

$$\boldsymbol{\tau} = \tau_{\alpha\beta} \mathbf{e}_\alpha \otimes \mathbf{e}_\beta \quad (2.8)$$

computed in the basis formed by the lattice axes in the deformed configuration. In (2.8), summation over the repeated indices is implied where $\alpha = a, b, c$, and $\beta = a, b, c$. $\mathbf{e}_a = \mathbf{R}^e \mathbf{a}$, $\mathbf{e}_b = \mathbf{R}^e \mathbf{b}$, $\mathbf{e}_c = \mathbf{R}^e \mathbf{c}$ are the unit vectors in the deformed configuration, and $\mathbf{a}$, $\mathbf{b}$, $\mathbf{c}$, are respectively the unit vectors along the a , b , c , lattice axes in the intermediate configuration. Notice that while computing the plastic slips, particularly in (2.7), we have ignored the elastic distortion of the crystal lattice by considering the elastic stretch of the stiff crystal as negligible, following [29]. Consequently, the basis formed by the $\mathbf{e}_a$, $\mathbf{e}_b$, $\mathbf{e}_c$ vectors is orthonormal, similar to $\mathbf{a}$, $\mathbf{b}$, $\mathbf{c}$. The coefficients of the Kirchhoff stress components $\tau_{\alpha\beta}$ in this linear relationship are given in Table 2.5. For example, for the (100)[001] slip system, substitution of $\mathbf{s} = \mathbf{e}_c$ and $\mathbf{m} = \mathbf{e}_a$, and (2.8) into (2.7) we obtain the Schmid stress for the (100)[001] system as $\tau = \tau_{ac}$. Another example could be given for the (110)[001] slip system, which is the third slip system in Table 2.5. In this case, $\mathbf{s} = \mathbf{e}_c$ and $\mathbf{m} = 0.55 \mathbf{e}_a + 0.83 \mathbf{e}_b$. Then we find $\tau = 0.55 \tau_{ac} + 0.83 \tau_{bc}$. Notice that, while calculating the unit vectors, particularly along the $[110]$ and $[1\bar{1}0]$ directions, the lattice constants of the PE $a = 7.4 \text{ \AA}$, $b = 4.9 \text{ \AA}$, $c = 2.5 \text{ \AA}$ [109] are employed.

In the polar region, lamellae extend along the $\mathbf{b}$ lattice axis under tensile loading. According to Table 2.5, τ_{bb} stress component originating from the extension or compression along the $\mathbf{b}$ lattice axis activates only the $\{110\}\langle 1\bar{1}0 \rangle$ slip systems. Therefore, in our model $\{110\}\langle 1\bar{1}0 \rangle$ operates as the sole plastic deformation mechanism for the extension of the polar region lamellae in the loading direction. $\{110\}\langle 1\bar{1}0 \rangle$ also facilitates the contraction of the equatorial lamellae in the model, along the transverse directions of the loading axis. Considering the low stresses obtained in polar region lamellae, activity of $\{110\}\langle 1\bar{1}0 \rangle$ is investigated in detail when discussing the slip system activities.

Slip system activities at an engineering strain of 0.16 for the spherulite with 50% crystallinity ratio are reported below. As the measure of activity, the accumulated plastic slip

$$\Gamma^\alpha(t) = \int_0^t |\dot{\gamma}^\alpha| dt \quad (2.9)$$

Table 2.5: Contribution factors of the Kirchhoff stress components resolved along the deformed lattice axes to the Schmid shear stress that activates the slip systems

Slip System	τ_{ac}	τ_{ab}	τ_{bc}	τ_{aa}	τ_{bb}	τ_{cc}
(100)[001]	1.	0	0	0	0	0
(010)[001]	0	0	1.	0	0	0
(110)[001]	0.55	0	0.83	0	0	0
(1 $\bar{1}$ 0)[001]	0.55	0	-0.83	0	0	0
(100)[010]	0	1.	0	0	0	0
(010)[100]	0	1.	0	0	0	0
(110)[1 $\bar{1}$ 0]	0	0.39	0	0.46	-0.46	0
(1 $\bar{1}$ 0)[110]	0	-0.39	0	0.46	-0.46	0

is employed for each slip system α , where $\dot{\gamma}^\alpha$ is the slip rate computed according to (2.6). In Table 2.6, relative activities of each slip system throughout the equatorial, polar, inclined regions and the whole model are presented, separately. The relative activity of the slip system α in the region Ω_r is

$$G_r^\alpha(t_{max})/G_r(t_{max}), \quad r = e, p, i, a, \quad (2.10)$$

where $G_r^\alpha(t) = \int_{\Omega_r} \Gamma^\alpha(t) dV$ is the total accumulated slip of the slip system α at time t , and $G_r(t) = \sum_{\alpha=1}^8 G_r^\alpha(t)$ is the total accumulated slip of all slip systems, in the region Ω_r , and t_{max} is the time when the maximum strain is reached at the end of the analysis. $\Omega_e, \Omega_p, \Omega_i$ are the equatorial, polar and inclined regions of the model, and $\Omega_a = \Omega_e \cup \Omega_p \cup \Omega_i$ is the domain of the whole model. In Table 2.6, combined activity of the three most active slip systems amounts to 83% of all crystalline slip in the model. These systems, from the most active to the least, are (100)[001], (100)[010] and $\{110\}\langle 1\bar{1}0 \rangle$. These slip systems, respectively, are the dominant slip systems of the equatorial, inclined and polar regions. (100)[010] is significantly active only in the inclined regions, on the other hand, activity of $\{110\}\langle 1\bar{1}0 \rangle$ and (100)[001] are significant throughout the model. While these slip systems cover the 97% of the slip in both the equatorial and the polar regions, they cover only the 77% of the slip in the inclined regions. Among the remaining three slip systems, $\{110\}[001]$ and (010)[001] chain slips are responsible from the 11% and 10% of the slip, respectively, in inclined

regions. The remaining system, (010)[100], has negligible activity throughout the model. Since (010)[100] and (100)[010] are orthogonal to each other, their coefficients in Table 2.5 are the same, and the observed activity difference between them is related with the difference between their slip resistances in Table 2.4.

Table 2.6: Relative activities $G_r^\alpha(t_{max})/G_r(t_{max})$ of slip systems within the equatorial, polar, inclined regions and the whole model at 0.16 engineering strain

	(100)[001]	(010)[001]	(110)[001]	(1 $\bar{1}$ 0)[001]	(100)[010]	(010)[010]	(110)[1 $\bar{1}$ 0]	(1 $\bar{1}$ 0)[110]
Equatorial region (%)	77.1	0.5	0.5	1.0	0.9	0.0	9.9	10.1
Polar region (%)	9.0	0.6	0.5	0.5	1.8	0.0	43.8	43.8
Inclined region (%)	19.0	10.2	5.6	5.7	42.8	1.1	7.7	7.9
Entire model (%)	32.4	7.5	4.2	4.4	31.3	0.8	9.6	9.8

In Figure 2.33, variation of the accumulated slip in the model with the macroscopic strain is presented for each slip system, separately. The (100)[001] is the first system that is activated, and its rate of activity decreases near the end of the analysis. Next, (100)[010] is activated and its rate of activity increases with strain. {110} $\langle$ 1 $\bar{1}$ 0 $\rangle$ activity starts approximately at the same macroscopic strain level with (100)[010], and its rate of activity is constant. The activity of the remaining slip systems, with higher slip resistances, (010)[001], {110}[001] and (010)[100] start later, at 0.04 macroscopic strain. The stiffness of the model decreases, as the more slip systems activate. Order of the slip systems from the most active to the least does not change throughout the analysis.

In Figure 2.34a, the variation of the total accumulated slip in the regions of the model is presented. The rate of the accumulated slip is approximately constant in the polar and equatorial regions, while it increases in the inclined region. The accumulated slip is initially the highest in the equatorial region, then it quickly increases in the inclined region, and the inclined region passes the equatorial region. According to Figure 2.34a, at the end of the analysis, more than 70% of the total accumulated slip is in the inclined region. In the actual spherulites, the inclined region occupies

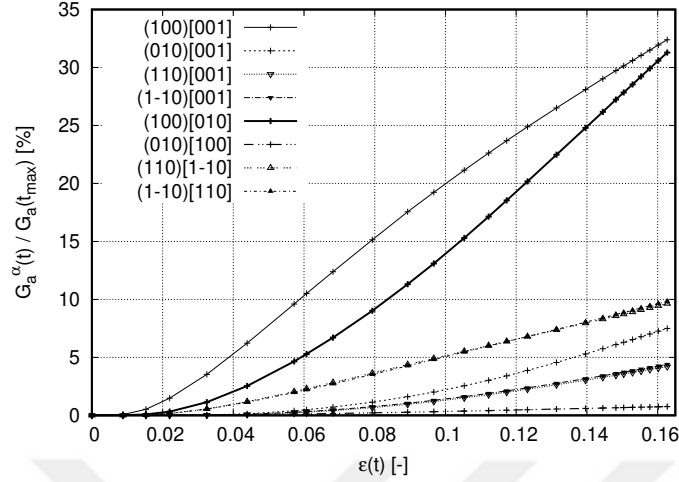


Figure 2.33: Variation of the accumulated slip of each slip system in the model with the macroscopic strain. Vertical axis is normalized by the value of the total accumulated slip in the model at the 0.16 engineering strain.

the greatest volume [5]. This is the case for the model as well, such that 74% of the lamellae volume of the model lies in the inclined region. In Figure 2.34b, accumulated slip per unit crystalline volume, called the specific accumulated slip, is presented for the regions of the model and the whole model. In Figure 2.34b, $V_c(\Omega_r)$ is the volume of the crystalline lamellae of the region r . In Figure 2.34b, crystalline slip starts in the equatorial region and then in the inclined and polar regions, almost at the same time. In the inclined and polar regions the specific accumulated slip is almost the same up to 0.04 engineering strain. Then the rate of increase of the specific accumulated slip in the polar region passes both the inclined and equatorial regions and continues to increase. Up to the engineering strain 0.08, the specific accumulated slip in the polar region is less than that of the whole model, then increases at a linear rate and approaches to the specific slip activity of the equatorial region. Starting from 0.1 engineering strain, the difference between the specific accumulated slip of all regions decreases, since the slip rate decreases in the equatorial region and increases in the other regions. Specific accumulated slip in Figure 2.34b could be considered as a measure that indicates which region plastically deforms the most. Plastically, equatorial region deforms the most, while the inclined region deforms the least. Lower level of plastic activity in inclined lamellae is related with the stack rotation phenomena, enabled with the deformation of the amorphous region. Polar regions passes the

model average at 0.08 engineering strain and almost catches the equatorial region at the end of the analysis, under the low stresses presented in Figure 2.32. Considering $\{110\}\langle 1\bar{1}0\rangle$ is responsible from the 88% of the plastic deformation in polar regions in Table 2.6, this slip system should be behind the soft plastic response of the polar regions.

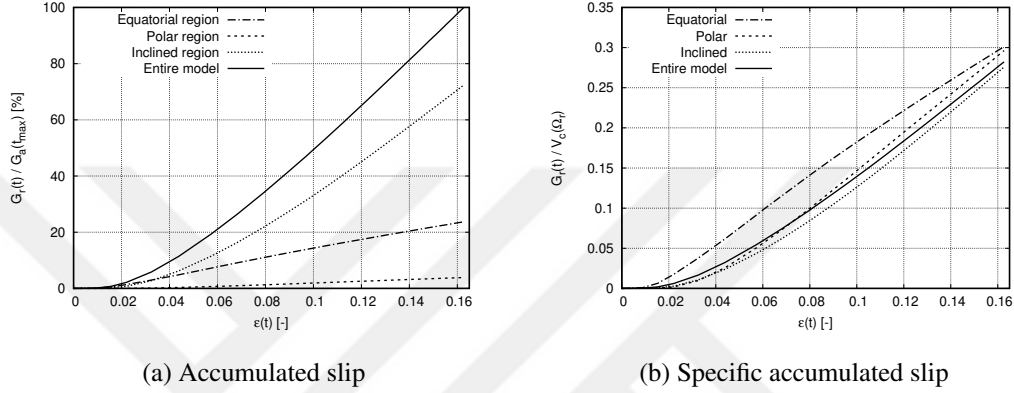


Figure 2.34: Variation of the total accumulated slip in the regions of the model with the macroscopic strain. In (a) vertical axis is normalized by the value of the total accumulated slip of all slip systems in the model at the 0.16 engineering strain, while in (b) vertical axis is normalized by the total volume of the lamellae in each region.

Slip system activities expressed in this section for the tensile loading are generally in agreement with the review of Lin and Argon [64] on the slip system activities, which is mainly based on the works of Allan and Bevis [5, 6] and Hay and Keller [44]. It should be noted that, unlike this study, these studies are conducted on 2D, i.e., disk like, spherulites deliberately formed within thin film samples to simplify the observations.

Hay and Keller [44] states activity of $(100)[001]$ in equatorial regions could explain the rotation of the chain axis towards the loading direction. Allan and Bevis [6] indicates that $\{110\}[001]$ and $\{110\}\langle 1\bar{1}0\rangle$ activity is a possible mechanism that could explain their observations on bright-field electron micrographs. In our model, the slip systems that have considerable activity in equatorial regions are $(100)[001]$ and $\{110\}\langle 1\bar{1}0\rangle$. $(100)[001]$ facilitates the deformation in the b plane of the lamellae, resulting in rotation of the chain axis towards the loading direction and thinning down

of the lamellae in the equatorial regions of the model. $\{110\}\langle\bar{1}\bar{1}0\rangle$, on the other hand, facilitates axial deformation along the b lattice axis. It enables the lamellae to shorten in equatorial region, which in turn facilitates interlamellar separation deformation mode of the almost incompressible amorphous phase in the model.

Allan and Bevis [6] considers that the chain slip and slip on the $\{110\}$ planes dominate the slip activity in the inclined regions. In other words, they do not expect high activity of the $(100)[010]$ and its orthogonal $(010)[100]$ slip systems in inclined region. In our model, however, although all slip systems are active in the inclined region, $(100)[010]$ is the dominant slip system. The high shear strains, along the b lattice axis, in the direction 45° away from the tensile axis is accommodated by the $(100)[010]$ slip in our model.

Allan and Bevis [6] specify $\{110\}\langle\bar{1}\bar{1}0\rangle$ as one of the major deformation mechanisms of the polar region. Indeed we find that $\{110\}\langle\bar{1}\bar{1}0\rangle$ is the most active slip system, responsible from the 88% of the crystalline slip activity in this region. We also find that $(100)[001]$ system takes up 9% of the slip activity in the polar regions. $(100)[001]$ activity is due to the lateral compressive stresses [80], originating from the Poisson's effect in polar region.

2.4.3.2 Compressive Loading of a Single Spherulite

In Figure 2.35, deformation of a selected set of lamellae under simple compression along x axis is presented, similar to Figure 2.29 of tensile loading. Contraction of the lamellae 3 and 7 that lie in the loading direction, extension of the lamellae 1, 5, 9, 12 that lie in the transverse direction, and rotation of the other lamellae around the nucleus towards the transverse direction are the noticeable features of the lamellae deformation in Figure 2.35. As in the simple tension case, the lamellae of the model can accommodate significant axial deformation. Rotation of the lamellae (2, 4, 6, 8, 10, 11, 13, 14) away from the loading direction is in line with the studies in literature [12].

In Figure 2.36, polar rr , $\theta\theta$, $r\theta$ components of the strain field on two planes in the model are presented, similar to the Figure 2.30 of the simple tension case. Inter-

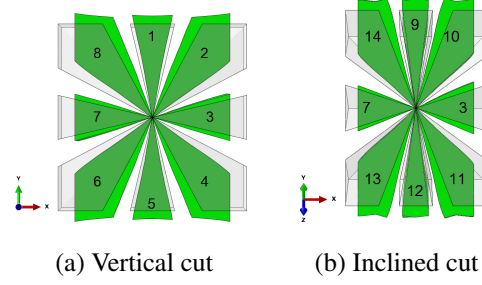


Figure 2.35: Undeformed (translucent) and deformed (solid) views of the selected lamellae of the model seen on the cuts passing through the nucleus. Viewing axes are aligned with the normal vector of the cross sections. Deformations are in true scale. In (b) cut plane normal is (0,1,1).

lamellar separation is observed near lamellae that grow in the loading direction in Figure 2.36(a), while in general, the strains in the amorphous phase is compressive. Since the highest strain components are in Figure 2.36(b), the interlamellar shear is dominant deformation mode of the amorphous phase.

In the following, slip system activities at an engineering strain of 0.18 for the spherulite with 50% crystallinity ratio are reported. In Table 2.7, relative activities of each slip system throughout the model are presented. In comparison to Table 2.6 of simple tension, relative activities of slip systems of the model do not change significantly in compression. As in the tensile loading case, (100)[001], (100)[010] and $\{110\}\langle\bar{1}\bar{1}0\rangle$ are the three most active slip systems in compressive loading, ordered from the most active to the least.

Table 2.7: Relative activities (%) of slip systems for simple compression, between 0 and 0.18 engineering strain

$(100)[001]$	$(010)[001]$	$(110)[001]$	$(\bar{1}\bar{1}0)[001]$	$(100)[010]$	$(010)[100]$	$(110)[\bar{1}\bar{1}0]$	$(\bar{1}\bar{1}0)[110]$
32.7	6.7	4.1	4.5	30.3	0.7	11.2	9.8

In Figure 2.37, variation of the accumulated slip in the model with the macroscopic strain is presented for each slip system, separately. The (100)[001] system is the

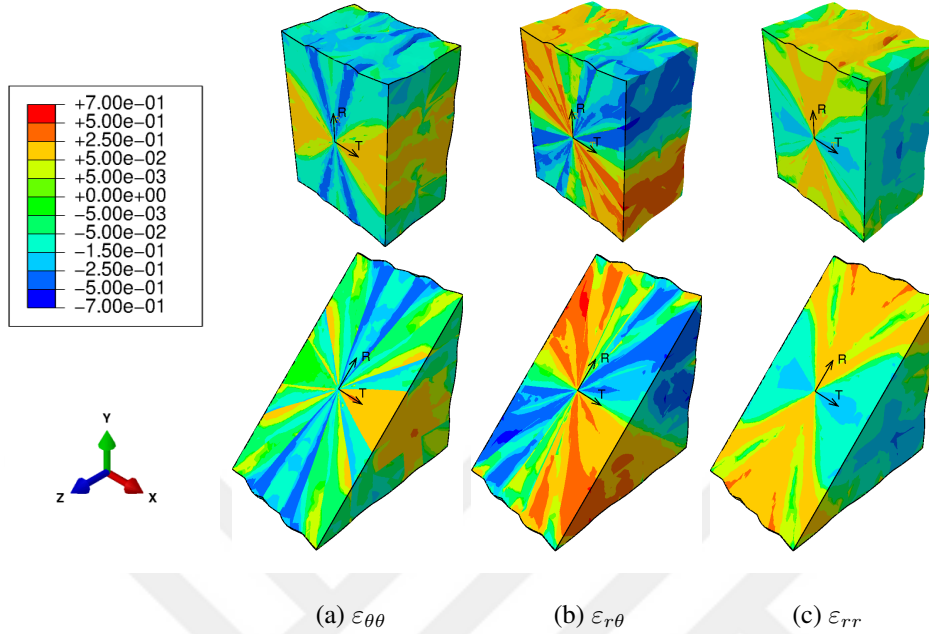


Figure 2.36: Distribution of the polar strain components for the simple compression analysis at 0.18 engineering strain. Strain components are in cylindrical coordinate systems, where origins are at the nucleus and z directions are along the normals of the cut planes. Normal vector of the inclined cuts are (0,1,1) in the Cartesian coordinate system shown.

first slip system that is activated. Rate of activity of the (100)[010] increases with strain. The $\{110\}\langle 1\bar{1}0\rangle$ activity starts approximately at the same macroscopic strain level with the activity of the (100)[010] system, and its rate also increases with increasing strain. Although in Figure 2.33 of the simple tension analysis, accumulated slip in $\{110\}\langle 1\bar{1}0\rangle$ systems were very similar, in Figure 2.37 accumulated slip in the (110)[$1\bar{1}0$] system is slightly more than the ($1\bar{1}0$)[110] system. Moreover, the rate of the accumulated slip in the $\{110\}\langle 1\bar{1}0\rangle$ systems is not constant as in simple tension, but increases in simple compression. The activity of the remaining slip systems, (010)[001], $\{110\}$ [001] and (010)[100] start later, at 0.04 macroscopic strain. Order of the slip systems from the most active to the least does not change throughout the analysis.

Figures 2.38 and 2.39 present contour plots of the chain and the transverse slip system activities, respectively. The compressive loading is parallel to the x-axis. The spatial

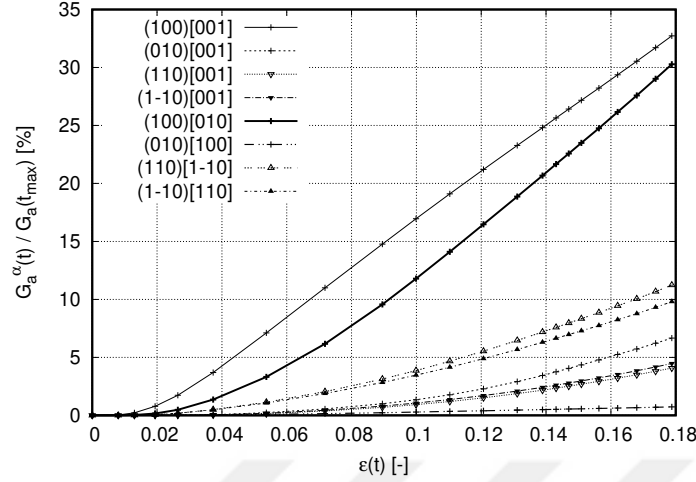


Figure 2.37: Variation of the accumulated slip of each slip system in the model with the macroscopic strain. Vertical axis is normalized by the value of the total accumulated slip in the model at the 0.18 engineering strain.

distribution of the slip system activity in the case of compression is very similar to that of the simple tension.

According to Figures 2.38b, 2.38c and 2.38d, (010)[001] and {110}[001] are active on the same lamellae set. These two slip systems have the same characteristic slip resistance and are both predominantly governed by the magnitude of τ_{bc} stress component according to Table 2.5. According to Table 2.6, the combined activity of {110}[001] systems is slightly higher than the activity of (010)[001]. While the Schmid stress of (010)[001] depends only on the τ_{bc} , Schmid stress of the {110}[001] depends on τ_{ac} as well. Therefore, the observed activity difference between these systems could be associated with contribution of the τ_{ac} stress component to the Schmid stress of the {110}[001].

Similar to the tensile loading, among the three most active slip systems, (100)[001] and {110}⟨1 $\bar{1}$ 0⟩ are active in all lamellae of the model, while (100)[010] is active only in the lamellae inclined with respect to the loading axis. In Figures 2.38 and 2.39, it is seen that (100)[001] and {110}⟨1 $\bar{1}$ 0⟩ are the only slip systems that are active in all lamellae of the model. While all slip systems show some activity in the inclined lamellae, (100)[001] and {110}⟨1 $\bar{1}$ 0⟩ are the only ones that are considerably active in the lamellae that are parallel or perpendicular to the compression axis.

Considering the lamellae growth is along the b lattice axis and the unit cell of PE is orthorhombic, geometrically it is evident that only the activity in $\{110\}\langle 1\bar{1}0\rangle$ systems could host the plastic deformation that extends or shrinks the lamellae. Therefore, the dominant activity of $\{110\}\langle 1\bar{1}0\rangle$ in the lamellae lying along the compression axis is expected. The activity observed in $(100)[001]$ slip system in these lamellae facilitates the plastic deformation that accommodates the lateral extension due to the Poisson's effect on the length scale of the spherulite.

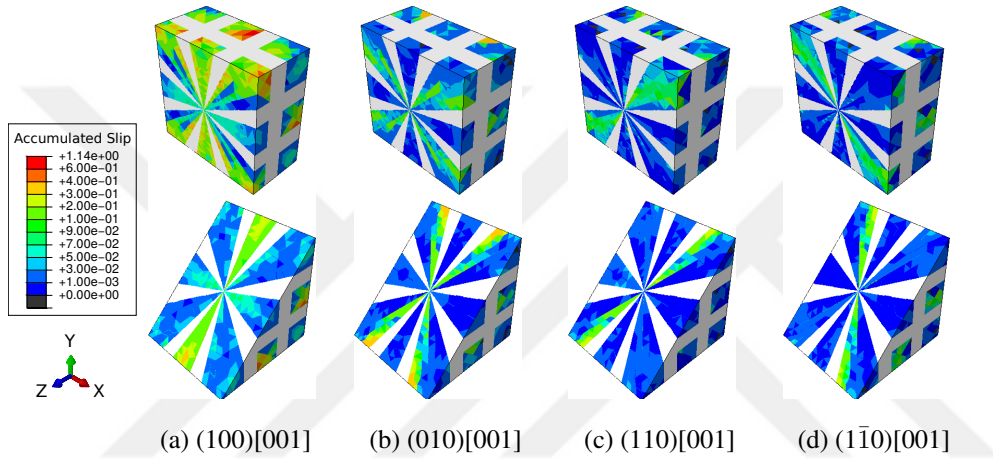


Figure 2.38: Distribution of the chain slip amounts for simple compression analysis. Loading is in the x direction. First row presents a vertical cut through the nucleus. Second row is an inclined cut, with a normal vector of $(0,1,1)$, through the nucleus.

Bartczak et al. [12] studied texture evolution of HDPE during uniaxial compression. Their samples have more than 70% crystallinity ratio and possess spherulites with 5 μm diameter. They report the overall crystallographic orientations of the samples as determined from wide angle X-ray scattering (WAXS). They observe that, (100) normals move towards the loading direction while (010) and (001) normals move away from it. They conclude that, activities of $(100)[001]$ and $(100)[010]$ systems, which have lower slip resistances than the other slip systems, explains this observation. They do not observe a strong orientation change of (110) normals neither towards nor away from the loading direction. Based on this, they indicate that no slip activity on (110) planes is detected. Finally they state a probable but limited activity of $(010)[001]$ slip system at the initial stages of deformation, based on the orientation change in (010) normals. The two most active slip systems expected by Bartczak et al. [12], namely

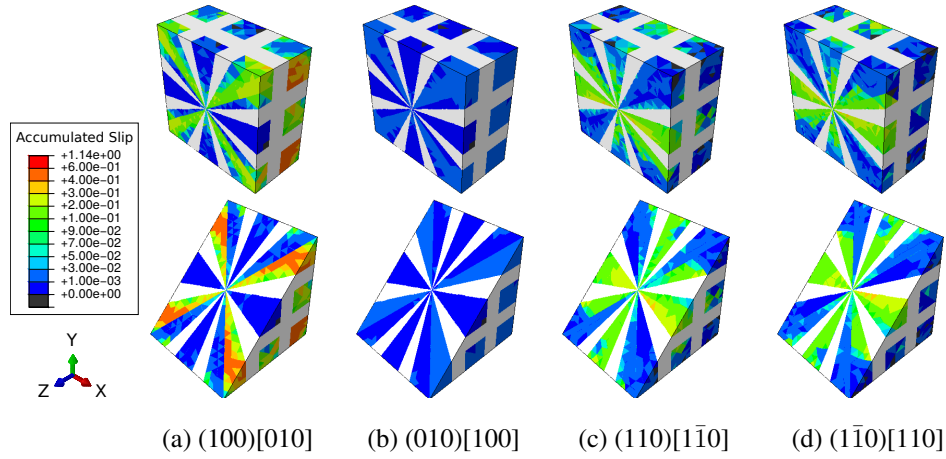


Figure 2.39: Distribution of the transverse slip amounts for simple compression analysis. Loading is in the x direction. First row presents a vertical cut through the nucleus. Second row is an inclined cut, with a normal vector of (0,1,1), through the nucleus.

(100)[001] and (100)[010], are responsible from the 63% of all crystallographic slip activity in our model. Considering the activity of (010)[001] as well, almost 70% of the plastic slip in the model is due to the slip systems detected by the cited study. The third most active slip system of the model, $\{110\} \langle 1\bar{1}0 \rangle$, which is responsible from the 21% of all crystalline slip is not detected in the study of Bartczak et al. [12]. To sum up, there is good agreement between our model and the experimental study [12] on the activity of all slip systems, except the ones on the $\{110\}$ planes, especially the $\{110\} \langle 1\bar{1}0 \rangle$ system.

2.4.3.3 Effect of the Degree of Crystallinity

In this section, the effect of crystallinity on the mechanical properties of the spherulite microstructure is shown. As mentioned in Section 2.4, crystalline phase regions of the model are composed of pyramids with square bases. The base dimensions of these pyramids are the same, regardless of the lamella group (FC, C, ME) they belong. By changing the base dimensions, models with volumetric crystallinity ratios of 50%, 62.5%, 75% and 87.5% are obtained as shown in Figure 2.40.

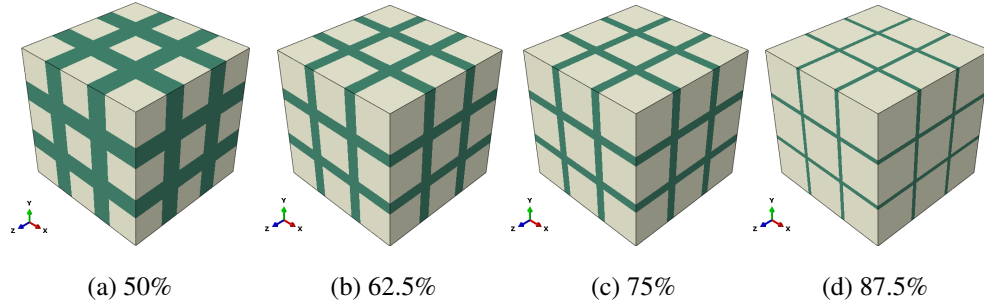


Figure 2.40: Views of 3d spherulite FEM models for different crystallinity ratios.

Variation of engineering stress-strain curves with crystallinity ratios under simple tension and compression loading conditions are given in Figure 2.41. For both loading cases, initial stiffness and yield stress increase with crystallinity ratio, while the post-yield hardening slope remains constant. These features are also reported in the numerical studies of Van Dommelen [109] and Ayoub et al. [9] for HDPE.

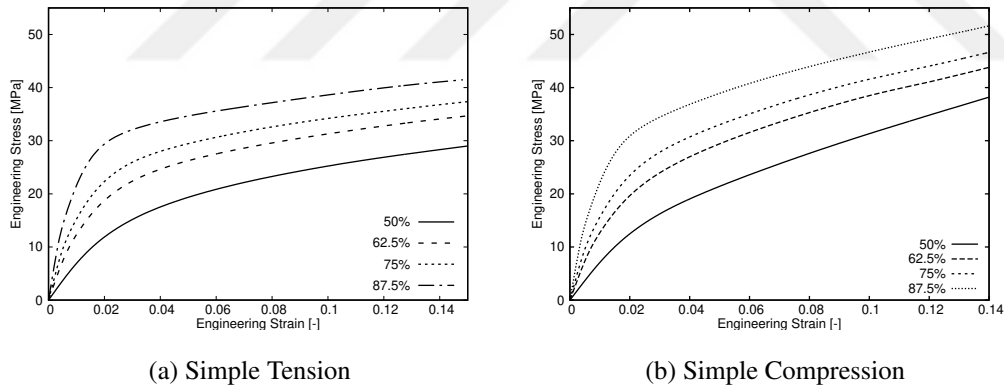


Figure 2.41: Variation of engineering stress strain curves of single spherulite FEM analyses with volumetric degree of crystallinity.

In Figure 2.42, variation of the yield stress and the Young's modulus with the degree of crystallinity of the present model, our previous disk-like spherulite model [79] and the experiments of Crist et al. [25] are presented. The data points in Figure 2.42 are obtained from the simple tension engineering stress-strain curves following Crist et al. [25]. Young's modulus is the slope of the initial tangent to the curve, while the yield stress is the stress corresponding to the point of intersection of the initial tangent and the tangent to the curve at approximately 15% strain. Data points of quenched

samples of Crist et al. [25] are excluded from Figure 2.42 considering there would not be enough time for the development of the spherulites. It is seen that the predictions of this study are in general agreement with the results of Crist et al. [25]. Especially the slopes in Figure 2.42 are in good agreement with [25]. In Figure 2.42 both the yield stress and the Young's modulus of the model is higher than that of the disk-like spherulite model. It should be noted that this study considers a single spherulite. On the other hand, the study of Crist et al. [25] is on macroscopic samples that are composed of multiple spherulites. As discussed in Hay and Keller [44] and also recently presented in Teixeira-Pinto et al. [101]; the macroscopic strain, in general, is accommodated by not a single spherulite but a collection of spherulites. Therefore, a multi-spherulitic model may be more representative for a comparison with the results of Crist et al. [25].

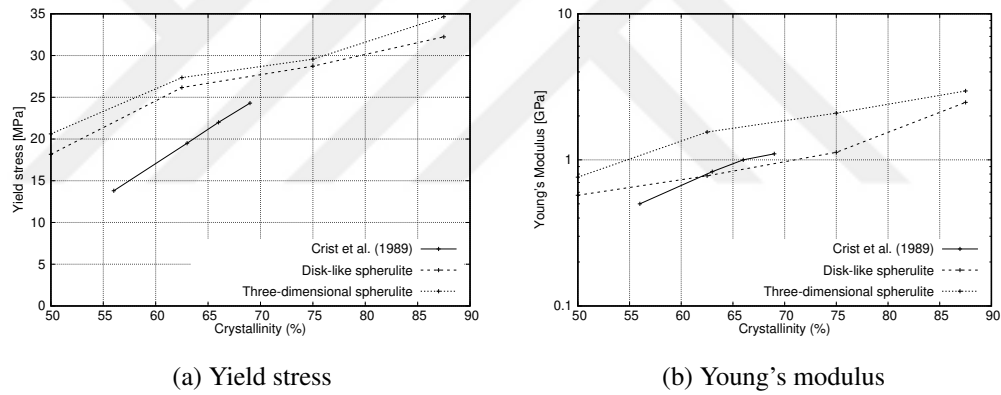


Figure 2.42: Comparison of findings of this study with experimental study of [25] on variation of macroscopic yield stress and Young's modulus with crystallinity ratio.

2.4.4 Discussion

A three-dimensional finite element model of a single spherulite, allowing consistent deformation of crystalline and amorphous phases within the length scale of the spherulite is studied. Simple tension and simple compression loading cases are analyzed, and the stress and strain distribution characteristics of the model are found to possess the kinematically necessary features of the spherulite deformation. Slip system activities throughout the spherulite and in the case of tensile loading within

the polar, the equatorial and the inclined regions are reported. These activities are generally in good agreement with the literature. In simple tension and simple compression, the slip in (100)[001], (100)[010] and $\{110\}\langle 1\bar{1}0\rangle$ systems amounts to more than 80% of the total slip system activity. These are the three most easily activated slip systems of the PE according to Table 2.4. The next most active slip system is the (010)[001], that accounts for 7% of the total slip activity for both loading conditions. The model enables consideration of arbitrary crystallinity ratios. Influence of crystallinity ratio on the macroscopic stress-strain response, the yield stress and the initial elastic modulus is investigated and a good agreement with the literature is found.

For both the disk-like and the three-dimensional spherulite models of this study, high activity of $\{110\}\langle 1\bar{1}0\rangle$ system in compression is not in good agreement with Bartczak et al. [12], and the polar region showing the least resistance to deformation in tensile loading is not in good agreement with Allan and Bevis [6] and Hay and Keller [44], and the review papers [64, 80]. These are the main differences of the results of this study with the experimental studies. The low deformation resistance of the polar regions in tension is associated with $\{110\}\langle 1\bar{1}0\rangle$ system as well, and will be explained here.

$\{110\}\langle 1\bar{1}0\rangle$ accounts for the almost 90% of plastic deformation in the polar regions of the model. One important constraint of the crystal plasticity framework is that, plastic flow directions are limited. Other slip systems have very limited activity in polar regions since $\{110\}\langle 1\bar{1}0\rangle$ are the only systems that could extend the lamellae along b lattice axis and can be activated with stress component τ_{bb} according to Appendix A and Table 2.5, respectively. In Table 2.5, coefficients of the normal components of the stress are the same for (110)[$1\bar{1}0$] and ($1\bar{1}0$)[110] systems. Therefore, when the principal stress directions correspond to the lattice axes, slip on both systems have the same sense and magnitude according to the employed flow rule (2.6). In Figure 2.39, the activities of these systems are very similar. The plastic velocity gradient composed of the simultaneous and equal rate of slip on (110)[$1\bar{1}0$] and ($1\bar{1}0$)[110] systems is

$$\mathbf{L}^p = \dot{\gamma} 0.92 (\mathbf{a} \otimes \mathbf{a} - \mathbf{b} \otimes \mathbf{b}) , \quad (2.11)$$

where $\mathbf{a}$ and $\mathbf{b}$ are the unit vectors along a and b lattice axes in the intermediate configuration, and $\dot{\gamma}$ is the slip rate. Notice that in this case, $\mathbf{L}^p$ is symmetric, and

therefore does not change the lattice orientation with the plastic spin. Recall that, in the polar region of the spherulites, the lattice axis b is always almost parallel to the principal extension direction, since the lamellar twist is along the lattice axis b . Even for the relatively complex orientation state of our model described in Section 2.4.1, since the b lattice axes are along the macroscopic loading direction, and $\mathbf{b}$ is an eigenvector of (2.11), the activity of $\{110\} \langle 1\bar{1}0 \rangle$ enables the unhindered plastic flow in the polar region lamellae along the macroscopic loading direction. Moreover, since the plastic spin is zero, the alignment of the flow direction with the extension direction is preserved with the flow. Therefore, especially compared to the equatorial region, the unhindered plastic flow is the reason of the lower stresses in the polar lamellae than that in the other regions of the model, see Figure 2.32, once $\{110\} \langle 1\bar{1}0 \rangle$ is activated.

As also pointed out in the review articles [64, 13] it is well documented in the literature that $\{110\}$ twinning is observed in PE crystals, and it is in strong competition with $\{110\} \langle 1\bar{1}0 \rangle$. Bartczak et al. [10] could not measure slip resistance of $\{110\} \langle 1\bar{1}0 \rangle$ since $\{110\}$ twinning is activated first in their single-crystal-textured samples. Wu et al. [116] found that the twinning and martensitic transformation are easier to activate than the slip in the $a-b$ plane. Although the contribution of twinning to the plastic strain is minor, twinning results in drastic changes in orientation, and could rotate the crystal to an orientation that is more favorable for crystallographic slip, as stated by Bartczak and Galeski [13]. Consequently, a considerable activity of $\{110\} \langle 1\bar{1}0 \rangle$ slip system and the soft polar regions in our numerical results could be associated with the simplification of considering the crystalline slip as the only plastic deformation mechanism of the PE crystal. On the other hand, it may also be harder to detect slip systems that operate together with other systems in such a way that the combined activity tends not to change the lattice orientation. Since the slip activities in bulk specimens are generally detected with WAXS measurements through the changes in crystal orientation [12].

2.5 Conclusion

The disk-like and three-dimensional finite element models of a single spherulite, allowing consistent deformation of the crystalline and amorphous phases within the

length scale of the spherulite are presented. Both models could consider arbitrary crystallinity ratios. While the number of crystalline regions is constant in the three-dimensional model, the response of the disk-like model converges with the increasing number of regions. The stress and strain distribution characteristics of the models are found to possess the kinematically necessary features of the spherulite deformation. Slip system activities throughout the models and in the case of tensile loading within the polar, the equatorial and the inclined regions are reported.

In the disk-like model, $\{110\} \langle 1\bar{1}0 \rangle$ is considerably active in both tensile and compressive deformation modes. Therefore, the high activity of $\{110\} \langle 1\bar{1}0 \rangle$ was attributed to the disk-like geometry. Recall that although Allan and Bevis [6] considers $\{110\} \langle 1\bar{1}0 \rangle$ as the major deformation mechanism of the polar regions, in their tensile experiments on disk-like spherulites, Bartczak et al. [12] under simple compression were not able to detect the activity of $\{110\} \langle 1\bar{1}0 \rangle$ on bulk specimens composed of three-dimensional spherulites. In order to verify this attribution, a three-dimensional model of a single spherulite is constructed, and the slip system activities are investigated in simple tension and compression. The high activity of $\{110\} \langle 1\bar{1}0 \rangle$ persists in the lamellae under the kinematical constraints of the three-dimensional model. Therefore, high activity of $\{110\} \langle 1\bar{1}0 \rangle$ is not an artifact of the geometry of our disk-like model.

Considerable activity of $\{110\} \langle 1\bar{1}0 \rangle$ slip system and the soft polar regions in our numerical results could be associated with the assumption of considering the crystalline slip as the only plastic deformation mechanism of the PE crystal. This assumption is frequently employed in the literature since the amount of plastic strain accommodated by the neglected twinning and martensitic transformations are not substantial, and we are not aware of any study that considers them for PE within the crystal plasticity framework. On the other hand, although the contribution of the twinning and martensitic transformation to the plastic strain is not much, as stated by Bartczak and Galeski [13], they result in drastic changes in the orientation and could rotate the crystal to an orientation that is more favorable for crystallographic slip.

CHAPTER 3

LAMINATE-BASED HOMOGENIZATION APPROACHES FOR NON-PARALLEL INTERFACES

3.1 Introduction

Similar to the metals, the nature of the plastic deformation of the semicrystalline polymers is mainly crystallographic and the deformation generally does not destroy the crystalline order [13]. After it was understood that the semicrystalline polymers possess the deformation mechanisms similar to that of the metals, approaches for obtaining homogenization based constitutive models of metals [90] have been also employed for modeling the semicrystalline polymers. However, application of those approaches to the SCPs were not a straightforward task. The PE crystal cannot accommodate arbitrary plastic deformations due to the existence of the covalently bonded strong polymer chains which remains intact during crystallographic slip. In order to accommodate arbitrary strains with crystallographic slip a crystal should have five independent slip systems considering the number of arbitrary components of a plastic strain tensor ϵ^p is five. Notice that since the plastic deformation preserves volume $\text{tr}(\epsilon^p) = 0$, one of the components is not arbitrary. As a result of this kinematic deficiency to accommodate plastic deformation, the polymer single crystals appear brittle. On the other hand, SCPs in bulk are quite ductile since the shear in the amorphous phase between the lamellae provides the necessary kinematical freedom [13].

In the literature, homogenization techniques are used to obtain the macroscopic response of polycrystalline materials. Among homogenization techniques, the Taylor approach, which assumes a uniform microscopic deformation field, provides an upper bound, and yields the stiffest response. Nevertheless, the Taylor approach leads

to good results for metals especially for cubic crystals, which have many equivalent slip systems with similar slip resistances. These crystals can support arbitrary plastic deformations. Since the same strain is enforced to all crystals in the Taylor approach, high stresses may be obtained for highly anisotropic crystals. Moreover, when rigid-plastic constitutive models are employed, the PE crystal becomes kinematically deficient, and the stresses may become unbounded [83]. This issue was addressed by Parks and Ahzi [83]. They proposed a constrained single crystal model in which an extension in the chain direction is prohibited by kinematic constraints. In the homogenization part, the macroscopic rate of deformation is assigned to each crystal by filtering out the components that violate the constraint condition. Therefore, they enabled the application of the Taylor approach to SCPs that do not support arbitrary plastic deformations. As an early study, they completely ignored the amorphous phase and assumed 100% crystallinity. Moreover, the elastic deformation of the crystals was also ignored by utilizing a rigid-viscoplastic formulation. In spite of these limitations, a reasonable agreement was obtained with the experiments on the texture evolution of high density polyethylene (HDPE). Lee et al. [60] reformulated the approach of Parks and Ahzi [83] and removed the requirement of explicit identification of the kinematic constraints. Similar to Parks and Ahzi [83], Yang and Chen [118] considered the crystalline phase as rigid viscoplastic. They employed a continuous orientation distribution for the chain axes in a unit sphere. The amorphous phase was considered in the model with its role on heterogeneous spin of crystals. They obtained good agreement with the stress-strain curves obtained from the tension and simple shear tests of HDPE.

Lee et al. [62] considered the basic building block of SCPs as a laminate composed of amorphous and crystalline phases sharing a common planar interface. They employed the lamination theory which provides the equilibrium of tractions and kinematical compatibility conditions on the plane that separates the phases. Their model is also rigid-plastic since rigid-viscoplastic constitutive models are employed for the phases. For the crystalline phase, the constrained single crystal model of Parks and Ahzi [83] was employed. In the amorphous phase a power law type flow rule was employed and orientational hardening was introduced through a back-stress tensor calculated according to the eight chain model of Arruda and Boyce [8]. In addition

to the Taylor approach, they employed Sachs approach and also proposed two Taylor/Sachs hybrid approaches. They obtained a good agreement with the experimental studies on texture evolution and stress-strain behavior of HDPE even though elasticity was fully ignored in their model. Van Dommelen et al. [109] introduced elasticity to the constitutive models of the both phases of the model of Lee et al. [62], and therefore enabled employment of a general crystal plasticity formulation instead of the constrained single crystal model of Parks and Ahzi [83]. In addition to the texture evolution and the stress-strain behavior, they obtained reasonably good predictions for the effect of the crystallinity ratio on the initial modulus and the yield strength. Sedighiamiri et al. [95, 94] added quantitative abilities to the model of Van Dommelen et al. [109] by adjusting the constitutive models of the phases. Moreover, three phase approaches are employed to determine the Young's modulus of PET [37] and HDPE [96]. In these studies, a rigid amorphous interface that lies between the crystalline phase and the amorphous phase was considered, and the lamination direction of the phases is the same.

To sum up, the lamination theory is frequently employed in order to introduce the amorphous phase to the SCP models. This approach provided the necessary kinematical freedom to the PE crystals especially with the experimentally observed interlamellar shear deformation mode of the amorphous phase. Therefore, laminate is considered as the basic building block of SCPs models in the literature that also include [2, 78] which employ the self-consistent scheme. However, it is well known that the response of a single laminate reduces to that of the Taylor and Sachs approaches in certain loading directions. Therefore, even though the Taylor approach could be employed to the laminates, the predictions of the Taylor approach were higher than the experiments both for the initial Young's modulus and the yield stress, and self-consistent schemes provided better predictions [62, 109].

Approaches similar to the lamination theory are also employed in constitutive models of metals, particularly in the so called grain cluster methods that take the interaction of adjacent grains into account and also reviewed in Roters et al. [90]. Stress and strain fields are typically uniform in each crystal within a cluster, and clusters are homogenized with the Taylor approach. According to Evers et al. [33], that is considered as a grain cluster model which takes the hardening induced by the geometrically

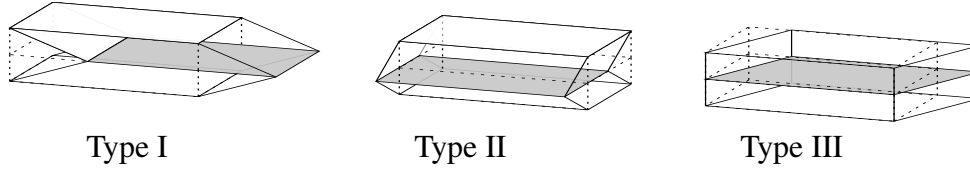


Figure 3.1: Relaxation modes of the LAMEL models, adapted from Van Houtte et al. [112]

necessary dislocations into account, the first study that employ Taylor approach with a bi-crystal is Ahzi et al. [4] and the context of which is the modeling of semicrystalline polymers. Grain cluster methods lead to the relaxation of the Taylor approach and in some cases provide better results. For instance, in the LAMEL model of Van Houtte et al. [111, 110], a cluster is composed of two crystals sharing a common interface. The Taylor approach is applied to the clusters, instead of the individual crystals. Therefore, the average strain in each cluster is the same with the others, strain may vary in the individual crystals without violating the kinematical compatibility on the interface as presented in Figure 3.1. The LAMEL model is employed for the rolling deformation mode, since the lamination direction of all clusters is the same and aligned with the normal of the rolling plane. Qualitative predictions of the rolling textures, especially of steel, are improved significantly with this approach. Van Houtte et al. [112] proposed the advanced LAMEL method, that extends the LAMEL method to more general deformation modes. There is also interest in taking the interaction between more than two crystals into account. For instance, the grain interaction (GIA) model of Crumbach et al. [27] is another grain cluster method, clusters of which are composed of eight crystals. Two dimensional view of four crystals of a cluster is presented in Figure 3.2. In the GIA model, shear in the A1 and A3 crystals are in opposite sense and cancel each other, while the kinematical misfit between A1 and A2 is motivated and penalized by the geometrically necessary dislocations.

Grain cluster methods take the interactions of the adjacent grains into account, importance of which is also qualitatively presented for the elastic properties by Bunge [20]. However, as pointed out by Van Houtte et al. [112] the following are the unrealistic features of these approaches. They may introduce misfits at least between the crystals that belong to different clusters as in the LAMEL method, or even between the

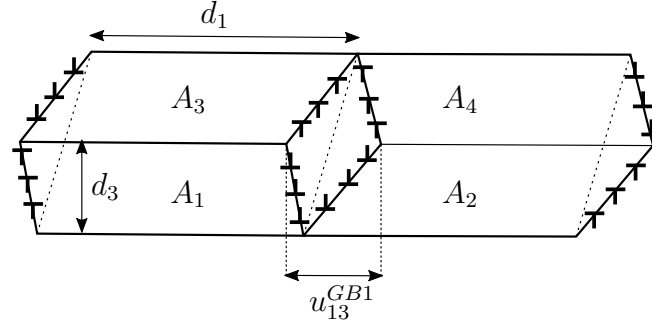


Figure 3.2: Two dimensional view of the cluster of the GIA model, adapted from Engler et al. [31]

crystals within the same cluster as in the GIA method. How many crystals a cluster is composed of is not physically justified. Interfaces within the cluster have special emphasis compared to the outer boundaries of a cluster which can be considered as interfaces that separate a cluster from the others. Finally, stress and strain fields are assumed to be uniform in a crystal of a cluster. While the in-cluster misfits are motivated and penalized with the geometrically necessary dislocations in the GIA model, Yadegari et al. [117], which also employ the uniform strain field assumption within a crystal in their generalized grain cluster method, simply consider them as the result of the kinematic assumptions of the model.

In this chapter we propose a set of methods that can be employed to construct clusters composed of arbitrary number of regions. As in the lamination theory [74], a planar interface lies between two adjacent regions, and deformation field of each region is uniform. It will be reviewed that in rank-1 laminates there exists a single interface where both the traction equilibrium and deformation continuity conditions are satisfied. However, in the proposed methods, multiple interfaces exist at the same length scale, and these conditions are not satisfied simultaneously on the interfaces. Instead, one of them is relaxed with a condition that minimizes the effective Helmholtz free energy. In other words, either the force equilibrium or the deformation continuity is present at all interfaces of the model. It can be shown that as the number of regions increases and the consecutive interfaces approach each other, the relaxed condition is better fulfilled. In classical lamination theory, the Hill-Mandel condition is trivially satisfied, such that the total mechanical work is conserved in the homogenization. In the proposed methods, the Hill-Mandel condition is trivially satisfied as well. As a

model problem disk-like spherulite morphology is employed, and the obtained results are compared with the FE model studied in Section 2.3 with a similar crystalline texture and the identical constitutive models and material parameters. In order to express applicability of the methods to different geometries, a model for the twisted-ribbon-shaped lamellae of spherulites is also presented.

3.1.1 Rank-1 Laminate

In the rank-1 laminate, schematically shown in Figure 3.3, there is perfect bonding and static equilibrium between the phases, separated by a planar interface. The unit normal vector of the interface in the reference configuration is denoted by $\mathbf{N}$, which is also the lamination direction. The mechanical properties of the laminate could change only along this direction. The deformation gradient $\mathbf{F}^I$ and the first Piola-Kirchhoff stress tensor $\mathbf{P}^I$ are taken as uniform in each phase I . Therefore,

$$(\mathbf{P}^2 - \mathbf{P}^1) \mathbf{N} = \mathbf{0} \quad (3.1)$$

is the equation of the static equilibrium on the interface. Any pair of linearly independent unit vectors $\mathbf{S}$ and $\mathbf{T}$ that satisfy $\frac{\mathbf{S} \times \mathbf{T}}{|\mathbf{S} \times \mathbf{T}|} = \mathbf{N}$ spans the interface plane in the reference configuration. Deformation gradients of the phases $\mathbf{F}^I$ should map these vectors to the same vectors in the reference configuration, in order to satisfy the kinematic compatibility

$$(\mathbf{F}^2 - \mathbf{F}^1) \mathbf{T} = \mathbf{0} \quad \text{and} \quad (\mathbf{F}^2 - \mathbf{F}^1) \mathbf{S} = \mathbf{0}. \quad (3.2)$$

Since both $\mathbf{S}$ and $\mathbf{T}$ are perpendicular to $\mathbf{N}$, (3.2) is satisfied when

$$\mathbf{F}^2 - \mathbf{F}^1 = \mathbf{w} \otimes \mathbf{N} \quad (3.3)$$

where $\mathbf{w}$ is an arbitrary vector in the current configuration. The displacement field of the laminate becomes continuous when (3.3), which is also known as the Hadamard's jump condition [81], is satisfied. Overall deformation gradient $\bar{\mathbf{F}}$ is computed by volume averaging

$$\sum_{i=1}^2 \nu^i \mathbf{F}^i = \bar{\mathbf{F}}, \quad (3.4)$$

where ν^i are the volume fractions of the phases, and $\sum_{i=1}^2 \nu^i = 1$. The following forms of the deformation gradients of the phases trivially satisfy both (3.3) and (3.4)

$$\mathbf{F}^1 = \bar{\mathbf{F}} - \frac{1}{\nu^1} \mathbf{m} \otimes \mathbf{N}, \quad \mathbf{F}^2 = \bar{\mathbf{F}} + \frac{1}{\nu^2} \mathbf{m} \otimes \mathbf{N}, \quad (3.5)$$

note that relationship between $\mathbf{m}$ and $\mathbf{w}$ is $\mathbf{m} = \frac{\nu^1 \nu^1}{\nu^1 + \nu^2} \mathbf{w}$. In the subsequent developments, we will consider that $\mathbf{w}$ depends on $\mathbf{m}$, and $\mathbf{m}$ is arbitrary. $\hat{\mathbf{F}}^i(\bar{\mathbf{F}}, \mathbf{m})$ given in (3.5), satisfy the kinematic requirements of the laminate in (3.2) and (3.4), for any value of $\bar{\mathbf{F}}$ or $\mathbf{m}$. Notice that, the unknowns of the laminate are the components of the deformation gradients $\mathbf{F}^1$ and $\mathbf{F}^2$ in (3.2) and (3.4), while the only unknowns in (3.5) are the components of the vector $\mathbf{m}$. Therefore, the number of unknowns decreases from 18 to 3, and the 15 unknowns of the laminate are eliminated in (3.5) to satisfy the 6 scalar equations in (3.2) and 9 scalar equations in (3.4).

According to (1.12), the average strain energy density function of the laminate is

$$\bar{\Psi}(\bar{\mathbf{F}}, \mathbf{I}^1, \mathbf{I}^2) = \min_{\mathbf{m}} \left\{ \sum_{i=1}^2 \nu^i \Psi^i(\hat{\mathbf{F}}^i(\bar{\mathbf{F}}, \mathbf{m}), \mathbf{I}^i) \right\}$$

where Ψ^i and $\mathbf{I}^i$ are the free energy functions and the internal state variables associated with the dissipative mechanisms of the phases, respectively. The necessary condition of the minimization leads to

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}} = (\mathbf{P}^2 - \mathbf{P}^1) \mathbf{N}$$

according to (3.5), and the average stress is

$$\bar{\mathbf{P}} = \frac{\partial \bar{\Psi}}{\partial \bar{\mathbf{F}}} = \sum_{i=1}^2 \nu^i \mathbf{P}^i,$$

as expressed in Section 1.3.1.

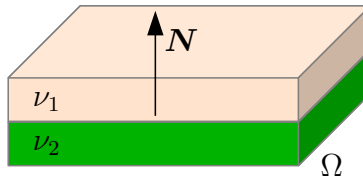


Figure 3.3: Schematic view of a rank-1 laminate

3.1.2 Sequential Laminate

A rank-1 laminate, that employs another rank-1 laminate in one of its phases as a constitutive model, is referred to as a rank-2 laminate. Similarly a rank-3 laminate employs a rank-2 laminate as a constitutive model, and so on. Laminates with ranks greater than 1 are called sequential laminates. In other words, sequential laminates are the laminates of laminates. In this section two-phase laminates schematically shown in Figure 3.3 are employed to obtain sequential laminates, where laminates are employed as constitutive models in the both phases of all laminates except in the innermost one, which is rank-1 by definition. Considering the volume averaging framework, employing a laminate as a constitutive model within another implies that the employed laminate is much smaller. When the modeled problem enables, sequential laminates can satisfy the principle of the separation of length scales. With their possibly infinite number of length scales that possess stress fields in equilibrium and compatible strain fields, sequential laminates are frequently employed in differential schemes of homogenization and can show the attainability of the homogenization bounds [30, 74].

In this part of the introduction, a preliminary modeling approximation of the disk-like spherulite FE model in Section 2.3 is presented with sequential laminates. We employ sequential laminates in a model that has non-parallel interfaces between the amorphous and crystalline regions, in order to solve the model with laminates. However, employment of sequential laminates violates the MMM principle of Hashin [42], since the interfaces of the disk-like spherulite model are actually at the same length scale. More realistic modeling approaches are presented in this chapter after the developed formulation in Section 3.2, and the results of this section are employed in the upcoming sections for comparison.

The schematic views of the model for different number of regions are presented in Figure 3.4. The even and odd numbered regions are amorphous and crystalline, respectively, and are also called slices. In the amorphous and crystalline regions of the model, the constitutive models and material parameters in Section 2.2 are employed. The lattice orientations in the crystalline regions are similar to the FE model of the disk-like spherulite given in Figure 2.7 for the 32 slices, except orientation of each

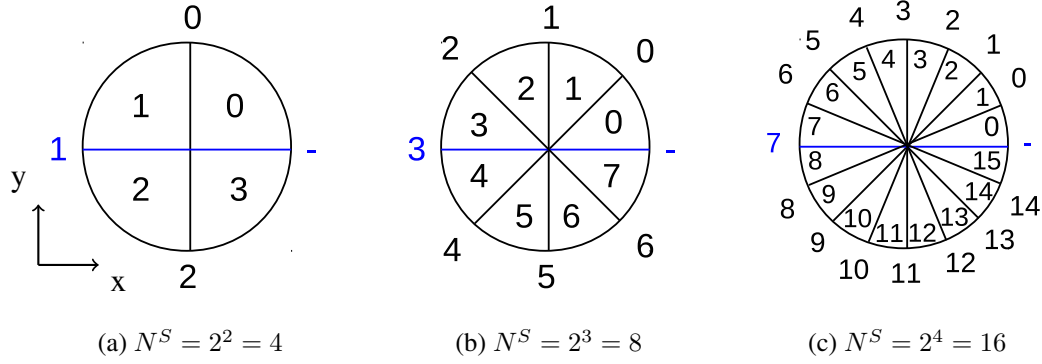


Figure 3.4: Schematic view of the slices and interfaces of the model for different number of slices

region is uniform and computed according to the coordinate of the centroid of the region.

3.1.2.1 Equations of the Sequential Laminate-based Preliminary Model

The equations of the sequential laminates could simply be described as the recursive application of the equations of the rank-1 laminates. It is known, however, the results and therefore the explicit equations of the sequential laminates depend on the order of lamination. For the models presented in Figure 3.4, selecting the interface of the highest-rank laminate defines the order of lamination. Interfaces 1, 3 and 7 are chosen for this purpose for the models with 4, 8 and 16 slices, respectively. Then, for example for the model with 4 slices, interface 1 belongs to a rank-2 laminate that employs two separate rank-1 laminates with interfaces 0 and 2, respectively. Since rank-1 laminates employ uniform stress and strain field assumptions, these fields are also uniform in the slices of the model. Then, the compatibility and equilibrium are satisfied between slices 0 and 1 at interface 0, and between slices 2 and 3 at interface 2. As for the interface 1, which is the interface of the rank-2 laminate, the compatibility and equilibrium are also satisfied but at the higher length scale, i.e. between the overall quantities of the two rank-1 laminates. It should be remarked that the interfaces of the highest-rank laminates in Figure 3.4 are not between two slices, which is discussed at the end of this section.

Table 3.1: Slices that appear in the equations of each interface

Interface no. (I)	Length scale (laminate rank)	Interface exists if	Counter-clockwise slices ($s(I)$)	Clockwise slices ($f(I)$)
0	1	$N^S \geq 4$	$\{0\}$	$\{1\}$
1	2		$\{0, 1\}$	$\{2, 3\}$
2	1		$\{2\}$	$\{3\}$
3	3	$N^S \geq 8$	$\{0, 1, 2, 3\}$	$\{4, 5, 6, 7\}$
4	1		$\{4\}$	$\{5\}$
5	2		$\{4, 5\}$	$\{6, 7\}$
6	1		$\{6\}$	$\{7\}$
7	4	$N^S \geq 16$	$\{0, 1, \dots, 7\}$	$\{8, 9, \dots, 15\}$
8	1		$\{8\}$	$\{9\}$
9	2		$\{8, 9\}$	$\{10, 11\}$
10	1		$\{10\}$	$\{11\}$
11	3		$\{8, 9, 10, 11\}$	$\{12, 13, 14, 15\}$
12	1		$\{12\}$	$\{13\}$
13	2		$\{12, 13\}$	$\{14, 15\}$
14	1		$\{14\}$	$\{15\}$

Below we will present the equations of the model explicitly in terms of the deformation gradient $\mathbf{F}^I$ and first Piola-Kirchhoff stress tensor $\mathbf{P}^I$ of slice I , where $I = 1, \dots, N^S$ and N^S is the number of slices. In Table 3.1, interface and slice numbers are consistent with Figure 3.4. Each row belongs to an interface and the length scale of an interface is in the second column. Value of this column also corresponds to the rank of the laminate the interface belongs to. The table is representative for models up to 16 slices. The rows of the table that correspond to models with different number of slices are indicated in the third column. Finally, the fourth and fifth columns present the slices that are considered to lie on the opposite sides of an interface. At the length scale of the interface, the volume averages of these slices satisfy the interface conditions as also expressed in (3.6).

Kinematical compatibility and traction equilibrium conditions are enforced at the length scale of the interfaces such that

$$\begin{aligned}
& \left(\frac{1}{\nu_s} \sum_{i \in s(I)} \nu^i \mathbf{F}^i - \frac{1}{\nu_f} \sum_{i \in f(I)} \nu^i \mathbf{F}^i \right) \mathbf{S}^j = \mathbf{0} \\
& \left(\frac{1}{\nu_s} \sum_{i \in s(I)} \nu^i \mathbf{F}^i - \frac{1}{\nu_f} \sum_{i \in f(I)} \nu^i \mathbf{F}^i \right) \mathbf{T}^j = \mathbf{0} \\
& \left(\frac{1}{\nu_s} \sum_{i \in s(I)} \nu^i \mathbf{P}^i - \frac{1}{\nu_f} \sum_{i \in f(I)} \nu^i \mathbf{P}^i \right) \mathbf{N}^j = \mathbf{0} \quad \forall j \in \{0, \dots, N^S - 2\}
\end{aligned} \tag{3.6}$$

where ν^I is the volume fraction of slice I in the reference configuration. $s(J)$ and $f(J)$ are the sets of the slice numbers that are considered to lie on the counter-clockwise and clockwise-sides of the interface J , as given in Table 3.1. $\nu_s = \sum_{i \in s(I)} \nu^i$ and $\nu_f = \sum_{i \in f(I)} \nu^i$ are the volume fraction of the slice-sets $s(I)$ and $f(I)$, respectively. For a given interface J , $\mathbf{N}^J$ is the unit normal and $\mathbf{S}^J, \mathbf{T}^J$ are a pair of linearly independent unit vectors that span the interface plane. Notice that half of the interfaces of the model, the even numbered interfaces, belong to the rank-1 laminates. For these interfaces both $s(I)$ and $f(I)$ have only single elements, and (3.6) reduces to the interface equations of the rank-1 laminates, i.e. (3.1) and (3.2). Therefore, the kinematical compatibility and equilibrium conditions are exactly satisfied between the adjacent slices on the even numbered interfaces.

The macroscopic deformation gradient $\bar{\mathbf{F}}$ is an input to the model and should be equal to the area average of the deformation gradient field of the model

$$\bar{\mathbf{F}} - \sum_{i=0}^{N^S} \nu^i \mathbf{F}^i = \mathbf{0}, \tag{3.7}$$

while the macroscopic first Piola-Kirchhoff stress $\bar{\mathbf{P}}$ is an output of the model

$$\bar{\mathbf{P}} - \sum_{i=0}^{N^S} \nu^i \mathbf{P}^i = \mathbf{0} \tag{3.8}$$

obtained by taking the volume average of the stress field of the model. The conditions (3.6) and (3.7) represent $9(N^S - 1) + 9 = 9N^S$ equations. Therefore, the number of the unknowns and the equations of the model are equal.

A shortcoming of the model, and a possible resolution is explained in this paragraph. According to the schematic views in Figure 3.4, all interfaces of the model, except the interface of the highest-rank laminate, are between two slices. This interface would no longer be necessary if the model is centrosymmetric. The orientation assignment of the crystalline slices could be modified to add centrosymmetry to the model, without deviating from the employed conditions of the crystalline texture of the PE described in Section 2.3. By applying right handed and left handed rotations respectively on centrosymmetric crystalline regions around b lattice axis, instead of only right-handed rotations, the models presented in Figure 3.4 could possess symmetry with respect to these interfaces (interfaces 1, 3, 7). Then only half of the model needs to be modeled, since both halves at the largest length scale would be identical.

3.1.2.2 Results of the Model

In Figure 3.4 even and odd numbered slices are amorphous and crystalline, respectively. For the amorphous and crystalline phases, the constitutive models and the material parameters presented in Section 2.2 are employed. PE crystal is oriented in the crystalline slices, similar to the disk-like FE model, such that the lattice axis b is in the x-y plane presented in Figure 3.4, and is along the radial direction at the center of the slice. In the crystalline regions of the FE model, the orientation of the lattice at an integration point depends on the coordinates of the integration point. Therefore, when the model is composed of a fewer number of slices, the crystalline orientations are considerably different than the FE model. In Figure 3.5, the response converges with the increasing number of slices, similar to the FE model in Section 2.3.

Since the response changes with the order of lamination, in Figure 3.6 uniaxial tension response of the models with 32 slices are presented, where the angle between the interface of the highest-rank laminate and the loading direction is $n\frac{\pi}{8}$. As expected, the stress-strain curves coincide for $n = 0$ and $n = 8$.

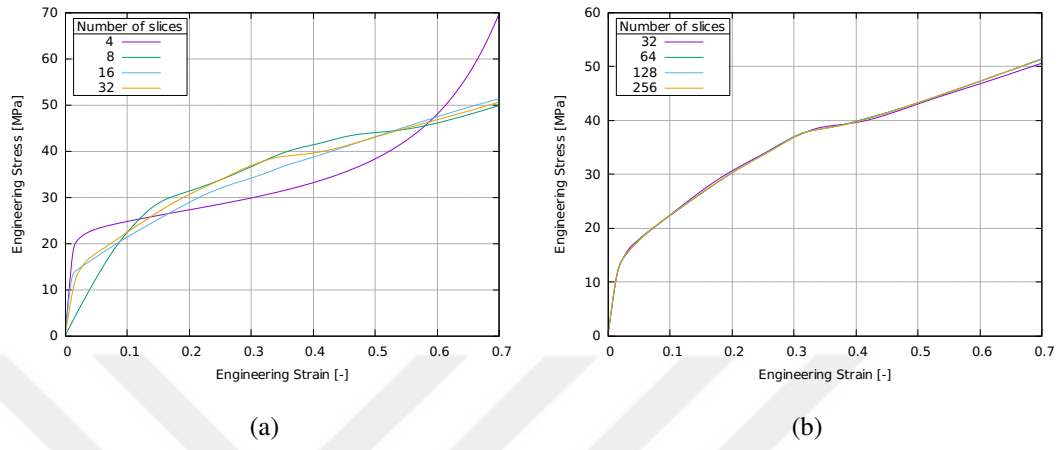


Figure 3.5: Variation of response under uniaxial tension with number of slices

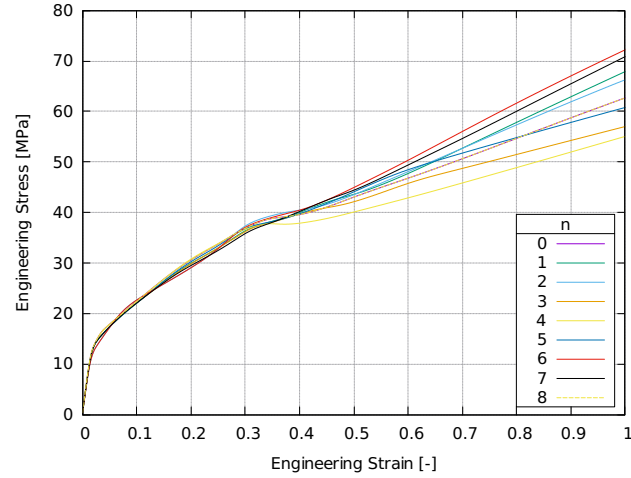


Figure 3.6: Variation of response with the orientation of the interface of the highest-rank laminate, for 32 slices. Angle between the loading direction and the interface is $\frac{\pi}{8}n$.

A preliminary comparison is presented in Figure 3.7, between the responses of the preliminary model and the disk-like FEM model in Section 2.3. The preliminary model has 64 slices and the interface of the highest-rank laminate is aligned with the loading direction. The obtained resemblance is promising.

In Figure 3.8 deformed views of the models with different number of slices at $\bar{F}_{11} = 1.7$ are presented. In the figure, dark and light colored regions are amorphous and crystalline, respectively. The loading direction and the top-most interface are aligned. In Figure 3.8, kinematical compatibility between adjacent regions is particularly worse on the interfaces of the top 2 high-rank laminates of the model. Between the adjacent regions, kinematical compatibility is exactly satisfied on the even numbered interfaces, all of which lie at the lowest length scale.

For the model with 32 slices, the variation of the deformed views with the orientation of the interface of the highest-rank laminate are presented in Figure 3.9. The model is not centrosymmetric because of the crystalline lattice orientation has a right handed rotation along the outward radial directions.

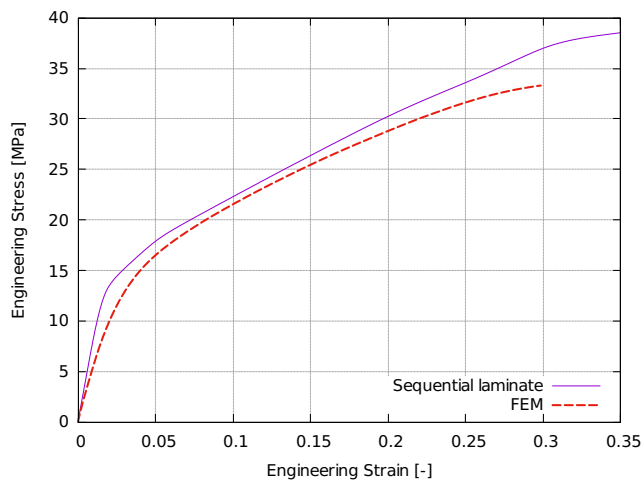


Figure 3.7: Responses of the model with 64 slices and the disk-like FE spherulite model

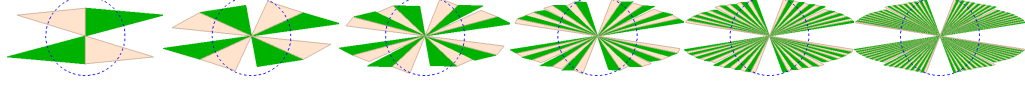


Figure 3.8: Deformed views of the models with 4 and up to 128 number of slices

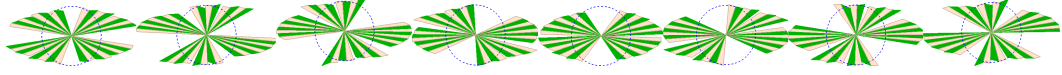


Figure 3.9: Variation of the deformed views with the orientation of the interface of the highest-rank laminate . Angle between the loading direction and the interface is $n\frac{\pi}{8}$. In the figure n increases from 0 to 7 from left to right.

The modeling approach for the disk-like spherulite morphology and the results presented in this section are only preliminary, and are not realistic since the separation of the length scales assumption that also depends on the MMM principle of Hashin is not fulfilled. Additionally, the response of the model depends on the order of the lamination. In the following, we will present approaches without these limitations, where all regions could be considered to lie at the same length scale.

3.2 General Formulation

Consider that a composite is discretized into N number of regions with planar interfaces. The volume fractions of the regions in the reference configuration are ν^I , where

$$\sum_{i=1}^N \nu^i = 1, \quad (3.9)$$

and the regions are referred to by a number $I \in \{1, \dots, N\}$. The deformation gradient $\mathbf{F}^I$ and the first Piola-Kirchhoff stress tensor

$$\mathbf{P}^I = \frac{\partial \Psi^I}{\partial \mathbf{F}^I} \quad (3.10)$$

fields of each region I are taken as uniform. Moreover, in (3.10), $\Psi^I(\mathbf{F}^I, \mathbf{I}^I)$ and $\mathbf{I}^I$ denote the free energy function and the associated internal state variables of the region, respectively. The deformation gradient $\mathbf{F}^I$ is taken as a function of the macroscopic deformation gradient $\bar{\mathbf{F}}$ and a set of vectors $\mathbf{m}^J$ $0 \leq J \leq L$

$$\hat{\mathbf{F}}^I(\bar{\mathbf{F}}, \mathbf{m}^1, \dots, \mathbf{m}^L) \quad \forall I \in \{1, 2, \dots, N\}, \quad (3.11)$$

where L is arbitrary and typically increases with the number of regions N . The macroscopic first Piola-Kirchhoff stress tensor $\bar{\mathbf{P}}$ and the deformation gradient $\bar{\mathbf{F}}$ are

$$\bar{\mathbf{P}} = \sum_{i=1}^N \nu^i \mathbf{P}^i \quad \text{and} \quad \bar{\mathbf{F}} = \sum_{i=1}^N \nu^i \mathbf{F}^i \quad (3.12)$$

according to the volume averaging framework, where the macroscopic first Piola-Kirchhoff stress tensor $\bar{\mathbf{P}}$ is also given as

$$\bar{\mathbf{P}} = \frac{\partial \bar{\Psi}}{\partial \bar{\mathbf{F}}} \quad (3.13)$$

in terms of the effective free energy function that we define as

$$\bar{\Psi} = \min_{\mathbf{m}^J} \sum_{J \in \{1, \dots, L\}} \nu^J \Psi^J(\mathbf{F}^J, \mathbf{I}^J) . \quad (3.14)$$

According to the $\hat{\mathbf{F}}^I$ in (3.11), $\bar{\Psi}$ is a function of the macroscopic deformation gradient $\bar{\mathbf{F}}$ and the internal state variables $\mathbf{I}^I$

$$\hat{\Psi}(\bar{\mathbf{F}}, \mathbf{I}^1, \dots, \mathbf{I}^N)$$

considering the minimization in (3.14) with respect to the vectors $\mathbf{m}^J$, and

$$\frac{\partial \bar{\Psi}}{\partial \bar{\mathbf{F}}} = \sum_{i=1}^N \nu^i \mathbf{P}^i : \frac{\partial \mathbf{F}^i}{\partial \bar{\mathbf{F}}} . \quad (3.15)$$

According to $\bar{\Psi}$ in (3.14), the definitions of $\bar{\mathbf{P}}$ in (3.12) and (3.13) are the same when

$$\frac{\partial \mathbf{F}^I}{\partial \bar{\mathbf{F}}} = \mathbb{I} \quad \forall I \in \{1, 2, \dots, N\} ,$$

where $\mathbb{I}$ is the fourth order identity tensor. Similar to the rank-1 laminates, it is assumed that each $\mathbf{m}^J$ vector appears only as a tensor product $\mathbf{m}^J \otimes \mathbf{M}^{I,J}$ in $\hat{\mathbf{F}}^I$, where the $\mathbf{M}^{I,J}(\nu^1, \dots, \nu^N, \mathbf{N}^1, \dots, \mathbf{N}^N)$ vectors are predetermined in reference coordinates, and $\mathbf{N}^J$ is the unit normal vector of the interface with number J . Therefore, the deformation gradients (3.11) are taken in the form

$$\hat{\mathbf{F}}^I(\bar{\mathbf{F}}, \mathbf{m}^1, \dots, \mathbf{m}^L) = \bar{\mathbf{F}} + \sum_{j=1}^L \mathbf{m}^j \otimes \mathbf{M}^{I,j} \quad \forall I \in \{1, 2, \dots, N\} . \quad (3.16)$$

It is important to observe that $\frac{\partial \mathbf{F}^I}{\partial \mathbf{m}^J}$ are known, and are constant since $\mathbf{F}^I$ are linear functions of $\mathbf{m}^J$ according to (3.16) and $\mathbf{M}^{I,J}$ are in reference coordinates. As

also expressed in the last paragraph of this section with examples, formulations with different properties could be obtained by defining $\hat{\mathbf{F}}^I$ functions in the form (3.16) that are also required to satisfy the second of (3.12). It could be emphasized once more that the explicit expressions of $\mathbf{M}^{I,J}$ in terms of the volume fractions ν^I and interface normals $\mathbf{N}^J$ are considered as known, and are unique to a specific formulation. In other words, the expression of $\mathbf{M}^{I,J}$ are specifically chosen in each formulation according to the desired characteristics. (3.5) of rank-1 laminates is an example that obeys (3.16), and in that particular case, all $\mathbf{M}^{I,J}$ vectors are parallel.

The necessary conditions

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^J} = \mathbf{0} \quad \forall J \in \{1, 2, \dots, L\} . \quad (3.17)$$

of the optimization problem in $\bar{\Psi}$ definition (3.14) evaluates to

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^J} = \sum_{i=1}^N \nu^i \mathbf{P}^i : \frac{\partial \mathbf{F}^i}{\partial \mathbf{m}^J} = \mathbf{0} \quad \forall J \in \{1, 2, \dots, L\} , \quad (3.18)$$

according to (3.17), and simplifies to

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^J} = \sum_{i=1}^N \nu^i \mathbf{P}^i \mathbf{M}^{i,J} = \mathbf{0} \quad \forall J \in \{1, 2, \dots, L\} , \quad (3.19)$$

according to (3.16). For a given $\bar{\mathbf{F}}$ and the set of internal state variables $\mathbf{I}^I$, the values of $\mathbf{m}^J$ vectors are solved to obtain the set of stress tensors $\mathbf{P}^I$ that satisfy (3.18), where $1 \leq I \leq N$. The optimality condition (3.19) that the stress set $\mathbf{P}^I$ $1 \leq I \leq N$ satisfies is independent of time, since $\frac{\partial \mathbf{F}^I}{\partial \mathbf{m}^J}$ are constant. In other words, the rate of change of (3.18) with time is

$$\sum_{i=1}^N \nu^i \dot{\mathbf{P}}^i : \frac{\partial \mathbf{F}^i}{\partial \mathbf{m}^J} = \sum_{i=1}^N \nu^i \dot{\mathbf{P}}^i : \mathbf{M}^{i,J} = \mathbf{0} \quad \forall J \in \{1, 2, \dots, L\} . \quad (3.20)$$

In Section 3.2.1, (3.20) will be employed to show that the general formulation is a consistent averaging scheme. It should be stated that, even though the minimization operation is employed in (3.14), we only enforce the stationarity condition (3.17) of optimization. Lopez-Pamies and Castaneda [67] also relax the minimization condition with the stationarity condition, and discuss the implications.

Note that the rank-1 laminate also lies within this formulation, where $\mathbf{M}^{1,1} = -\nu^2 \mathbf{N}$ and $\mathbf{M}^{2,1} = \nu^1 \mathbf{N}$ for $N = 2$ according to (3.5), and $\mathbf{N}$ is also constant even when

$N > 2$ among all $\mathbf{F}^I$ and $\mathbf{m}^J$. Interfaces are taken as an abstract concept in this section, rather than physical features of a material, as follows. All interfaces are not required to be associated with a vector $\mathbf{m}^J$. Neither the number of interfaces, nor the number of $\mathbf{m}^J$ vectors, i.e. L , is limited, and choosing $L = 0$ would result in a Taylor-type homogenization. In that case the number of interfaces or orientation of their normal vectors $\mathbf{N}^I$ does not play any role in the formulation. On the other hand, for a given N , there may be a large L and a large number of linearly independent $\mathbf{M}^{I,J}$ vectors that may result in a Sachs-type homogenization. This statement is partially supported by the results in Section 3.5, and needs verification, while the reasoning is as follows. Since (3.19) is satisfied for any and arbitrary $\mathbf{M}^{I,J}$ when the stresses $\mathbf{P}^I$ are the same for all I , Sachs-type homogenization $\mathbf{P}^I = \bar{\mathbf{P}}, \forall I \in \{1, 2, \dots, N\}$ also satisfies (3.19), and a large L could provide the necessary kinematical freedom between $\mathbf{F}^I$. It should be noted that the linear independence of $\mathbf{M}^{I,J}$ is clearly not a requirement of the formulation, considering the rank-1 laminate could also be expressed in this formulation.

3.2.1 Micro– and Macro– Work Rate Balance and Consistency as an Averaging Scheme

For non-linear problems, the stress power between the length scales is conserved when

$$\bar{\mathbf{P}} : \dot{\bar{\mathbf{F}}} = \sum_{i=1}^N \nu^i \mathbf{P}^i : \dot{\mathbf{F}}^i. \quad (3.21)$$

Moreover, an averaging scheme is considered to be consistent when the values of the $\bar{\mathbf{F}}, \bar{\mathbf{P}}$ pairs in

$$\bar{\mathbf{P}} = \frac{\partial \bar{\Psi}}{\partial \bar{\mathbf{F}}} \quad \text{and} \quad \bar{\mathbf{F}} = \frac{\partial \bar{\Phi}}{\partial \bar{\mathbf{P}}},$$

are the same, where

$$\begin{aligned} \bar{\Psi}(\bar{\mathbf{F}}, \mathbf{I}^1, \dots, \mathbf{I}^N) &= \langle \Psi(\mathbf{F}, \mathbf{I}) \rangle, \quad \bar{\Phi}(\bar{\mathbf{P}}, \mathbf{I}^1, \dots, \mathbf{I}^N) = \langle \Phi(\mathbf{P}, \mathbf{I}) \rangle \\ \text{and} \quad \bar{\mathbf{F}} &= \langle \mathbf{F} \rangle, \quad \bar{\mathbf{P}} = \langle \mathbf{P} \rangle \end{aligned} \quad (3.22)$$

In this section, the brackets $\langle \cdot \rangle$ indicates volume averaging in the micro-scale. For instance $\langle \Psi(\mathbf{F}, \mathbf{I}) \rangle = \sum_{i=1}^N \nu^i \Psi^i(\mathbf{F}^i, \mathbf{I}^i)$ in (3.22). In general, the complementary strain energy function $\Phi(\mathbf{P}, \mathbf{I})$ is obtained from a known strain energy function

$\Psi(\mathbf{F}, \mathbf{I})$ with the Legendre transformation

$$\Phi(\mathbf{P}, \mathbf{I}) = \mathbf{P} : \mathbf{F} - \Psi(\mathbf{F}, \mathbf{I}) , \quad (3.23)$$

where

$$\frac{\partial \Phi}{\partial \mathbf{I}} = - \frac{\partial \Psi}{\partial \mathbf{I}} . \quad (3.24)$$

Observe that, the differential of complementary free energy $d\Phi(\mathbf{P}, \mathbf{I}) = \mathbf{F} : d\mathbf{P} + \frac{\partial \Phi}{\partial \mathbf{I}} \cdot d\mathbf{I}$ is obtained through the Legendre transformation, when the differential of free energy $d\Psi = \mathbf{P} : d\mathbf{F} + \frac{\partial \Psi}{\partial \mathbf{I}} \cdot d\mathbf{I}$ is substituted into the differential of (3.23)

$$d\Phi = \mathbf{P} : d\mathbf{F} + \mathbf{F} : d\mathbf{P} - d\Psi .$$

For the homogeneous equivalent composites that could be obtained within the general formulation of Section 3.2, we already have the relationship

$$\frac{\partial \bar{\psi}}{\partial \bar{\mathbf{F}}} = \bar{\mathbf{P}} ,$$

and, in the following, we will show that

$$\bar{\mathbf{P}} : \dot{\bar{\mathbf{F}}} = \langle \mathbf{P} : \dot{\mathbf{F}} \rangle , \quad \bar{\mathbf{F}} : \dot{\bar{\mathbf{P}}} = \langle \mathbf{F} : \dot{\mathbf{P}} \rangle \quad \text{and finally} \quad \frac{\partial \bar{\phi}}{\partial \bar{\mathbf{P}}} = \bar{\mathbf{F}} ,$$

which would also satisfy the corresponding Legendre transformation at the macroscopic scale

$$\bar{\phi}(\bar{\mathbf{P}}) = \bar{\mathbf{P}} : \bar{\mathbf{F}} - \bar{\psi}(\bar{\mathbf{F}}) .$$

Let us first show that the stress power is conserved between the length scales, i.e. (3.21). According to the first of (3.22), the rate of change of the effective free energy with respect to time could be written as

$$\begin{aligned} \dot{\bar{\Psi}} &= \sum_{i=1}^N \left(\frac{\partial \bar{\Psi}}{\partial \mathbf{F}^i} : \dot{\mathbf{F}}^i + \frac{\partial \bar{\Psi}}{\partial \mathbf{I}^i} \cdot \dot{\mathbf{I}}^i \right) \\ &= \sum_{i=1}^N \nu^i \left(\frac{\partial \Psi^i}{\partial \mathbf{F}^i} : \dot{\mathbf{F}}^i \right) + \sum_{i=1}^N \frac{\partial \bar{\Psi}}{\partial \mathbf{I}^i} \cdot \dot{\mathbf{I}}^i + \sum_{j=1}^L \frac{\partial \bar{\Psi}}{\partial \mathbf{m}^j} \cdot \dot{\mathbf{m}}^j , \end{aligned} \quad (3.25)$$

where in the second line the dependence of $\mathbf{F}^I$ on the macroscopic deformation gradient $\bar{\mathbf{F}}$ and $\mathbf{m}^J$ vectors is introduced, according to (3.16) as follows. In the second line, the first term employs that for any region I , $\mathbf{F}^I$ only appears in Ψ^I in the first of (3.22), and in the last term it is not necessary to write the derivatives $\frac{\partial \bar{\Psi}}{\partial \mathbf{F}^I}$ explicitly.

According to (3.17), the last term in (3.25) is zero, and substitution of respectively (3.10) and (3.12) provides

$$\dot{\bar{\Psi}} = \bar{\mathbf{P}} : \dot{\bar{\mathbf{F}}} + \sum_{i=1}^N \nu^i \frac{\partial \Psi^i}{\partial \mathbf{I}^i} \cdot \dot{\mathbf{I}}^i. \quad (3.26)$$

By subtracting (3.25) from (3.26), we obtain the desired work criterion (3.21).

It is known that, $\bar{\mathbf{F}} : \dot{\bar{\mathbf{P}}} = \langle \mathbf{F} : \dot{\mathbf{P}} \rangle$ is trivially satisfied by $\bar{\mathbf{P}} : \dot{\bar{\mathbf{F}}} = \langle \mathbf{P} : \dot{\mathbf{F}} \rangle$ when the stress field is in equilibrium and the strain field is compatible [76]. However, neither the equilibrium nor the compatibility is postulated in Section 3.2. Nevertheless, the general formulation in Section 3.2 also satisfies this condition and provides consistent averaging schemes as follows. According to (3.23)

$$\bar{\Psi} + \bar{\Phi} = \sum_{i=1}^N \nu^i \mathbf{P}^i : \mathbf{F}^i, \quad (3.27)$$

and the rate of change of (3.27) with time is

$$\begin{aligned} \dot{\bar{\Psi}} + \dot{\bar{\Phi}} &= \sum_{i=1}^N \nu^i \left(\dot{\mathbf{P}}^i : \mathbf{F}^i + \mathbf{P}^i : \dot{\mathbf{F}}^i \right) \\ &= \sum_{i=1}^N \nu^i \left[\dot{\mathbf{P}}^i : \left(\bar{\mathbf{F}} + \sum_{j=1}^L \mathbf{m}^j \otimes \mathbf{M}^{i,j} \right) + \mathbf{P}^i : \left(\dot{\bar{\mathbf{F}}} + \sum_{j=1}^L \dot{\mathbf{m}}^j \otimes \mathbf{M}^{i,j} \right) \right] \\ &= \dot{\bar{\mathbf{P}}} : \bar{\mathbf{F}} + \bar{\mathbf{P}} : \dot{\bar{\mathbf{F}}} + \sum_{i=1}^N \nu^i \left[\sum_{j=1}^L \mathbf{m}^j \cdot \left(\dot{\mathbf{P}}^i \mathbf{M}^{i,j} \right) + \sum_{j=1}^L \dot{\mathbf{m}}^j \cdot \left(\mathbf{P}^i \mathbf{M}^{i,j} \right) \right] \\ &= \dot{\bar{\mathbf{P}}} : \bar{\mathbf{F}} + \bar{\mathbf{P}} : \dot{\bar{\mathbf{F}}} + \sum_{j=1}^L \left[\mathbf{m}^j \cdot \sum_{i=1}^N \nu^i \dot{\mathbf{P}}^i \mathbf{M}^{i,j} + \dot{\mathbf{m}}^j \cdot \sum_{i=1}^N \nu^i \mathbf{P}^i \mathbf{M}^{i,j} \right] \\ &= \dot{\bar{\mathbf{P}}} : \bar{\mathbf{F}} + \bar{\mathbf{P}} : \dot{\bar{\mathbf{F}}} \end{aligned}$$

where the second line is according to (3.16), and the identities (3.19) and (3.20) are employed in the fourth line. Additionally, substituting first (3.26) and then (3.24) provides

$$\dot{\bar{\Phi}} = \bar{\mathbf{F}} : \dot{\bar{\mathbf{P}}} + \sum_{i=1}^N \nu^i \frac{\partial \Phi^i}{\partial \mathbf{I}^i} \cdot \dot{\mathbf{I}}^i. \quad (3.28)$$

Comparison of (3.28) with

$$\dot{\bar{\Phi}} = \sum_{i=1}^N \nu^i \left(\mathbf{F}^i : \dot{\mathbf{P}}^i + \frac{\partial \Phi^i}{\partial \mathbf{I}^i} \cdot \dot{\mathbf{I}}^i \right)$$

that follows from the second of (3.22) reveals that

$$\bar{\mathbf{F}} : \dot{\bar{\mathbf{P}}} = \sum_{i=1}^N \nu^i \mathbf{F}^i : \dot{\mathbf{P}}^i .$$

Moreover,

$$\frac{\partial \bar{\Phi}}{\partial \bar{\mathbf{P}}} = \bar{\mathbf{F}}$$

according to the differential of (3.28)

$$d\bar{\Phi} = \bar{\mathbf{F}} : d\bar{\mathbf{P}} + \sum_{i=1}^N \nu^i \frac{\partial \Phi^i}{\partial \mathbf{I}^i} \cdot d\mathbf{I}^i .$$

Therefore, consistent averaging schemes are obtained with the proposed general formulation. It is also numerically verified that the models solved with the approaches based on the general formulation satisfy the

$$\bar{\mathbf{P}} : \dot{\bar{\mathbf{F}}} = \sum_{i=1}^N \nu^i \mathbf{P}^i : \dot{\mathbf{F}}^i \quad \text{and} \quad \bar{\mathbf{P}} : \bar{\mathbf{F}} = \sum_{i=1}^N \nu^i \mathbf{P}^i : \mathbf{F}^i$$

identities.

3.3 Kinematically Compatible Formulation

In this section, compared to Section 3.1.2, we aim to satisfy the kinematical compatibility on all interfaces of the model. We extend the formulation of the rank-1 laminate such that more than one lamination direction could be employed at the single length scale. In that case, the kinematic compatibility between the regions exist when (3.3) is written for all of the $N - 1$ number of interfaces

$$\mathbf{F}^{I+1} - \mathbf{F}^I = \mathbf{m}^I \otimes \mathbf{N}^I \quad \forall I \in \{1 \dots N - 1\} \quad (3.29)$$

between the N number of regions, where $\mathbf{F}^I$ are the deformation gradients of the regions, and $\mathbf{N}^I$ are the lamination directions also presented in Figure 3.10 for $N = 3$. While there are two regions and one jump vector $\mathbf{m}$ in the rank-1 laminate in Figure 3.3, there are N regions and $N - 1$ jump vectors $\mathbf{m}^I$ in this case.

In terms of the deformation gradient of the first region $\mathbf{F}^1$ and $\mathbf{m}^I$

$$\mathbf{F}^I = \mathbf{F}^1 + \sum_{i=1}^{I-1} \mathbf{m}^i \otimes \mathbf{N}^i . \quad (3.30)$$

according to (3.29), where

$$\mathbf{F}^1 = \bar{\mathbf{F}} - \sum_{i=1}^{N-1} [A^i \mathbf{m}^i \otimes \mathbf{N}^i]$$

could be obtained from the substitution of (3.30) into the second of (3.12), and

$$A^I = \left(\sum_{i=I+1}^N \nu^i \right). \quad (3.31)$$

Then, the expression for the deformation gradients of the regions, which satisfies both the second of (3.12) and (3.29) for arbitrary $\bar{\mathbf{F}}$ and $\mathbf{m}^I$ are

$$\mathbf{F}^I = \bar{\mathbf{F}} - \sum_{i=1}^{N-1} [A^i \mathbf{m}^i \otimes \mathbf{N}^i] + \sum_{i=1}^{I-1} \mathbf{m}^i \otimes \mathbf{N}^i \quad \forall I \in \{1 \dots N-1\}. \quad (3.32)$$

We employ (3.32), in the formulation presented in Section 3.2. Therefore, instead of the static equilibrium which is satisfied when $(\mathbf{P}^{i+1} - \mathbf{P}^i) \mathbf{N}^i = \mathbf{0}$, $\forall i \in \{1, \dots, N-1\}$, $\mathbf{m}^I$ are solved to satisfy the necessary conditions for the optimality of the effective free energy

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^J} = \mathbf{0} \quad \forall J \in \{1, \dots, N-1\}. \quad (3.33)$$

We find

$$\left(\frac{\partial \mathbf{F}^I}{\partial \mathbf{m}^J} \right)_{ijk} = \begin{cases} -A^J \delta_{ik} N_j^J & J > I \\ (1 - A^J) \delta_{ik} N_j^J & \text{otherwise} \end{cases} \quad (3.34)$$

according to (3.32), which is required in (3.15). By substituting (3.34) into (3.18) we obtain

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = \left(-A^I \sum_{i=1}^N \nu^i \mathbf{P}^i + \sum_{i=I+1}^N \nu^i \mathbf{P}^i \right) \mathbf{N}^I = \mathbf{0} \quad \forall I \in \{1, \dots, N-1\}, \quad (3.35)$$

which should be satisfied by the vectors $\mathbf{m}^I$ as discussed in Section (3.2). Below, we will simplify (3.35) and give a physical interpretation that slightly resembles the

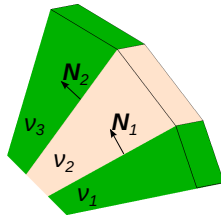


Figure 3.10: Structure with three regions and two lamination directions

interface conditions of sequential laminates in the third line of (3.6). By employing (3.12) and (3.31) in (3.35), we obtain

$$\boxed{\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = \left(\sum_{i=I+1}^N \nu^i \tilde{\mathbf{P}}^i \right) \mathbf{N}^I = \mathbf{0} \quad \forall I \in \{1, 2, \dots, N-1\}} \quad (3.36)$$

where

$$\tilde{\mathbf{P}}^I = \mathbf{P}^I - \bar{\mathbf{P}} \quad (3.37)$$

is the fluctuating part of the first Piola-Kirchhoff stress in region I . Note that according to (3.37)

$$\sum_{i=1}^N \nu^i \tilde{\mathbf{P}}^i = \mathbf{0}, \quad (3.38)$$

therefore,

$$\left(\sum_{i=I+1}^N \nu^i \tilde{\mathbf{P}}^i \right) \mathbf{N}^I = - \left(\sum_{i=1}^I \nu^i \tilde{\mathbf{P}}^i \right) \mathbf{N}^I = \mathbf{0} \quad \forall I \in \{1, 2, \dots, N-1\}. \quad (3.39)$$

Then $\mathbf{m}^I$ could also be solved to satisfy

$$\boxed{\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = \left(\sum_{i=1}^I \nu^i \tilde{\mathbf{P}}^i \right) \mathbf{N}^I = \mathbf{0} \quad \forall I \in \{1, 2, \dots, N-1\}} \quad (3.40)$$

instead of (3.36).

Although (3.36) and (3.40) can be used to find the values of $\mathbf{m}^I$, they are not explicit expression in terms of the stresses of the slices $\mathbf{P}^I$, since they refer to $\bar{\mathbf{P}}$. Let us substitute (3.37) and the first of (3.12) into (3.40)

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = \left[\sum_{i=1}^I \nu^i \mathbf{P}^i - \left(\sum_{i=1}^I \nu^i \right) \sum_{i=1}^N \nu^i \mathbf{P}^i \right] \mathbf{N}^I = \mathbf{0} \quad \forall I \in \{1, 2, \dots, N-1\}$$

and simplify the common terms

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = \left[\left(1 - \sum_{i=1}^I \nu^i \right) \sum_{i=1}^I \nu^i \mathbf{P}^i - \left(\sum_{i=1}^I \nu^i \right) \sum_{i=I+1}^N \nu^i \mathbf{P}^i \right] \mathbf{N}^I = \mathbf{0} \quad \forall I \in \{1, 2, \dots, N-1\} \quad (3.41)$$

Notice that according to (3.9),

$$\left(1 - \sum_{i=1}^I \nu^i \right) = \sum_{i=I+1}^N \nu^i \quad (3.42)$$

substituting (3.42) into (3.41) and dividing (3.41) by $\left(\sum_{i=I+1}^N \nu^i\right) \left(\sum_{i=1}^I \nu^i\right)$ we obtain

$$\boxed{\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = \left[\frac{1}{\sum_{i=1}^I \nu^i} \sum_{i=1}^I \nu^i \mathbf{P}^i - \frac{1}{\sum_{i=I+1}^N \nu^i} \sum_{i=I+1}^N \nu^i \mathbf{P}^i \right] \mathbf{N}^I = \mathbf{0}} \quad (3.43)$$

$$\forall I \in \{1, 2, \dots, N-1\}$$

Note that the first and second terms on in (3.43) are the volume average of the stresses on the left and right hand-side of the interface I , where the interface I is considered to separate the model into two halves. Therefore, according to (3.43), the traction vectors computed with the average stress of the regions on the left hand side of an interface is required to be equal to the average stress of the regions on the right hand side of the interface. Notice also that for $N = 2$, conditions (3.36), (3.40) and (3.43) reduce to the static equilibrium $(\mathbf{P}^1 - \mathbf{P}^2) \mathbf{N}^1 = \mathbf{0}$. Therefore, the rank-1 laminate formulation is recovered for $N = 2$.

3.3.1 Satisfaction of Static Equilibrium Conditions

As the number of slices increases, the normal vectors of consecutive interfaces approach to each other such that $\mathbf{N}^{K+1} \approx \mathbf{N}^K$ and $\mathbf{N}^{K-1} \approx \mathbf{N}^K$. In that case, writing (3.36) for the interfaces $K-1$ and $K+1$ provides

$$\begin{aligned} \left(\sum_{i=K+1}^N \nu^i \tilde{\mathbf{P}}^i + \nu^K \tilde{\mathbf{P}}^K \right) \mathbf{N}^{K-1} &= \mathbf{0} \\ \left(\sum_{i=K+1}^N \nu^i \tilde{\mathbf{P}}^i - \nu^{K+1} \tilde{\mathbf{P}}^{K+1} \right) \mathbf{N}^{K+1} &= \mathbf{0} \end{aligned} \quad (3.44)$$

dividing the first and second equations in (3.44) by ν^K and ν^{K+1} respectively, and replacing $\mathbf{N}^{K-1}$ and $\mathbf{N}^{K+1}$ with $\mathbf{N}^K$ in (3.44) yields

$$\begin{aligned} \left(\frac{1}{\nu^K} \sum_{i=K+1}^N \nu^i \tilde{\mathbf{P}}^i + \tilde{\mathbf{P}}^K \right) \mathbf{N}^K &\approx \mathbf{0} \\ \left(\frac{1}{\nu^{K+1}} \sum_{i=K+1}^N \nu^i \tilde{\mathbf{P}}^i - \tilde{\mathbf{P}}^{K+1} \right) \mathbf{N}^K &\approx \mathbf{0}, \end{aligned} \quad (3.45)$$

and since

$$\left(\sum_{i=K+1}^N \nu^i \tilde{\mathbf{P}}^i \right) \mathbf{N}^K = \mathbf{0}$$

according to (3.45)

$$\begin{aligned}\tilde{\mathbf{P}}^K \mathbf{N}^K &\approx \mathbf{0} \\ -\tilde{\mathbf{P}}^{K+1} \mathbf{N}^K &\approx \mathbf{0} .\end{aligned}\tag{3.46}$$

Adding the equations in (3.46) yields

$$\left(\tilde{\mathbf{P}}^K - \tilde{\mathbf{P}}^{K+1} \right) \mathbf{N}^K \approx \mathbf{0} .\tag{3.47}$$

Substituting (3.37) into (3.47) provides

$$\left(\mathbf{P}^K - \mathbf{P}^{K+1} \right) \mathbf{N}^K \approx \mathbf{0} ,$$

which indicates that the static equilibrium is approximately satisfied when $\mathbf{N}^{K+1} \approx \mathbf{N}^K$ and $\mathbf{N}^{K-1} \approx \mathbf{N}^K$. Therefore, when $\mathbf{N}^{K+1} = \mathbf{N}^K$ and $\mathbf{N}^{K-1} = \mathbf{N}^K$ the static equilibrium is exactly satisfied on the interface K , which indicates that the formulation reduces to that of the rank-1 laminates when all interfaces are parallel, as expected.

3.3.2 Bonded End Interfaces

In the formulation presented in Section 3.3, for N number of regions it is only possible to enforce the kinematic and equilibrium equations for $N - 1$ interfaces. Notice that the number of unknowns of N regions is only sufficient for the conditions of the $N - 1$ number of interfaces in Section 3.3. This situation posed a difficulty when modeling, for instance, the circular geometry presented in Figure 3.11. In Figure 3.11, there are 32 regions and 32 interfaces.

In this section, the formulation is extended to account for N number of interfaces with N number of slices. In addition to (3.29), the functional form of the deformation gradient $\hat{\mathbf{F}}^i(\bar{\mathbf{F}}, \mathbf{m}^1, \mathbf{m}^2, \dots)$ of each region of the model should also satisfy the kinematic compatibility condition between the first and the last region

$$\begin{aligned}(\mathbf{F}^N - \mathbf{F}^1) \mathbf{T}^N &= \mathbf{0} \\ (\mathbf{F}^N - \mathbf{F}^1) \mathbf{S}^N &= \mathbf{0}\end{aligned}\tag{3.48}$$

where $\mathbf{T}^N$ and $\mathbf{S}^N$ are a pair of independent vectors in the plane of the interface between regions 1 and N . Substituting $\mathbf{F}^1$ and $\mathbf{F}^N$ computed according to (3.32),

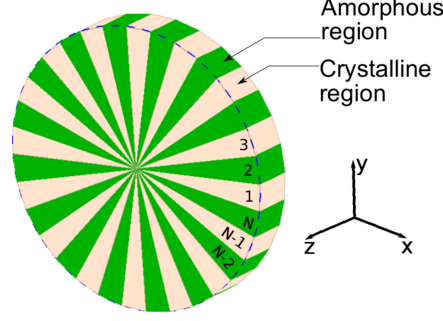


Figure 3.11: An example geometry with N number of slices and N number of interfaces

into (3.48) we obtain

$$\begin{aligned} \left(\sum_{i=1}^{N-1} \mathbf{m}^i \otimes \mathbf{N}^i \right) \mathbf{T}^N &= \mathbf{0} \\ \left(\sum_{i=1}^{N-1} \mathbf{m}^i \otimes \mathbf{N}^i \right) \mathbf{S}^N &= \mathbf{0} \end{aligned} \quad (3.49)$$

conditions. Notice that all interface normals are in the plane of the circular face of the cylinder in Figure 3.11. In this special case, one of the conditions in (3.48) is satisfied by selecting $\mathbf{S}^N$ as the normal vector of the circular face. $\mathbf{S}^N$ could be selected in this way, since this direction lies in the plane of the last interface. Then we only need to satisfy

$$\left(\sum_{i=1}^{N-1} \mathbf{m}^i \otimes \mathbf{N}^i \right) \mathbf{T}^N = \mathbf{0}. \quad (3.50)$$

To this end we single out $\mathbf{m}^{N-1}$ and take it to the other side of the equation

$$\mathbf{m}^{N-1} = \frac{-1}{(\mathbf{N}^{N-1} \cdot \mathbf{T}^N)} \left(\sum_{i=1}^{N-2} \mathbf{m}^i \otimes \mathbf{N}^i \right) \mathbf{T}^N. \quad (3.51)$$

According to the relationship between the $\mathbf{m}^{N-1}$ and the remaining $N - 2$ number of $\mathbf{m}^I$ in (3.51), $\mathbf{m}^{N-1}$ is no longer arbitrary. Therefore, there are only $N - 2$ unknown jump vectors $\mathbf{m}^I$ and

$$\hat{\mathbf{m}}^{N-1}(\mathbf{m}^1, \dots, \mathbf{m}^{N-2}) = - \sum_{i=1}^{N-2} \frac{\mathbf{N}^i \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} \mathbf{m}^i.$$

Then, the deformation gradients of each region and their derivatives with respect to

the jump vectors $\mathbf{m}^I$ are

$$\mathbf{F}^I = \bar{\mathbf{F}} - \left(\sum_{i=1}^{N-2} [A^i \mathbf{m}^i \otimes \mathbf{N}^i] + A^{N-1} \hat{\mathbf{m}}^{N-1} \otimes \mathbf{N}^{N-1} \right) + \sum_{i=1}^{I-1} \mathbf{m}^i \otimes \mathbf{N}^i$$

$$\left(\frac{\partial \mathbf{F}^I}{\partial \mathbf{m}^J} \right)_{ijk} = \begin{cases} -A^J \delta_{ik} N_j^J + \frac{\mathbf{N}^J \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} A^{N-1} \delta_{ik} N_j^{N-1} & J > I \\ (1 - A^J) \delta_{ik} N_j^J + \frac{\mathbf{N}^J \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} A^{N-1} \delta_{ik} N_j^{N-1} & \text{otherwise} \end{cases} \quad (3.52)$$

for $I < N$, while for $I = N$

$$\mathbf{F}^I = \bar{\mathbf{F}} - \left(\sum_{i=1}^{N-2} [A^i \mathbf{m}^i \otimes \mathbf{N}^i] + A^{N-1} \hat{\mathbf{m}}^{N-1} \otimes \mathbf{N}^{N-1} \right) + \sum_{i=1}^{I-2} \mathbf{m}^i \otimes \mathbf{N}^i + \hat{\mathbf{m}}^{N-1} \otimes \mathbf{N}^{N-1}$$

$$\left(\frac{\partial \mathbf{F}^I}{\partial \mathbf{m}^J} \right)_{ijk} = \begin{cases} -A^J \delta_{ik} N_j^J + \frac{\mathbf{N}^J \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} (A^{N-1} - 1) \delta_{ik} N_j^{N-1} & J > I \\ (1 - A^J) \delta_{ik} N_j^J + \frac{\mathbf{N}^J \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} (A^{N-1} - 1) \delta_{ik} N_j^{N-1} & \text{otherwise} \end{cases} . \quad (3.53)$$

Substitution of (3.52) and (3.53) into (3.15) results in

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = -A^I \sum_{i=1}^N \nu^i \mathbf{P}^i \mathbf{N}^I + \sum_{i=I+1}^N \nu^i \mathbf{P}^i \mathbf{N}^I + \frac{\mathbf{N}^I \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} \left(A^{N-1} \sum_{i=1}^N \nu^i \mathbf{P}^i - \nu^N \mathbf{P}^N \right) \mathbf{N}^{N-1} ,$$

and substituting the first of (3.12) provides

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = -A^I \bar{\mathbf{P}} \mathbf{N}^I + \sum_{i=I+1}^N \nu^i \mathbf{P}^i \mathbf{N}^I + \frac{\mathbf{N}^I \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} (A^{N-1} \bar{\mathbf{P}} - \nu^N \mathbf{P}^N) \mathbf{N}^{N-1} ,$$

where, according to (3.31) $A^{N-1} = \nu^N$, and employing (3.37) we write

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = \sum_{i=I+1}^N \nu^i \tilde{\mathbf{P}}^i \mathbf{N}^I - \frac{\mathbf{N}^I \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} \nu^N \tilde{\mathbf{P}}^N \mathbf{N}^{N-1} .$$

Then, the necessary condition for the optimality of $\bar{\Psi}$ is

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = \sum_{i=I+1}^N \nu^i \tilde{\mathbf{P}}^i \mathbf{N}^I - \frac{\mathbf{N}^I \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} \nu^N \tilde{\mathbf{P}}^N \mathbf{N}^{N-1} = \mathbf{0} \quad \forall I \in \{1, \dots, N-2\} . \quad (3.54)$$

Moreover,

$$\boxed{\sum_{i=1}^I \nu^i \tilde{\mathbf{P}}^i \mathbf{N}^I + \frac{\mathbf{N}^I \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} \nu^N \tilde{\mathbf{P}}^N \mathbf{N}^{N-1} = \mathbf{0} \quad \forall I \in \{1, \dots, N-2\}} \quad (3.55)$$

is also an optimality condition for $\bar{\Psi}$, since it is equivalent to (3.54), according to (3.38).

Notice that (3.55) is written only for $N-2$ interfaces, the interfaces with numbers from 1 to $N-2$, in this derivation. We need to show that (3.55) is satisfied on the remaining two interfaces $N-1$ and N , in order to express that there are not any special interfaces in the model and all interfaces of the model are equal. Substitution of $I = N-1$ into (3.55) yields

$$\sum_{i=1}^{N-1} \nu^i \tilde{\mathbf{P}}^i \mathbf{N}^{N-1} + \frac{\mathbf{N}^{N-1} \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} \nu^N \tilde{\mathbf{P}}^N \mathbf{N}^{N-1},$$

which evaluates to $\mathbf{0}$ according to (3.38). Substitution of $I = N$ into (3.55) yields

$$\sum_{i=1}^N \nu^i \tilde{\mathbf{P}}^i \mathbf{N}^I + \frac{\mathbf{N}^N \cdot \mathbf{T}^N}{\mathbf{N}^{N-1} \cdot \mathbf{T}^N} \nu^N \tilde{\mathbf{P}}^N \mathbf{N}^{N-1}$$

which also evaluates to $\mathbf{0}$ according to (3.38) and since $\mathbf{N}^N \cdot \mathbf{T}^N = \mathbf{0}$.

Following the same steps in Section 3.3.1 for (3.55), it could be shown that the static equilibrium is approached as the number of regions increases with the finer discretization of the disk.

3.4 Statically Equilibrated Formulation

In Section 3.3, kinematic compatibility is satisfied on all interfaces, while the static equilibrium is relaxed. In this section, we satisfy the static equilibrium on all interfaces, while the kinematic compatibility is relaxed. To this end, we will require the optimizers of the macroscopic Helmholtz free energy $\bar{\Psi}$ in (3.14) to satisfy the static equilibrium

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = (\mathbf{P}^{I+1} - \mathbf{P}^I) \mathbf{N}^I = \mathbf{0} \quad \forall I \in \{1 \dots N-1\}. \quad (3.56)$$

Then, we need to find the $\hat{\mathbf{F}}^I (\bar{\mathbf{F}}, \mathbf{m}^1, \dots, \mathbf{m}^{N-1})$ that leads to (3.56). According to (3.56), terms containing $\mathbf{m}^I$ should appear only in $\mathbf{F}^{I+1}$ and $\mathbf{F}^I$. In other words, $\mathbf{F}^I$ only depends on $\mathbf{m}^{I-1}$ and $\mathbf{m}^I$, i.e. $\hat{\mathbf{F}}^I (\bar{\mathbf{F}}, \mathbf{m}^{I-1}, \mathbf{m}^I)$. Therefore, comparison of

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = \nu^{I+1} \mathbf{P}^{I+1} : \frac{\partial \mathbf{F}^{I+1}}{\partial \mathbf{m}^I} + \nu^I \mathbf{P}^I : \frac{\partial \mathbf{F}^I}{\partial \mathbf{m}^I}, \quad (3.57)$$

which is according to (3.14), with (3.56) provides

$$\begin{aligned} \nu^{I+1} \mathbf{P}^{I+1} : \frac{\partial \mathbf{F}^{I+1}}{\partial \mathbf{m}^I} &= \mathbf{P}^{I+1} \mathbf{N}^I \\ \nu^I \mathbf{P}^I : \frac{\partial \mathbf{F}^I}{\partial \mathbf{m}^I} &= -\mathbf{P}^I \mathbf{N}^I. \end{aligned} \quad (3.58)$$

Writing (3.58) in indicial notation yields

$$\begin{aligned} P_{ij}^{I+1} \left[\nu^{I+1} \left(\frac{\partial \mathbf{F}^{I+1}}{\partial \mathbf{m}^I} \right)_{ijk} - \delta_{ik} N_j^I \right] &= 0_k \\ P_{ij}^I \left[\nu^I \left(\frac{\partial \mathbf{F}^I}{\partial \mathbf{m}^I} \right)_{ijk} + \delta_{ik} N_j^I \right] &= 0_k \end{aligned}$$

which should be satisfied for all values of $\mathbf{P}^{I+1}$ and $\mathbf{P}^I$. Therefore,

$$\begin{aligned} \nu^{I+1} \left(\frac{\partial \mathbf{F}^{I+1}}{\partial \mathbf{m}^I} \right)_{ijk} - \delta_{ik} N_j^I &= 0_k \\ \nu^I \left(\frac{\partial \mathbf{F}^I}{\partial \mathbf{m}^I} \right)_{ijk} + \delta_{ik} N_j^I &= 0_k \end{aligned}$$

and consequently

$$\begin{aligned} \left(\frac{\partial \mathbf{F}^{I+1}}{\partial \mathbf{m}^I} \right)_{ijk} &= \frac{1}{\nu^{I+1}} \delta_{ik} N_j^I \\ \left(\frac{\partial \mathbf{F}^I}{\partial \mathbf{m}^I} \right)_{ijk} &= -\frac{1}{\nu^I} \delta_{ik} N_j^I. \end{aligned}$$

Then, from the Fundamental Theorem of Calculus

$$\begin{aligned} \mathbf{F}^{I+1} &= \int \frac{\partial \mathbf{F}^{I+1}}{\partial \mathbf{m}^I} d\mathbf{m}^I = \frac{1}{\nu^{I+1}} \mathbf{m}^I \otimes \mathbf{N}^I + \hat{\mathbf{C}}^{I+1} (\bar{\mathbf{F}}, \mathbf{m}^{I+1}) \\ \mathbf{F}^I &= \int \frac{\partial \mathbf{F}^I}{\partial \mathbf{m}^I} d\mathbf{m}^I = -\frac{1}{\nu^I} \mathbf{m}^I \otimes \mathbf{N}^I + \hat{\mathbf{C}}^I (\bar{\mathbf{F}}, \mathbf{m}^{I-1}) \end{aligned} \quad (3.59)$$

where $\hat{\mathbf{C}}^I$ and $\hat{\mathbf{C}}^{I+1}$ are integration constants. Then, comparing equations in (3.59), we find that

$$\mathbf{F}^{I+1} = \int \frac{\partial \mathbf{F}^{I+1}}{\partial \mathbf{m}^I} d\mathbf{m}^I + \int \frac{\partial \mathbf{F}^{I+1}}{\partial \mathbf{m}^{I+1}} d\mathbf{m}^{I+1} + \hat{\mathbf{C}} (\bar{\mathbf{F}})$$

and in order to be consistent with (3.16), we define

$$\hat{C}(\bar{F}) = \bar{F}.$$

Then

$$\mathbf{F}^{I+1} = \bar{F} + \frac{1}{\nu^{I+1}} (\mathbf{m}^I \otimes \mathbf{N}^I - \mathbf{m}^{I+1} \otimes \mathbf{N}^{I+1})$$

and therefore

$$\mathbf{F}^I = \bar{F} + \frac{1}{\nu^I} (\mathbf{m}^{I-1} \otimes \mathbf{N}^{I-1} - \mathbf{m}^I \otimes \mathbf{N}^I) \quad \forall I \in \{2, \dots, N-1\} \quad (3.60)$$

Thus, the general definition of $\mathbf{F}^I$ to be employed as (3.16) is

$$\mathbf{F}^I = \begin{cases} \bar{F} - \frac{1}{\nu^I} \mathbf{m}^I \otimes \mathbf{N}^I & I = 1 \\ \bar{F} + \frac{1}{\nu^I} \mathbf{m}^I \otimes \mathbf{N}^I & I = N \\ \bar{F} + \frac{1}{\nu^I} (\mathbf{m}^{I-1} \otimes \mathbf{N}^{I-1} - \mathbf{m}^I \otimes \mathbf{N}^I) & \text{otherwise.} \end{cases} \quad (3.61)$$

3.4.1 Satisfaction of Kinematical Compatibility Conditions

It could be seen that, as the normal vectors of the consecutive interfaces approach to each other, the deformation gradients of the adjacent regions approach to be rank-1 connected. For instance for $N-1 > I > 2$

$$\mathbf{F}^{I+1} - \mathbf{F}^I = \left(\frac{1}{\nu^{I+1}} + \frac{1}{\nu^I} \right) \mathbf{m}^I \otimes \mathbf{N}^I - \frac{1}{\nu^{I+1}} \mathbf{m}^{I+1} \otimes \mathbf{N}^{I+1} - \frac{1}{\nu^I} \mathbf{m}^{I-1} \otimes \mathbf{N}^{I-1} \quad (3.62)$$

according to (3.61). When $\mathbf{N}^{I+1} \approx \mathbf{N}^I$ and $\mathbf{N}^{I-1} \approx \mathbf{N}^I$ then

$$\mathbf{F}^{I+1} - \mathbf{F}^I \approx \left[\left(\frac{1}{\nu^{I+1}} + \frac{1}{\nu^I} \right) \mathbf{m}^I - \frac{1}{\nu^{I+1}} \mathbf{m}^{I+1} - \frac{1}{\nu^I} \mathbf{m}^{I-1} \right] \otimes \mathbf{N}^I \quad (3.63)$$

where (3.63) is approximately a rank-1 relationship between $\mathbf{F}^I$ and $\mathbf{F}^{I+1}$. Moreover, when all $\mathbf{N}^I$ vectors lie in a plane, as also expressed in Section 3.3.2 for the disk-like spherulite model, the compatibility is exactly satisfied for a particular vector $\mathbf{S}$ that is the normal to all $\mathbf{N}^I$ vectors. This could be seen by multiplication of (3.63) with $\mathbf{S}$.

3.4.2 Formulation with Bonded End Interfaces

As in Section 3.3.2, the formulation given in Section 3.4 is extended to support N number of interfaces between N number of slices. Therefore, one more equation

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^N} = (\mathbf{P}^1 - \mathbf{P}^N) \mathbf{N}^N = \mathbf{0}$$

is introduced to (3.56), and the optimizers of the effective free energy should satisfy

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = (\mathbf{P}^{I+1} - \mathbf{P}^I) \mathbf{N}^I = \mathbf{0} \quad \forall I \in \{1 \dots N\} \quad (3.64)$$

where we employ the

$$(\cdot)^{N+1} = (\cdot)^1$$

convention in this section. Following the same steps in Section 3.4, we arrive at

$$\mathbf{F}^I = \bar{\mathbf{F}} + \frac{1}{\nu^I} (\mathbf{m}^{I-1} \otimes \mathbf{N}^{I-1} - \mathbf{m}^I \otimes \mathbf{N}^I) . \quad (3.65)$$

Notice that (3.65) is identical to (3.61), except there are not any special cases for the first and last regions. Therefore, (3.65) also provides better approximations to the rank-1 connection between the regions with finer discretization, and if there is a special vector $\mathbf{S}$ that is perpendicular to all $\mathbf{N}^I$, the kinematical compatibility is satisfied at all interfaces for $\mathbf{S}$.

3.5 Combinations of the Kinematically Compatible and Statically Equilibrated Formulations

In this section two approaches are presented that are based on the formulations presented in Sections 3.3 and 3.4.

3.5.1 Weighted Hybrid Formulation

In Sections 3.3 and 3.4, we satisfy respectively the kinematic compatibility or the static equilibrium condition, and relax the other. In this section, we introduce a scalar α that takes values between zero and one, which could be considered as a material

parameter that determines which physically motivated condition should be more precisely satisfied. In all interfaces, the precisely satisfied condition is the kinematic compatibility for $\alpha = 1$ and the static equilibrium for $\alpha = 0$.

Let us refer to the functions given in (3.32) and (3.61) as $\mathbf{F}^{K,I}$ and $\mathbf{F}^{S,I}$ respectively. Superscripts K and S denote the formulations that exactly satisfy the kinematic compatibility, and the static equilibrium respectively. In this section we define $\hat{\mathbf{F}}^I$ for N number of regions as

$$\begin{aligned}\hat{\mathbf{F}}^I(\bar{\mathbf{F}}, \mathbf{m}^1, \dots, \mathbf{m}^{N-1}) &= \alpha \hat{\mathbf{F}}^{K,I}(\bar{\mathbf{F}}, \mathbf{m}^1, \dots, \mathbf{m}^{N-1}) \\ &+ (1 - \alpha) \hat{\mathbf{F}}^{S,I}(\bar{\mathbf{F}}, \mathbf{m}^1, \dots, \mathbf{m}^{N-1}).\end{aligned}$$

Then, according to (3.15), the optimizers of the effective free energy should satisfy,

$$\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^I} = \sum_{i=1}^N \nu^i \mathbf{P}^i : \left(\alpha \frac{\partial \hat{\mathbf{F}}^{K,I}}{\partial \mathbf{m}^I} + (1 - \alpha) \frac{\partial \hat{\mathbf{F}}^{S,I}}{\partial \mathbf{m}^I} \right) \quad (3.66)$$

which evaluates to

$$\alpha \left(\sum_{i=1}^I \nu^i \tilde{\mathbf{P}}^i \right) \mathbf{N}^I + (1 - \alpha) (\mathbf{P}^{I+1} - \mathbf{P}^I) \mathbf{N}^I = \mathbf{0} \quad \forall I \in \{1, \dots, N-1\},$$

according to (3.40) and (3.56), respectively.

The number of the vectors $\mathbf{m}^I$ are not equal between the statically equilibrated and kinematically compatible formulations when the last interfaces are bonded as presented in Sections 3.3.2 and 3.4.2. Therefore, this approach could not be directly employed to the formulations with bonded end interfaces.

3.5.2 Overly Equilibrated Formulation

The formulations in Sections 3.3 and 3.4 could also be combined in such a way that the $\mathbf{m}^I$ vectors of the kinematically compatible and statically equilibrated formulations, respectively $\mathbf{m}^{K,I}$ and $\mathbf{m}^{S,I}$, are considered as separate sets of vectors

$$\begin{aligned}\hat{\mathbf{F}}^I(\bar{\mathbf{F}}, \mathbf{m}^{K,1}, \dots, \mathbf{m}^{K,N-1}, \mathbf{m}^{S,1}, \dots, \mathbf{m}^{S,N-1}) &= \hat{\mathbf{F}}^{K,I}(\bar{\mathbf{F}}, \mathbf{m}^{K,1}, \dots, \mathbf{m}^{K,N-1}) \\ &+ \hat{\mathbf{F}}^{S,I}(\bar{\mathbf{F}}, \mathbf{m}^{S,1}, \dots, \mathbf{m}^{S,N-1}).\end{aligned} \quad (3.67)$$

Then, the optimizers of the effective free energy in (3.15) should satisfy

$$\begin{aligned}\frac{\partial \bar{\Psi}}{\partial \mathbf{m}^{K,I}} &= \sum_{i=1}^N \nu^i \mathbf{P}^i : \frac{\partial \hat{\mathbf{F}}^{K,I}}{\partial \mathbf{m}^{K,I}} = \mathbf{0} \\ \frac{\partial \bar{\Psi}}{\partial \mathbf{m}^{S,I}} &= \sum_{i=1}^N \nu^i \mathbf{P}^i : \frac{\partial \hat{\mathbf{F}}^{S,I}}{\partial \mathbf{m}^{S,I}} = \mathbf{0},\end{aligned}$$

which evaluates to

$$\begin{aligned}\left(\sum_{i=1}^I \nu^i \tilde{\mathbf{P}}^i \right) \mathbf{N}^I &= \mathbf{0} \quad \forall I \in \{1, \dots, N-1\} \\ (\mathbf{P}^{I+1} - \mathbf{P}^I) \mathbf{N}^I &= \mathbf{0} \quad \forall I \in \{1, \dots, N-1\}\end{aligned} \quad (3.68)$$

as presented in Sections 3.3 and 3.4. According to (3.68), this formulation satisfies the static equilibrium exactly. Moreover the energetically motivated additional equilibrium conditions in (3.36) or (3.55), are also satisfied. This formulation could be employed for both free end interfaces and bonded end interfaces, since the equality of the number of jump vectors of the kinematically compatible and statically equilibrated formulations is not required. The number of jump vectors in (3.67) is presented for the free end interfaces, and should be changed accordingly for the bonded end interfaces case.

3.6 Results

This section focuses on the main observations of the results of the individual models in Sections 3.3.2 and 3.4.2. As a model problem the disk-like spherulite, also solved with FEM in Section 2.3 is employed. Models of the disk-like spherulite is obtained with the four different formulations presented in Sections 3.3 and 3.4. The geometry is discretized into finite number of alternating amorphous and crystalline regions as presented in Figure 3.11. Orientations of the lattice of the crystalline regions are assigned as described in Section 2.3 with a cylindrical coordinate system. Since the stress and strain fields are uniform in each region, orientations are assigned according to the coordinates of the centroids of the regions. The constitutive models and the material parameters in Section 2.2, that were also employed in Section 2.3, are used in amorphous and crystalline regions of the models.

Until Section 3.6.7, only the results of simple tension deformation mode is presented. Loading is applied in the x direction by enforcing the $\bar{F}_{11}$ component of the deformation gradient to obtain a constant engineering strain rate of $1 \times 10^{-3} \text{ s}^{-1}$. Employed driver function requires normal components of the first Piola-Kirchhoff stresses in y and z directions $\bar{P}_{22}$ and $\bar{P}_{33}$ to be equal to zero by varying the $\bar{F}_{22}$ and $\bar{F}_{33}$ components of the macroscopic deformation gradient. In Section 3.6.7, response of the models with bonded end interfaces is presented in tension, compression and simple shear deformation modes together with that of the FEM model in Section 2.3. Finally, a model for the twisted-ribbon-shaped lamellae of spherulites is presented.

3.6.1 Determination of the Necessary Number of Regions

The models presented in this chapter discretize a geometry into arbitrary number of regions. In Figures 3.12 and 3.13, variation of the tensile response with the number of regions is presented, for the kinematically compatible and statically equilibrated models with bonded end interfaces, respectively. The response of the both models converge as the number of regions increase. Deformed views of the models with different number of slices are also presented in the figures. The number of regions is selected according to the convergence of the response, similar to the mesh convergence studies of FE models.

Recall that in disk-like spherulite models, the normal vectors of all interfaces are perpendicular to the e_3 basis vector. Therefore, according to (3.16) for any slice of any disk-like spherulite model that is based on the general formulation in Section 3.2, the components F_{13} , F_{23} and F_{33} of the deformation gradient tensor are equal to the respective component of the macroscopic deformation gradient tensor $\bar{\mathbf{F}}$, i.e. $F^I e_3 = \bar{\mathbf{F}} e_3 \forall I$. As also will be expressed in Section 3.6.2, this is actually one of the necessary conditions of the kinematical compatibility between the slices. Since the deformation gradient components F_{31} , F_{32} can vary between the slices, there is a significant amount of deformation variation in the out-of-plane direction between the slices, as can be seen in the deformed views in Figures 3.12 and 3.13.

For different number of slices, the distribution of the P_{11} component of the first Piola-Kirchhoff stress tensor in the kinematically compatible and statically equilibrated

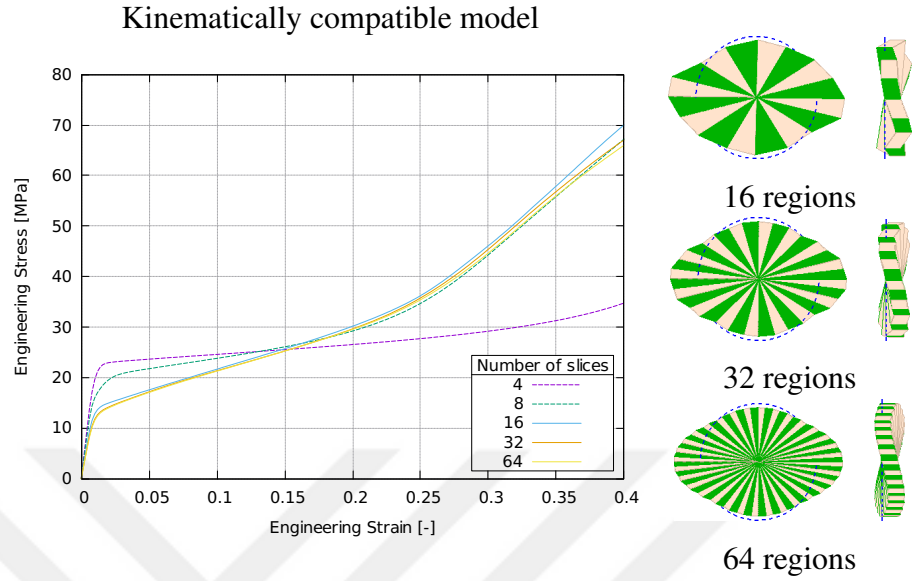


Figure 3.12: Variation of the response of the kinematically compatible model with the number of slices. On the right, deformed views are presented where the dark and light regions indicate the amorphous and crystalline phases, respectively.

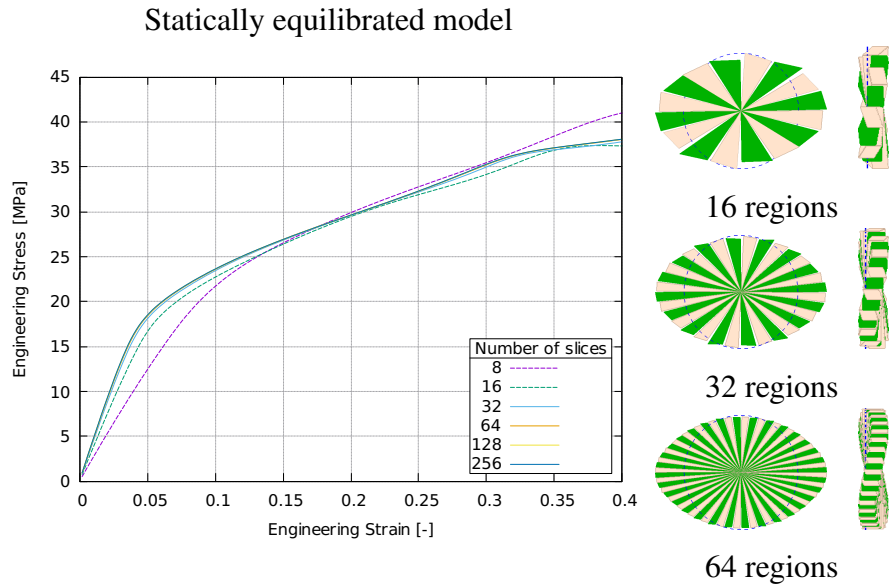


Figure 3.13: Variation of the response of the statically equilibrated model with the number of slices in simple tension. On the right, deformed views are presented where the dark and light regions indicate the amorphous and crystalline phases, respectively.

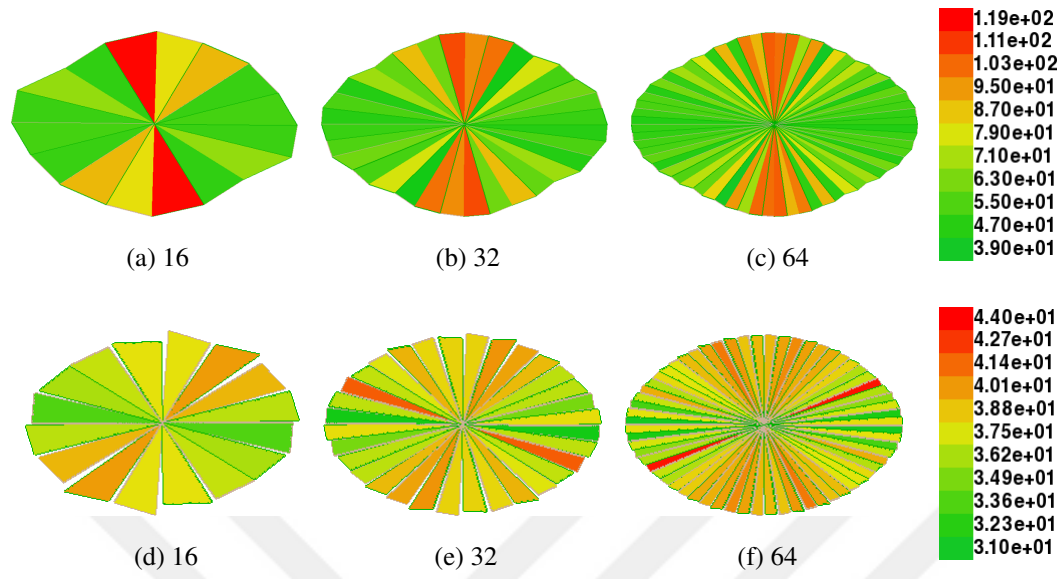


Figure 3.14: Distribution of the P_{11} stress component on the deformed geometries of the kinematically compatible (a-c) and statically equilibrated (d-f) models for different number of regions.

models are presented in Figure 3.14. In the kinematically compatible model, the P_{11} components are quite different in the equatorial region. Static equilibrium requires the P_{11} components to be the same for an interface with normal vector in the x direction in Figure 3.11. As the number of slices increases, P_{11} components become similar. In the statically equilibrated model, P_{11} components are the same between the regions separated by the interface with normal vector in the x direction for all slice numbers, as expected. The variation of the P_{33} component of the first Piola-Kirchhoff tensor among the slices is presented in Figure 3.15 for the kinematically compatible and statically equilibrated formulations. According to Figure 3.15, P_{33} stress components are quite different between the slices. Notice that, according to the conditions of simple tension the volume average of the P_{33} stress components should be equal to zero in the model. Therefore, both the positive and negative values of this component are present in Figure 3.15.

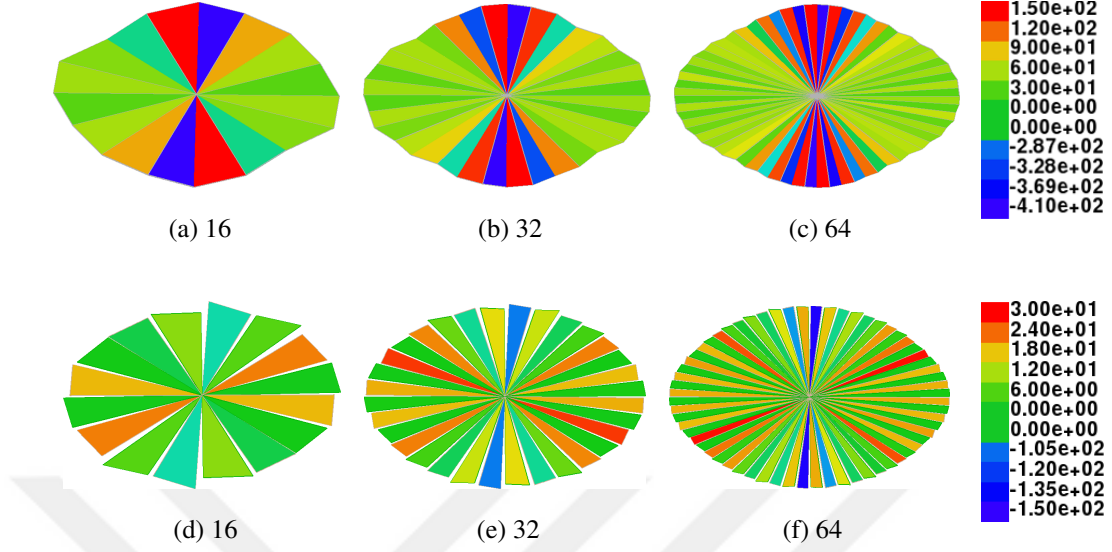


Figure 3.15: Distribution of the P_{33} stress component on the deformed geometries of the kinematically compatible (a-c) and statically equilibrated (d-f) models for different number of regions.

3.6.2 Variation of the Compatibility and Equilibrium Errors of the Formulations with the Number of Regions

For an interface between regions I and $I + 1$, errors for the static equilibrium and kinematical compatibility could respectively be defined as

$$\gamma_S = \frac{\|(\mathbf{P}^{I+1} - \mathbf{P}^I) \mathbf{N}\|}{\|\bar{\mathbf{P}}\|} \quad \text{and} \quad \gamma_K = \frac{\|(\mathbf{F}^{I+1} - \mathbf{F}^I) \mathbf{T}\| + \|(\mathbf{F}^{I+1} - \mathbf{F}^I) \mathbf{S}\|}{\|\bar{\mathbf{E}} + \mathbf{1}\|},$$

where $\mathbf{T}$ and $\mathbf{S}$ vectors span the plane of the interface such that $\mathbf{T} \times \mathbf{S} = \mathbf{N}$, and $\bar{\mathbf{E}}$ is the Green-Lagrange strain tensor computed with $\bar{\mathbf{F}}$, and $\mathbf{1}$ is the second order identity tensor. Below, the selected $\mathbf{S}$ vectors are along the out-of-plane direction of the disks, therefore as expressed in Section 3.3.2, the term that contain $\mathbf{S}$ vector in γ_K is zero.

In Figure 3.16, the variation of the maximums among the errors of all interfaces, $\max_I \gamma_S^I$ and $\max_I \gamma_K^I$, are presented for the kinematically compatible and statically equilibrated models, respectively. As the number of regions is doubled, the maximum error decreases by one half. In Figure 3.17, the sum of the errors $\sum_I \gamma_S^I$ and $\sum_I \gamma_K^I$ are presented. According to Figures 3.16 and 3.17, the errors decrease with

the increasing number of regions, while their sum remains almost constant.

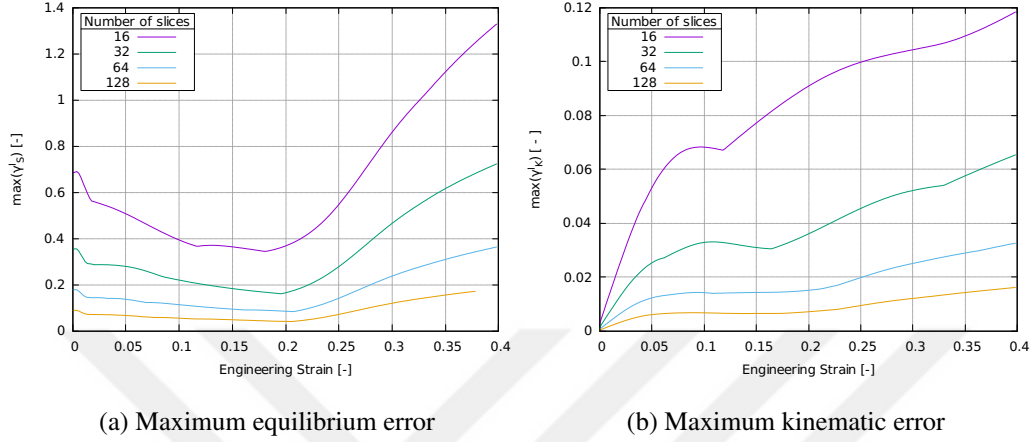


Figure 3.16: Maximum errors of kinematically compatible and statically equilibrated models

It should be noted that the physical interface conditions, i.e. kinematical compatibility or static equilibrium, are replaced by their energetically motivated counterparts that are proposed in Sections 3.3 and 3.4, since the constant strain field assumption in each region does not have the required number of degrees of freedom. Notice that increasing the number of regions by one also increases the number of interfaces by one. Nine components of the deformation gradient of the new region are the introduced additional degrees of freedom of the model. Static equilibrium and kinematical compatibility on the new interface, on the other hand, also requires nine scalar conditions according to (3.1) and (3.2). Therefore, even in the limit, the model does not have the necessary kinematic freedom and therefore the errors in Figure 3.17 do not decrease further. On the other hand, the errors approach to zero on the individual interfaces, since the $\mathbf{T}^I$, $\mathbf{S}^I$, $\mathbf{N}^I$ vectors of the adjacent interfaces approaches to each other. Since there exists a residual error in both the kinematically compatible and statically equilibrated models, as the number of regions increases their response does not necessarily approach to the same curve. In fact in Section 3.6.1 response of the models converges to different curves as presented in Figure 3.18.

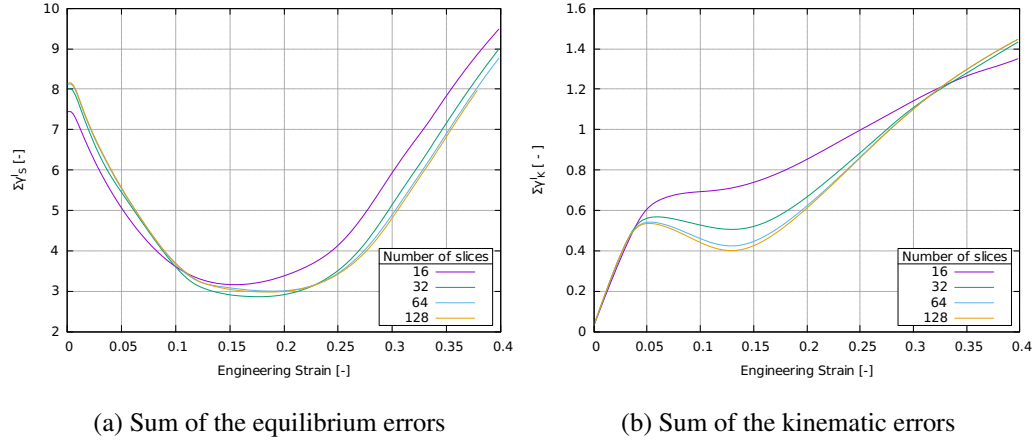


Figure 3.17: Total errors of the statically equilibrated and kinematically compatible models

3.6.3 Comparison with the Sequential Laminate-based Preliminary Model

For the homogenization methods for polycrystals, it is known that more stiff responses are obtained with more strict kinematical constraints. Responses of the Taylor approach and the sequential laminate based approach of Section 3.1.2 are compared with the models of this section in Figure 3.18. In sequential laminate based approach, kinematical compatibility is exactly satisfied on half of the interfaces of the model, specifically the even numbered interfaces in Table 3.1. In the kinematically compatible model, the kinematical compatibility is satisfied on all interfaces. Therefore, the kinematically compatible model should be closer to the Taylor approach than the sequential laminate. Similarly, sequential laminate should be closer to the Taylor bound than the statically equilibrated model, since the kinematical compatibility is relaxed on all interfaces of the statically equilibrated model. These expectations, which are expected only for the elastic range since the models employ more general constitutive models than rigid-plasticity, are satisfied according to the responses in Figure 3.18.

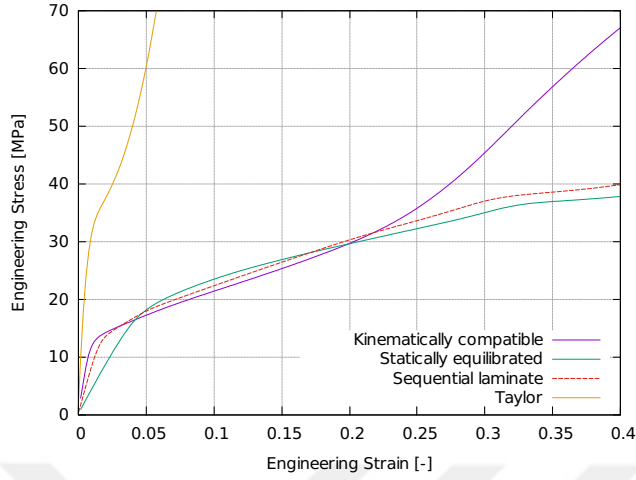


Figure 3.18: Responses of the models with bonded end interfaces in simple tension

3.6.4 Isotropy of the Models with Bonded End Interfaces

In Figure 3.19 the variation of the response with the loading direction in simple tension is presented, where the loading direction remains on the plane of the disk and is aligned with an interface of the model. As explained in Sections 3.3.2 and 3.4.2, the same equations are satisfied on all interfaces when the bonded formulations are employed. In Figure 3.19 it is verified that the response of the bonded models does not depend on n , therefore a single curve is presented. On the other hand, different responses are obtained with the models with free end interfaces, for different angles between the loading direction and the free end interfaces. Nevertheless responses of the models with bonded and free ends are similar up to 20% engineering strain.

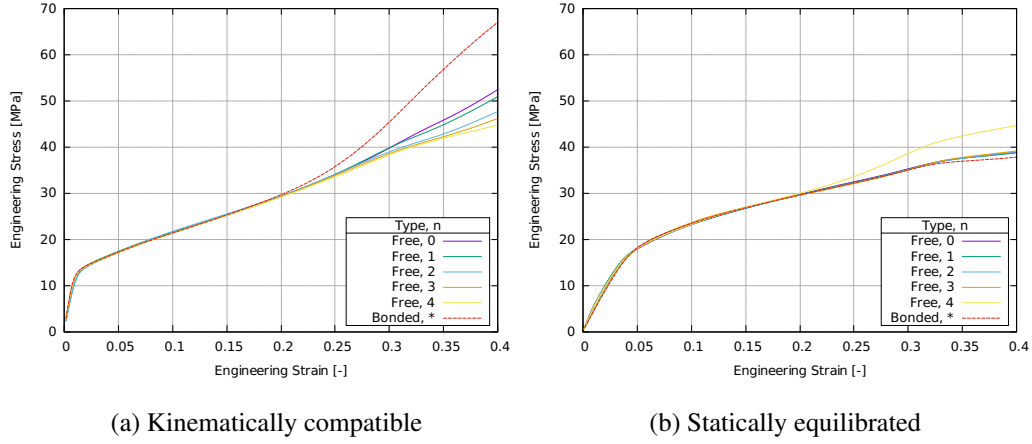


Figure 3.19: Variation of response with the loading direction of disk-like models with 32 slices. Angle between the loading direction and the last interface is $\frac{\pi}{8}n$.

3.6.5 Responses of the Combined Formulations

Variation of the weighted combination of the models with free end interfaces is presented in Figure 3.20. Response of the weighted hybrid model is similar to the kinematically compatible model for $\alpha = 0.9$, and similar to the statically equilibrated model for $\alpha = 0.1$.

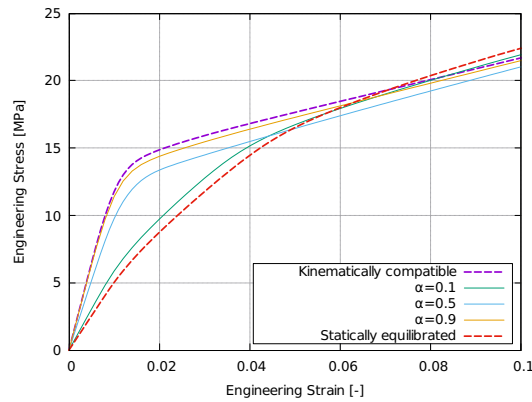


Figure 3.20: Variation of the simple tension response with the weighting factor α defined in Section 3.5.1

Response of the overly equilibrated model of Section 3.5.2 is presented in Figure 3.21

for bonded end interfaces and 32 slices. Responses of the kinematically compatible and statically equilibrated models with bonded end interfaces and the sequential laminate approach are also in Figure 3.21 for comparison. Overly equilibrated model provides the softest response. The interface normals $\mathbf{N}^I$ span the $x - y$ plane in the modeled disk-like spherulite geometry in Figure 3.11. In the overly equilibrated model, the components of the first Piola-Kirchhoff tensor in the $x - y$ plane, P_{11} , P_{22} , P_{12} and P_{21} are uniform throughout the model. Since both kinematically compatible and statically equilibrated model satisfy the kinematical compatibility for a special vector $\mathbf{S}$, that is along z direction in this case, F_{13} , F_{23} and F_{33} components are the same in all slices of the model. Therefore, the model is a Taylor-Sachs hybrid, that employs Taylor approach in the subspace with the projection tensor $\mathbb{P} = \mathbf{e}_3 \otimes \mathbf{e}_i \otimes \mathbf{e}_i \otimes \mathbf{e}_3$ and Sachs approach in the other subspace with projection tensor $\mathbb{P} = \mathbb{I} - \mathbf{e}_3 \otimes \mathbf{e}_i \otimes \mathbf{e}_i \otimes \mathbf{e}_3$, where Einstein's summation convention is implied. These projection tensors are identical to that are given for rank-1 laminates by Van Dommelen et al. [109], when their interface normal is $\mathbf{n} = \mathbf{e}_3$. Note that we define the double contraction operation as $\mathbf{A} : \mathbf{B} = A_{ij}B_{ij}$, while Van Dommelen [107] employs the convention $\mathbf{A} : \mathbf{B} = A_{ij}B_{ji}$. They expressed the Taylor-Sachs hybrid approaches in terms of these projection tensors, where each approach introduces a symmetric second order tensor as an additional unknown in a self-consistent-like scheme. Considering the hybrid models of Van Dommelen et al. [109] or Lee et al. [61], employing rank-1 laminates in the regions of the overly equilibrated model could also be a realistic approach. To sum up, the presented general formulation in Section 3.2 has a wide response-range comparable to the wide range the sequential laminates are known to provide [74]. However, the stress and strain fields are not necessarily compatible in the formulation, unlike sequential laminates.

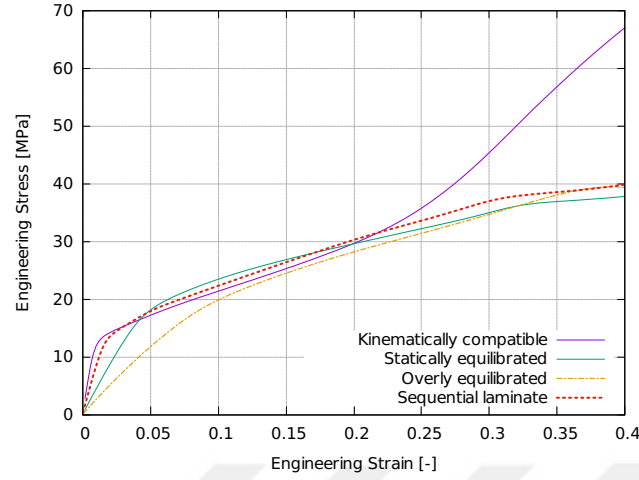


Figure 3.21: Uniaxial tension responses of different bonded formulations and the sequential laminate

3.6.6 Influence of the Degree of Crystallinity

In the model, the crystallinity ratio is taken into account by employing different central angles in the amorphous and crystalline regions as shown in Figure 3.22. In Figure 3.24, the initial Young's modulus and the yield strength of the curves in Figure 3.23 are compared with the experimental results of Crist et al. [25]. The experimental results generally lie between the results of the kinematically compatible and the statically equilibrated models in Figure 3.24.

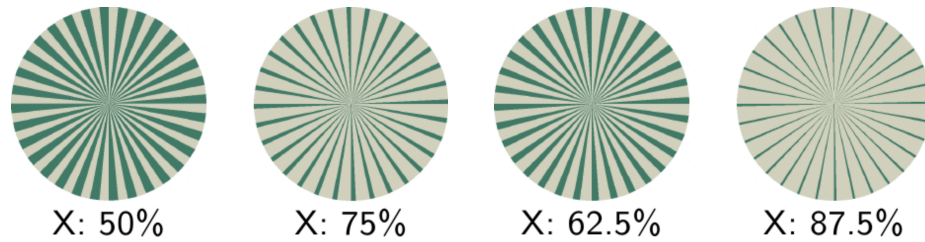


Figure 3.22: Views of the model for different crystallinity ratios. Dark and light colored regions are amorphous and crystalline, respectively.

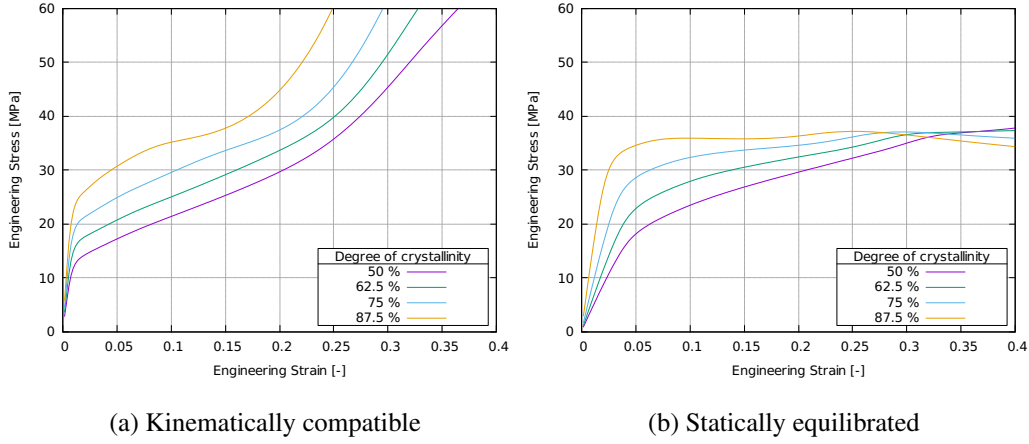


Figure 3.23: Variation of the responses of the models with bonded end interfaces with the degree of crystallinity in simple tension for 32 slices

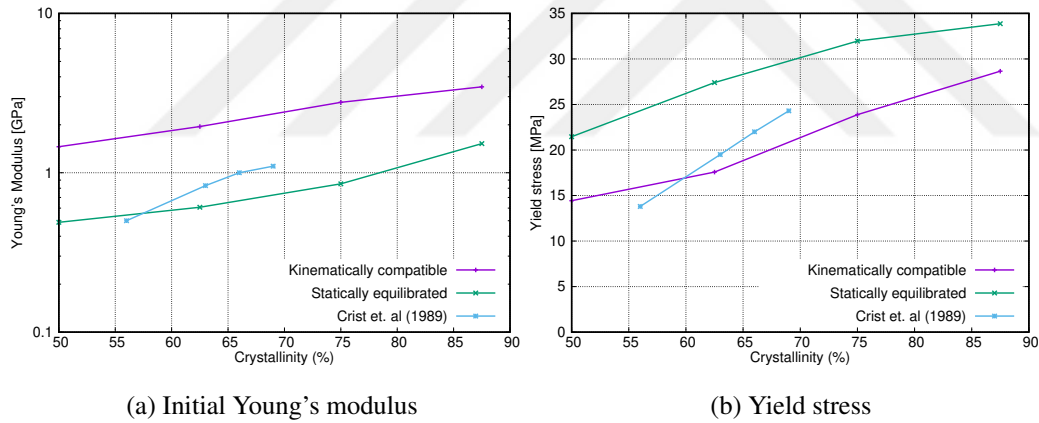


Figure 3.24: Comparison of the Young's modulus and the yield stress obtained from the disk like spherulite models and the experimental study of Crist et al. [25] on bulk specimens with spherulitic microstructure

3.6.7 Comparison with FEM

In literature, FEM with boundary conditions that satisfy the Hill's energy criterion is also employed to obtain the homogenized response of a volume element. Notice that, when a boundary condition is selected, such as linear displacement, as expressed in Section 1.3.1 the unknown of the model is the value of the macroscopic strain (or

stress) that will be enforced on the model, and the output of the model is the volume average of the strain (or stress) field. Moreover, the neighborhood information is also present in the FEM models, and the stress and strain fields satisfy equilibrium and compatibility. Thus, FEM provides full-field solutions, that are considered among the most realistic homogenization results. Therefore, approximations of the other homogenization methods are compared with the full-field simulations in the literature [71, 58, 59].

In order for the response of the disk-like spherulite FEM model presented in Section 2.3 to be comparable with that of the models in this chapter, boundary conditions that satisfy Hill's energy criterion should be employed in the disk-like FEM model. Therefore, the boundary conditions assigned to the internal nodes of the model in Section 2.3.1 are removed except the fixed nucleus. Periodic boundary conditions are assigned to the sides of the FEM model. Two versions of the boundary conditions are studied, that differ on the boundary conditions on the faces of the model with normals aligned with the z axis. In the first and second versions, traction free boundary conditions and periodic boundary conditions are employed, respectively. Therefore, two FEM analyses per deformation mode is presented in Figure 3.25, and the one that reaches the highest deformation level is compared with the statically equilibrated and kinematically compatible formulations with bonded end interfaces. Responses are in general agreement.

As discussed in 1.3.1, the importance of the selected boundary condition diminishes for statistically representative RVEs. However, the disk-like spherulite model is not a statistically representative RVE by itself, as expressed in Section 2.3.4.1. Moreover, periodic boundary conditions are not realistic, nevertheless, the response of the model does not strongly depend on the boundary conditions.

In Figure 3.27, in simple shear, the normal component of the stress on the shear plane is presented for the studied formulations and the FEM models. According to the coordinate system of Section 2, in order to be consistent with the results of Section 2.3.4.4 especially on the slip system activity, shear is applied along the x direction on the plane with the normal along y axis. Therefore, the normal component of stress on the shear plane is $\bar{P}_{22}$ component of the average first Piola-Kirchhoff stress tensor. In Fig-

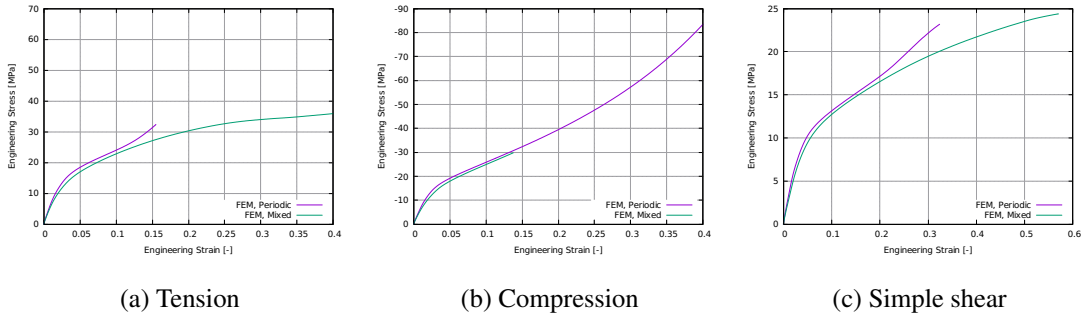


Figure 3.25: Influence of boundary conditions on the FEM analyses

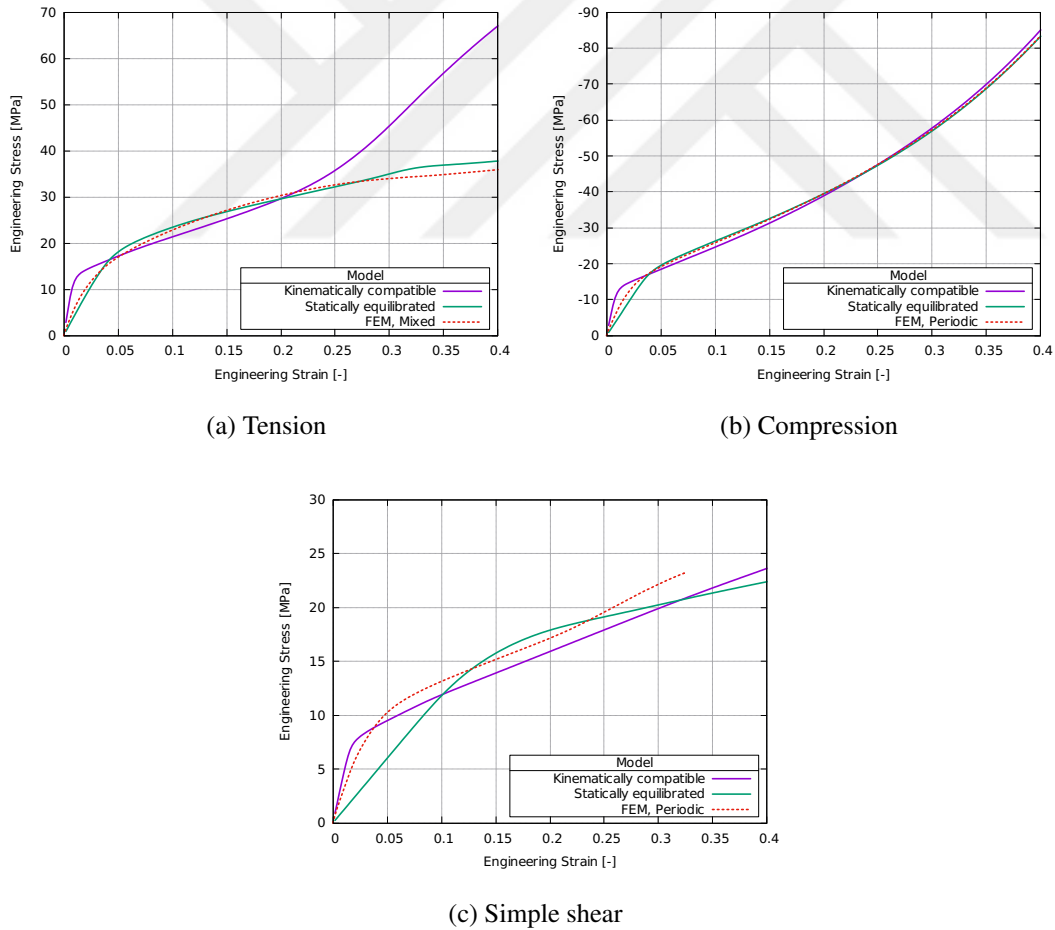


Figure 3.26: Comparison of the response of the models with bonded end interfaces and the FEM model

ure 3.27, the statically equilibrated formulation is in better agreement with the FEM models up to approximately 20% engineering strain, beyond which FEM models also differ among each other. In Figure 3.28, deformed views of the models are presented. In Figure 3.29 and 3.30, spatial variation of the slip system activities are presented. For the simple tension, slip system activities are in very good agreement with FEM results presented in Figure 2.16. The agreement with FEM results for the simple compression presented in Figure 2.18 is also quite good. However, while the slip system activities in FEM models are very similar in tension and compression as presented in Figures 2.16 and 2.18, in Figure 3.29 an unexpected $\{110\} \langle 1\bar{1}0 \rangle$ activities is observed on lamellae perpendicular to the loading axis in compression. According to the deformed views under compression in Figure 3.28, the crystalline slices perpendicular to the compression axis extend at least as much as the amorphous slices, and we have expressed in Section 2.4.3.1, the slip system $\{110\} \langle 1\bar{1}0 \rangle$ facilitates the extension or contraction of the lamellae. On the contrary, deformed views of the FEM model in Figure 2.11 indicate that the crystalline slices extend less than the amorphous slices. These differences could be attributed to the uniform strain assumption and the either exactly or approximately satisfied rank-1 connection between the slices. As for the simple shear, the slip system activities are different than that of presented in Figure 2.19. Nevertheless, slip activity of kinematically compatible formulation is also in good agreement with the experimental study of Bartczak et al. [11] discussed in Section 2.3.4.4, since the experimental study is on bulk specimens and distribution of slip system activities within a spherulite is not discussed. Statically equilibrated formulation finds $(100)[010]$ as the least active slip system, which is not consistent with Bartczak et al. [11], that reports it as the second most active slip system. To sum up, slip system activities and the stress-strain response of the kinematically compatible formulation are in better agreement with the FEM model, compared to the statically equilibrated formulation. In FEM analyses, the stress and strain fields are variable along the radius of the spherulite, which could explain the difference of the slips system activities in simple shear between the FEM models and the formulations presented in this section. It could be mentioned that, while we employ the schematic view of a disk in this chapter, another schematic representation that is also equally valid is a circular thin ring which is a disk of arbitrary radius the centroid of which is aligned with the centroid of a large disk-shaped void.

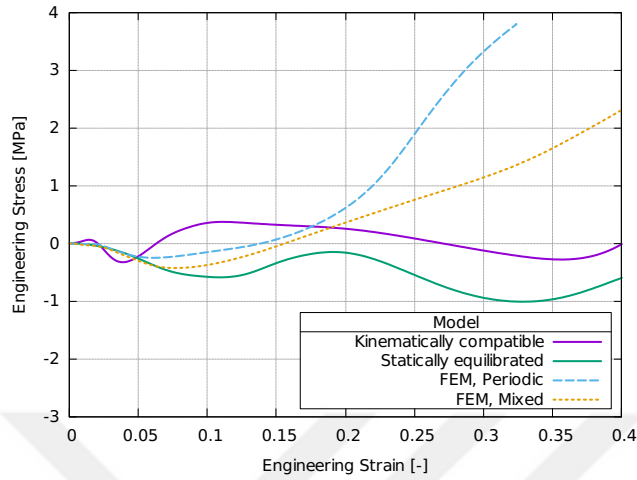


Figure 3.27: Comparison of the normal component of stress on the shear plane in simple shear analyses of the models

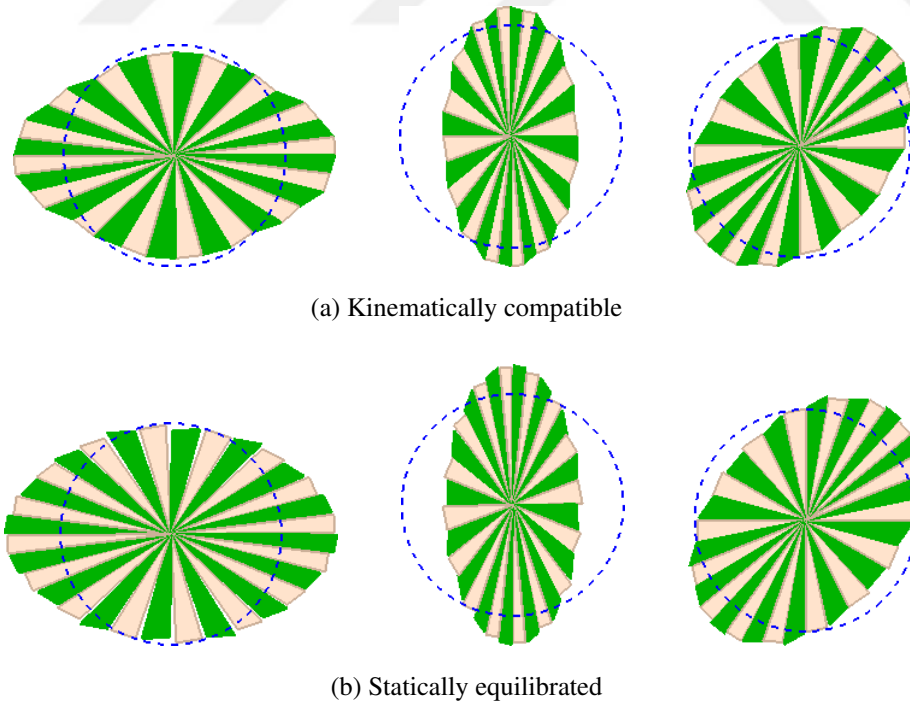


Figure 3.28: Deformed views of the models at 40% engineering strain under simple tension, compression and shear, respectively

As mentioned in Section 1.3.1, volume is also a quantity that is considered in the volume-averaging framework. In the small strain framework, when (1.16) is satisfied, volume change computed according to the microscopic strain field and the macroscopic strain $\bar{\epsilon}$, is the same, i.e. $\sum_{i=0}^N \nu^i \text{tr}(\epsilon^i) = \text{tr}(\bar{\epsilon})$, since trace is a linear operator, where we employed the notation of Section 1.3.2. Therefore, for small strain kinematics, the volume is trivially conserved in the transition between the length scales. However, this is not the case for the large-strain framework, since the determinant operator employed in the corresponding condition $\sum_{i=0}^N \nu^i \det(\mathbf{F}^i) = \det(\bar{\mathbf{F}})$ is non-linear. Van Dommelen et al. [109], satisfy this criterion in the expense of relaxing both (1.3) and (1.7), while we neglect the equality of volume between the length scales and exactly satisfy (1.3) and (1.7). It is known that only for large RVEs the microscopic and macroscopic volumes are equal for boundary value problems that employ the uniform traction boundary conditions (1.2), while the linear displacement or the periodic boundary conditions always satisfy the equality [102]. In Figure 3.31, in simple tension, the variation of the model volume with the engineering strain are presented according to the microscopical and macroscopic deformation gradients, respectively for the FEM models and our models. In the figure, current model volume is normalized to the reference volume. As expected, the microscopic and macroscopic volumes are not equal for the FE model that employ a traction-free boundary condition, where it should be noted that the microscopic volume corresponds to the actual volume of the model that could also be calculated according to the model boundary. Deviation between the macroscopic and microscopic volumes in our simplified disk-like spherulite models is in the same range with that of the FE model, and is negligible in a small deformation range, as expected. It should be mentioned that, the equality between the microscopic and macroscopic volumes is more significant when the conservation of mass in the transition between the length scales is particularly important [102]. In order to emphasize the comparability of the responses of FE analyses with the responses of our formulations, the evolutions of the external work, i.e. $\int \bar{\mathbf{P}} : \dot{\bar{\mathbf{F}}} dt$ are presented in Figure 3.32. Recall that, since special boundary conditions are employed in the FE analyses, Hill's energy criterion (1.9) is satisfied in FE models, as in our models.

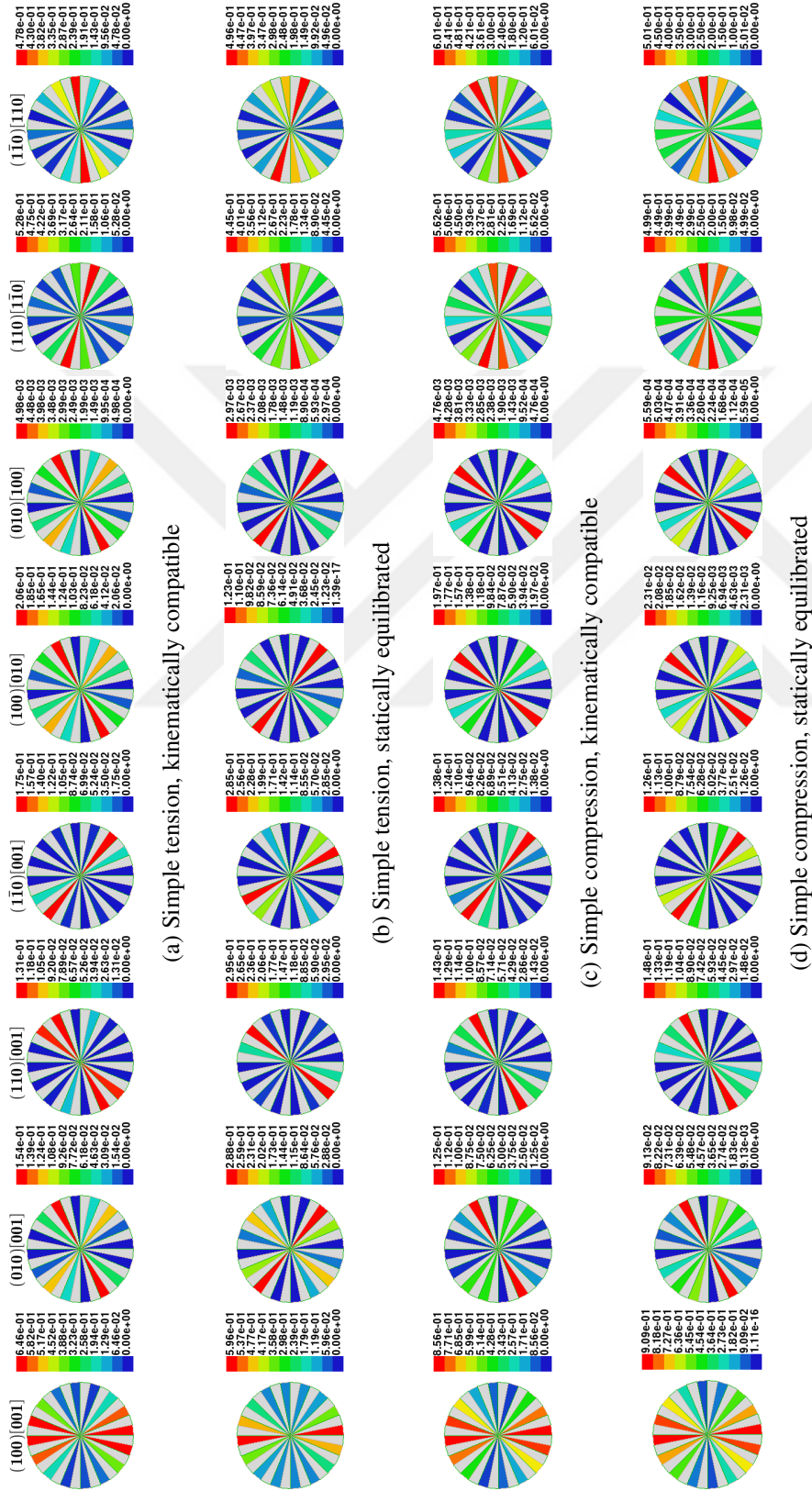
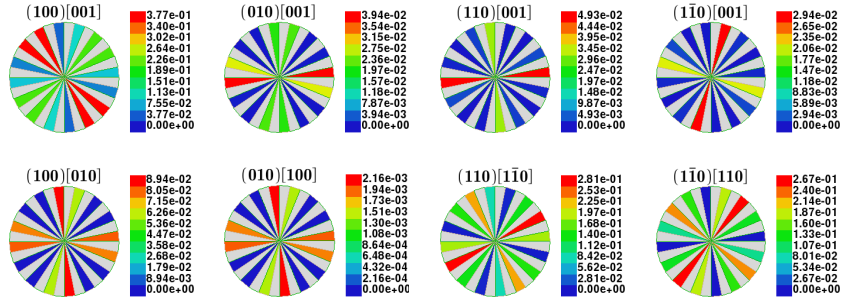
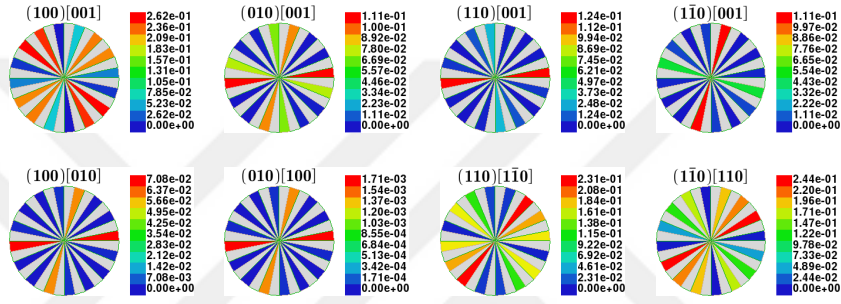


Figure 3.29: Distribution of the accumulated slip at 40% engineering strain for the simple tension and compression and studied formulations

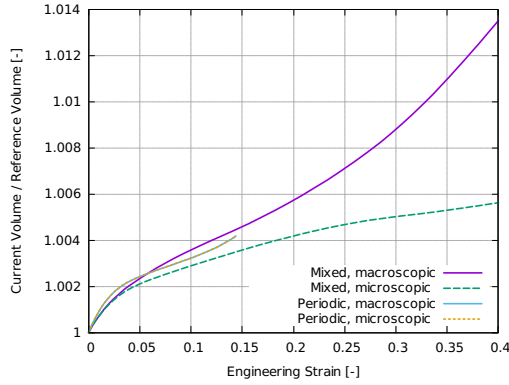


(a) Kinematically compatible

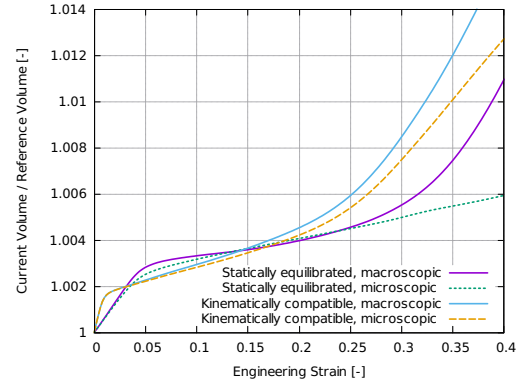


(b) Statically equilibrated

Figure 3.30: Distribution of the accumulated slip at 40% engineering strain for simple shear and studied formulations

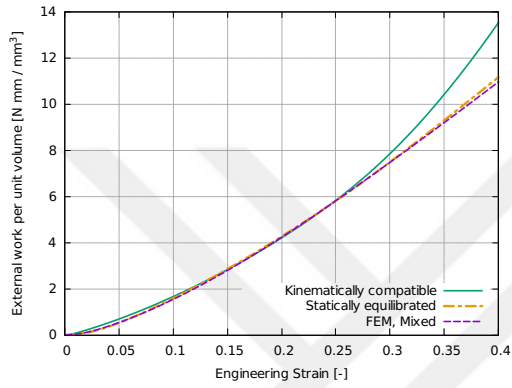


(a) Disk-like spherulite FEM models

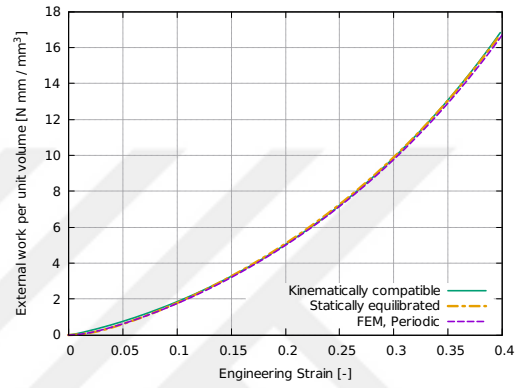


(b) Simplified disk-like spherulite models

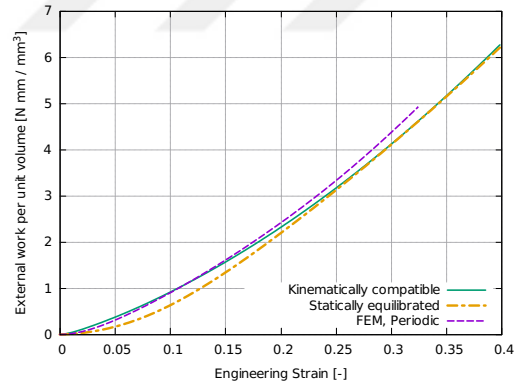
Figure 3.31: In simple tension, for FE models with different boundary conditions and our models, comparison of the volume evolution according to the microscopic and macroscopic deformation gradients



(a) Tension



(b) Compression



(c) Simple shear

Figure 3.32: Comparison of the external work of the models with bonded end interfaces and the FEM model

The simple tension analysis of the statically equilibrated formulation presented in Figure 3.26 is composed of 200 fixed-size load increments, and a single threaded Fortran implementation completes the analysis in 7.3 seconds on a 2.3 GHz Intel central processing unit (CPU). The evolution of the residuals of the driver iterations in the last increment is as follows: (6.37E+00, 4.23E-03, 1.29E-08) MPa. The evolution of the residuals of the interface condition iterations, respectively, in the first and last driver iterations of this increment are (3.79E+02, 2.96E+00, 9.13E-02, 1.02E-04, 1.05E-09) MPa and (9.30E-02, 1.93E-07) MPa. Compared to the first iteration, the last iteration starts with a lower residual since the enforced macroscopic deformation gradient is more similar to that is solved in the previous increment. The evolution of the residuals indicate quadratic convergence rates to the solutions. There are also the iterations of the constitutive model of the crystalline phase. In order to obtain faster solutions, the equations of the constitutive models of the phases could be introduced to the equation system of the interface conditions as in Van Dommelen [107]. The models are quite fast since an additional slice introduces only 3 scalar equations, i.e. (3.55) or (3.64) depending on the formulation, where the corresponding unknowns are the components of a vector that is similar to the jump vectors of the rank-1 laminates. Therefore, the solution speed is considered to be governed by the speed of the constitutive models of the phases. For comparison, the simple tension FEM analysis in Figure 3.26 reaches 40% engineering strain in 1.9 hours in 487 variable-size load increments with a single thread on the same CPU.

3.6.8 Modeling Ribbon-like Lamellae of Spherulites

In order to demonstrate that the proposed formulations could be employed in different geometries, a model for the twisted lamellar microstructure is presented in this section. The thin ribbon-like lamellae of spherulites, that are also schematically shown in Figure 2.1, grow from the nucleus often with a periodic twist. The lamellar twisting is considered as the reason of the concentric ring-shaped periodic extinction bands along the radius shown in Figure 3.33, where the distance between the rings corresponds to the half of the twist period [26]. Keller and Sawada [51] reports a twisting period of $2\text{ }\mu\text{m}$ for the lamellae in a bulk sample of polyethylene. Toda et al. [104] proposed a proportional relationship between the ring period and the width of the

lamellae, and experimentally measured these quantities in PE spherulites within thin film samples. According to the measurements of Toda et al. [104] we have considered $0.26 \mu\text{m}$ as a suitable width of a lamella with a twisting period of $2\mu\text{m}$. The helicoid with $2\mu\text{m}$ twisting period and $0.26 \mu\text{m}$ width is shown in Figure 3.34a by employing $-0.13 \leq u \leq 0.13$ and $P = 1/\pi$ in the parametric function of the helicoid which reads

$$h(u, \theta) = \langle u \cos(\theta), u \sin(\theta), P\theta \rangle \quad (3.69)$$

in Cartesian coordinates. Since the formulations presented in this chapter allow only for two neighbors per region, the actually helicoidal lamella is simplified and represented as a helix, that is also shown in Figure 3.34a. The parametric function of the helix reads

$$h(\theta) = \langle R \cos(\theta), R \sin(\theta), P\theta \rangle \quad (3.70)$$

where $R = 0.13/2$ is the radius and $P = 1/\pi$ is the pitch of the helix presented in Figure in 3.34a. Any value of R in the range $[-0.13, 0.13]$ would also lie in the helicoid shown in Figure 3.34a. The helix should be discretized into finite number of regions in order to be solved with the formulations presented in this chapter. The normal vectors of the interfaces between the regions are taken as tangent to the helix

$$\mathbf{N}(\theta) = \frac{dh(\theta)}{d\theta} = \langle -R \sin(\theta), R \cos(\theta), P \rangle, \quad \theta \in \left\{ 1 \frac{2\pi}{N}, 2 \frac{2\pi}{N} \dots, N \frac{2\pi}{N} \right\}.$$

This approach leads to the assumption of uniform strain along the width and thickness of the ribbon shaped lamellae, since the strain field is uniform in each region. A view of the planar interfaces between the regions of the helix discretized into 32 regions is presented in Figure 3.34b. While the geometry of a lamella consists of many periods of twists, it is sufficient to discretize only a single period of the helix where $0 < \theta \leq 2\pi$, since the periodic growth of the helix could be taken into account with the bonded formulations. A visualization of the discretized geometry is presented in Figure 3.34c. In Figure 3.34, the thickness of the lamella, which is typically between $10 - 40 \text{ nm}$, is presented as 50 nm for visual clarity. It should be remarked that the shape and the dimensions of the interfaces will not take part in the solution and the dimensions of these rectangular interfaces only help to distinguish the breadth from the thickness of the lamella in the figures.

While a lamella is a crystalline structure, it is also surrounded by an amorphous phase,

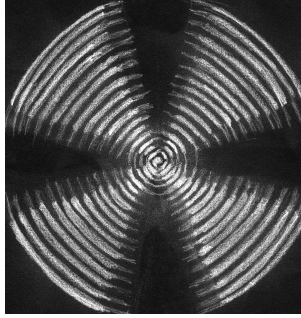


Figure 3.33: A sketch of a single spherulite in a PE thin film based on polarized light microscopy images of Hay and Keller [44]

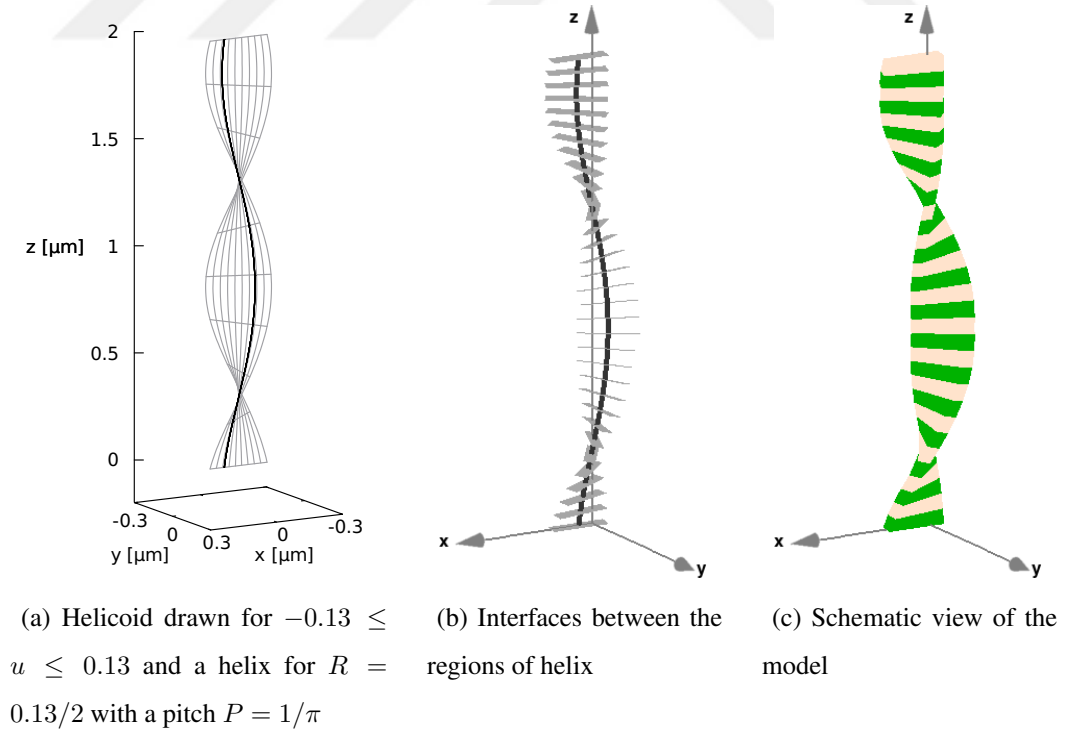


Figure 3.34: The discretized helical geometry of the model

and $\{201\}$ crystallographic plane is the interface between these phases. Therefore, a rank-1 laminate that employs the amorphous and the crystalline phase constitutive models and material parameters presented in Section 2.2 in its phases is employed as the constitutive model of each region. Crystallinity ratio of the laminate is taken as 50 %. The laminate of each region is oriented such that, the b lattice axis of the crystalline phase is aligned with the tangent vectors

$$\frac{dh(\theta)}{d\theta} = \langle -R\sin(\theta), R\cos(\theta), P \rangle \quad (3.71)$$

to the curve in (3.70) at the centroid of each region. Moreover, the interface normals between the amorphous and crystalline phases of the rank-1 laminates are aligned with the binormal vector $\frac{dh(\theta)}{d\theta} \times \frac{d^2h(\theta)}{d\theta^2}$ of the curve [1], which is perpendicular to the tangent vector in (3.71). The normal vector of the $\{201\}$ crystallographic plane, which is also aligned with the interface plane normal of the rank-1 laminate, is along the thickness direction of the helix based geometry presented in Figure 3.34c. Equal area projection pole figures of the crystallographic axes are presented in Figure 3.35. In Figure 3.35 it is seen that the b lattice axes are close to the z axis, which is the growth direction of the lamella as presented in Figure 3.34c. The circular symmetry of the poles in Figure 3.34c is a result of the regular twist of the helicoid and its discretization to equal sized regions. As also expressed by Rosenthal et al. [89], in the context of the lamellae of polymers, the main difference between the helicoidal and helical lamellae is that the radial growth direction is not physically contained in the helical lamellae, since a helix revolves around an empty cylinder. Therefore, while the b lattice direction is in the growth direction of a helicoidal lamellae, in a helix b lattice direction is tilted with respect to the growth direction of the lamellae. Idealization of the helicoid geometry with a helix could be an oversimplification, and perhaps may not be an appropriate idealization for the lamellae of spherulites in polyethylene. Nevertheless, there are semicrystalline polymers that are composed of helical lamellae. Rosenthal et al. [89] showed that the spherulites in thin films of polypropylene adipate are composed of helical lamellae and the b lattice direction is not along the radial direction in that case.

The statically equilibrated formulation with the bonded end interfaces, presented in Section 3.4.2 is employed for the solution of the discretized helix. The variation of the uniaxial response under simple tension and compression along the z axis with

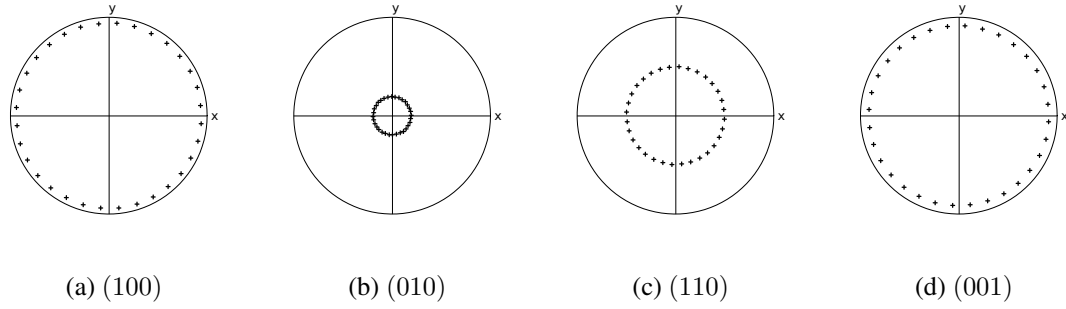


Figure 3.35: Initial crystallographic texture for the model with 32 slices

the number of regions N is presented in Figure 3.36. According to Figure 3.36, discretization of the helix into a relatively small number of regions is sufficiently accurate in both deformation modes, since an increase in the number of regions vary the orientations of the crystals only in a narrow range according to the well defined pattern presented in Figure 3.35. For the helix with 64 regions, another homogenized response of the constitutive models of the regions is obtained with the Taylor approach and also presented in Figure 3.35. In the compressive deformation mode, engineering stress-strain curves of the statically equilibrated formulation is much more softer than that of the Taylor approach, compared to the tensile response. The difference between the deformation modes is also clear in Figures 3.37 and 3.38, where the deformed views of the solutions for the Taylor approach and the statically equilibrated model with 32 regions are presented. The tensile and compressive responses are compared in Figure 3.39, where the variation of the absolute values of the Cauchy stress component along the loading direction with the true axial strain are plotted. Due to the crystallographic and morphological texture evolution phenomena which are accounted for respectively by the crystal plasticity constitutive model and the rank-1 laminate, the tensile and compressive responses are expected to differ especially at the later stages of the deformation even when the Taylor approach is employed. According to Figure 3.39 the statically equilibrated formulation, which satisfies static equilibrium and also approximately satisfy the kinematic compatibility of the helical morphology, provides a model with a high tension-compression asymmetry.

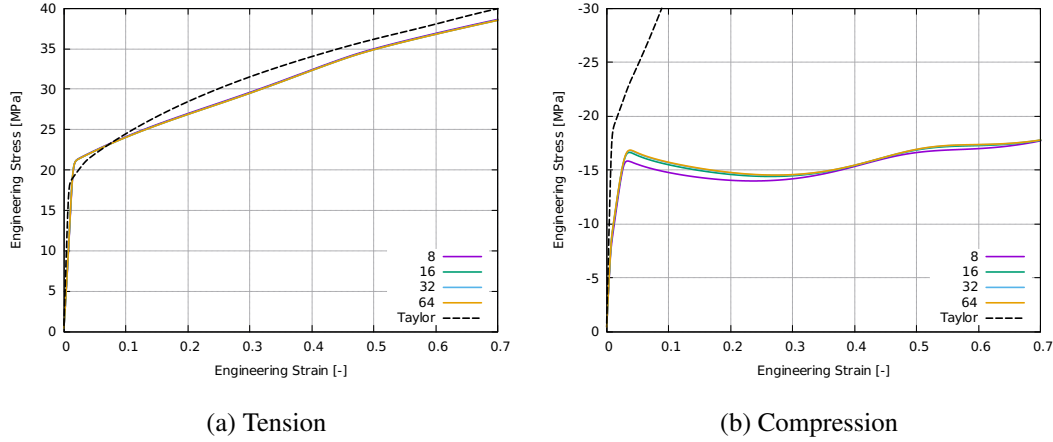


Figure 3.36: Variation of the response with number of regions of a single twisted lamellae under uniaxial loading along the growth axis

As far as we know, the present model is the only study in literature that discretize a twisted lamella and employ crystal plasticity. The actual lamellae of the semicrystalline polymers behave differently under axial tension and compression, since the lamellae that lie along the principal compression axis typically kink and take the shape of a chevron in transmission electron micrographs [12, 24, 14]. Although the mechanism behind the kinking is not completely understood, there is interest in the literature to propose models that takes this phenomenon into account. As a recent study, Lin et al. [65] consider that sinusoidal buckling instability occurs in semicrystalline polymers and introduces micro-buckling as a deformation mode to their model, which takes the basic building block of the semicrystalline polymers as a three phase structure that represents lamellar stacks embedded in an amorphous matrix. It should be mentioned that, kinking is also considered to be associated with the interlamellar separation deformation mode of the amorphous phase [13]. According to this view, since the increase of the distance between the lamellae could extend the tie chains up to their chain-length, kinking could be a result of the high stresses at the local bridges formed by the tie-molecules between the lamellae. The influence of the localized tie-chain bridges between the lamellae is not considered in the present model.

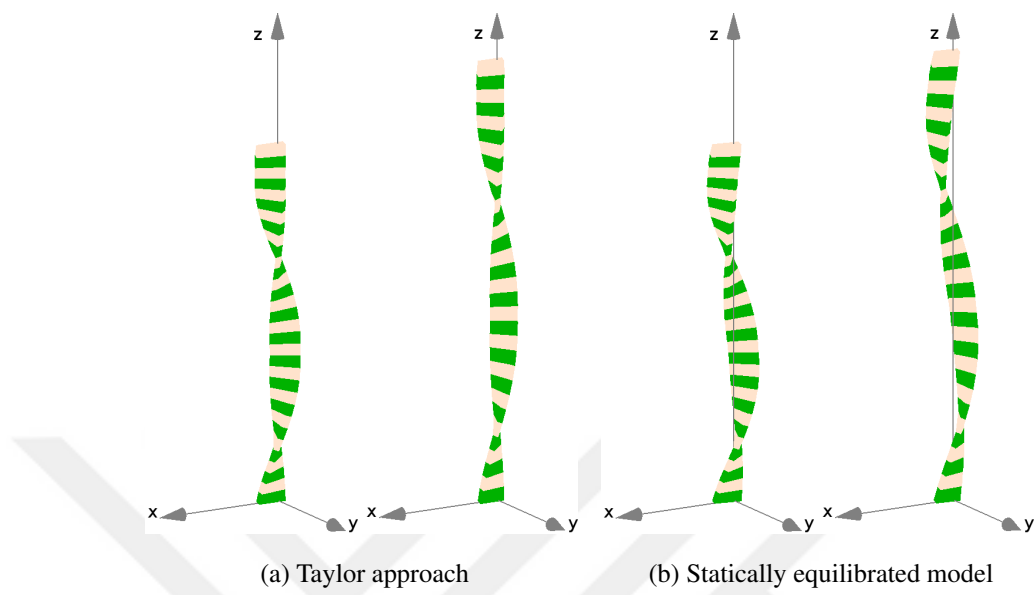


Figure 3.37: Deformed views at 35 and 70 % engineering strain under simple tension along z axis

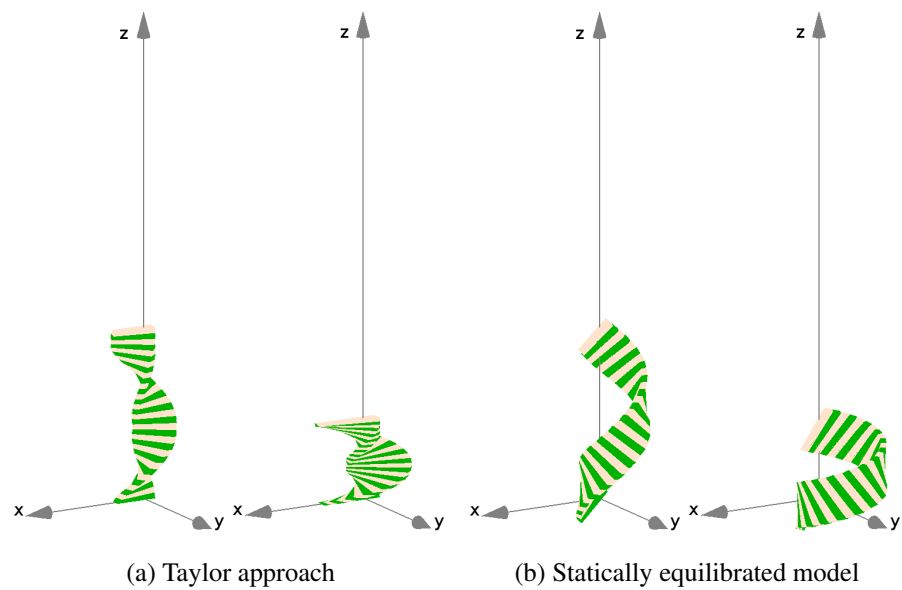
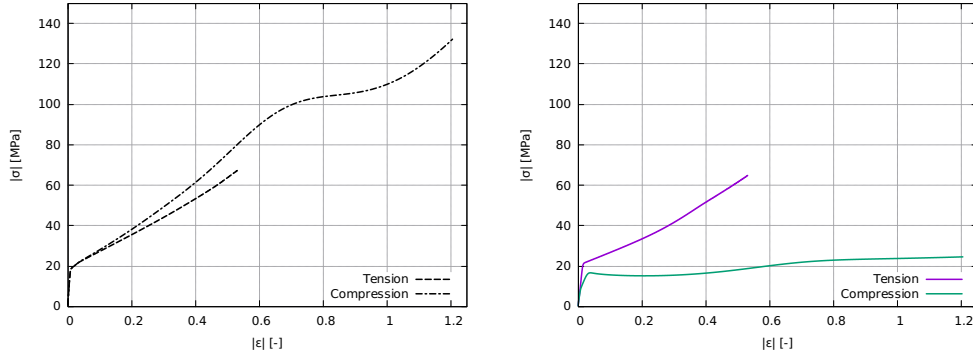


Figure 3.38: Deformed views at 35 and 70 % engineering strain under simple compression along z axis



(a) Taylor approach

(b) Statically equilibrated model with bonded end faces

Figure 3.39: True stress-strain curves for tension and compression. Notice that 70 % engineering strain corresponds to 0.53 and -1.2 true strain in tension and compression, respectively.

3.7 Conclusion

We propose methods and its several versions, that can be employed in homogenization based constitutive models for inelastic multiphase materials under large deformation. The method allows the phases to be inelastic and extends the lamination theory to accommodate more than one lamination direction. The method can be used with geometries discretized into regions separated by planar interfaces and each region can have at most two neighbors. Similar to the lamination theory, a uniform field assumption is employed in each region of the model. Different from the sequential laminates, all interfaces between regions are considered to lie at the same length scale, and there are versions of the model that provides statically admissible stress field or kinematically compatible strain field throughout the discretized geometry. However, in order to satisfy the Hill's energy criterion with the limited number of degrees of freedom, when the stress field is statically admissible the strain field is only weakly kinematically admissible and vice versa. Since the kinematical compatibility and static equilibrium is not satisfied together, the model could be considered as a polycrystal homogenization approach, such as Taylor and Sachs approaches, their hybrids [62] or the self-consistent scheme rather than a full-field homogenization approach

such as FEM when employed with appropriate boundary conditions. We do not see any reason that prevents the employment of the statically equilibrated and kinematically compatible formulations for regions with more than two interfaces. The procedures followed for finding the formulations with bonded end interfaces potentially could also be employed to take into account more than two interfaces per region. However, as discussed in Section 3.2, if too many interfaces and a limited number of regions are employed, kinematically compatible and statically equilibrated formulations could potentially reduce to the Taylor and Sachs approaches, respectively.

The method is similar to the grain-cluster methods such that the interaction of the adjacent regions is considered on planar interfaces with uniform fields in each region. In order to consider the influence of the neighborhood of a crystal in homogenization, the statistical information on the neighborhoods is required. Since the proposed method can be employed with discretized geometries, the required neighborhood information is obtained from the well characterized morphologies of the modeled material, for instance the spherulite morphology of PE. The necessary number of regions for the discretization is identified from the convergence criterion of the stress-strain response. A special emphasis given on the integrity of the modeled morphology may motivate the significance attributed to the internal interfaces over the model boundaries.

Arbitrary constitutive models could be employed in each region, and the relative size of each region is also arbitrary. Therefore, the influence of the degree of crystallinity is studied by employing different central angles for the amorphous and crystalline regions of the model. Response of the model, that is composed of only 32 regions with uniform fields, is comparable with the FEM model under the studied simple tension, compression and shear deformation modes. Additionally, twisted ribbon shaped lamellae of the PE is also modeled, where the bonded end interfaces in this case enforces infinite length of the lamellae growing with a continuous periodic twist.



CHAPTER 4

A MODEL WITH NON-UNIFORM STRAIN FIELD BETWEEN LAMINATES

In this chapter, a modeling approach based on a kinematically compatible strain field in the cylindrical coordinate system is presented for small strain kinematics. The strain field could be employed in a region between the interfaces of two rank-1 laminates, that are homogenized using the Taylor approach. In Section 4.1, the equations of a laminate are given in cylindrical coordinates at small strains. In Section 4.2, uniform strain fields of the phases of laminates homogenized with the Taylor-type homogenization are presented in terms of the formulation in Section 4.1. In Section 4.3, the definition of regions formed between interfaces of adjacent laminates and a model that employs a kinematically compatible strain field in these regions are presented. In the studied model, the orientations and the material models of the laminates are similar to that of the disk-like spherulite model in Section 2.3. Numerical results are presented in Section 4.4. A more general strain field that could support interaction laws other than Taylor-approach and conclusions are given in Sections 4.5 and 4.6, respectively.

4.1 A Representation of the Equations of the Rank-1 Laminates at Small Strains

The rank-1 laminate is presented in Section 3.1.1. In this section, the laminate is presented in cylindrical coordinates with the small strain assumption. For a given macroscopic deformation gradient, a vector that identifies the jump in the deformation gradient field is presented as the unknown of a laminate with a certain orientation in Section 3.1.1. In this section, the individual components of this jump vector are

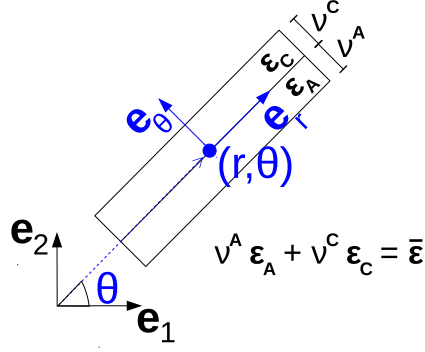


Figure 4.1: Schematic representation of the laminate, where the interface normal is the polar basis vector e_θ

specified in the cylindrical coordinate system, and are considered as the unknowns of the laminate. As a convention, the unit normal vector $\mathbf{n}$ of the interface is considered to be aligned with the e_θ basis vector of the cylindrical coordinate system

$$\mathbf{n} = e_\theta \quad (4.1)$$

as in Figure 4.1. The rank-1 laminate schematically presented in Figure 4.1 is composed of two half spaces with uniform stress and strain fields and shares a planar interface. In this section we will refer to these half-spaces, or regions, with the letters A and C , after the amorphous and crystalline phases of semicrystalline polymers, respectively.

The constant strain in the phases of the laminate can be decomposed into the macroscopic and the fluctuation parts

$$\epsilon^A = \bar{\epsilon} + \tilde{\epsilon}^A, \quad \epsilon^C = \bar{\epsilon} + \tilde{\epsilon}^C \quad (4.2)$$

where ϵ^A and ϵ^C are constant strain tensors in the amorphous and crystalline phases, respectively. $\bar{\epsilon}$ is the enforced macroscopic strain. The volume average of the strain of the phases is equal to the macroscopic strain

$$\nu^A \epsilon^A + \nu^C \epsilon^C - \bar{\epsilon} = \mathbf{0} \quad (4.3)$$

where $\nu^A + \nu^C = 1$ relationship exists between the volume fractions. Substitution of (4.2) into (4.3) yields

$$\nu^A \tilde{\epsilon}^A + \nu^C \tilde{\epsilon}^C = \mathbf{0}. \quad (4.4)$$

According to (4.4), without loosing generality, $\tilde{\epsilon}^A$ and $\tilde{\epsilon}^C$ could be written in terms of an arbitrary tensor $\tilde{\epsilon}$ as

$$\tilde{\epsilon}^A = \frac{1}{\nu^A} \tilde{\epsilon} \quad \text{and} \quad \tilde{\epsilon}^C = -\frac{1}{\nu^C} \tilde{\epsilon}. \quad (4.5)$$

By substituting (4.5) into (4.2) we obtain the representation of the uniform strain fields of the phases in terms of $\tilde{\epsilon}$

$$\epsilon^A = \bar{\epsilon} + \frac{1}{\nu^A} \tilde{\epsilon}, \quad \epsilon^C = \bar{\epsilon} - \frac{1}{\nu^C} \tilde{\epsilon}. \quad (4.6)$$

Notice that the representations of ϵ^A and ϵ^C in (4.6) trivially satisfy the kinematical condition in (4.3). Now we will specify $\tilde{\epsilon}$. Recall that, as stated in Section 3.1.1, only a rank-1 jump is allowed between the deformation gradients of the regions of a laminate, as a consequence of the kinematical compatibility along the interface. Therefore, $\tilde{\epsilon}$ should be the symmetric part of a rank-1 tensor

$$\tilde{\epsilon} = \text{sym}(\mathbf{m} \otimes \mathbf{n}) \quad (4.7)$$

where $\mathbf{n}$ is the unit normal vector of the interface and $\mathbf{m}$ is an arbitrary vector. Let us specify $\mathbf{m}$ in terms of components

$$\mathbf{m} = \alpha \mathbf{e}_\theta + \gamma \mathbf{e}_r + \beta \mathbf{e}_3, \quad (4.8)$$

where α , β and γ are arbitrary scalars. Then

$$\tilde{\epsilon} = \alpha \mathbf{e}_\theta \otimes \mathbf{e}_\theta + \text{sym}(\gamma \mathbf{e}_r \otimes \mathbf{e}_\theta + \beta \mathbf{e}_3 \otimes \mathbf{e}_\theta) \quad (4.9)$$

where (4.1) and (4.8) are substituted into (4.7). Since $\mathbf{e}_r$, $\mathbf{e}_\theta$ and $\mathbf{e}_3$ form an orthonormal basis and α , β and γ are arbitrary, $\mathbf{m}$ is still arbitrary. Note that when a single laminate is considered, selection of the $\mathbf{e}_\theta$ basis vector as the interface normal $\mathbf{n}$ in (4.1) is a convention rather than an assumption. By substituting (4.9) into (4.6), we obtain

$$\begin{aligned} \epsilon^A &= \bar{\epsilon} + \frac{\alpha}{\nu^A} \mathbf{e}_\theta \otimes \mathbf{e}_\theta + \text{sym} \left(\frac{\gamma}{\nu^A} \mathbf{e}_r \otimes \mathbf{e}_\theta + \frac{\beta}{\nu^A} \mathbf{e}_3 \otimes \mathbf{e}_\theta \right) \\ \epsilon^C &= \bar{\epsilon} - \frac{\alpha}{\nu^C} \mathbf{e}_\theta \otimes \mathbf{e}_\theta - \text{sym} \left(\frac{\gamma}{\nu^C} \mathbf{e}_r \otimes \mathbf{e}_\theta + \frac{\beta}{\nu^C} \mathbf{e}_3 \otimes \mathbf{e}_\theta \right). \end{aligned} \quad (4.10)$$

Then, for a given macroscopic strain $\bar{\epsilon}$, there are three scalar unknowns in the laminate. These unknowns are solved to satisfy the traction equilibrium condition

$$(\sigma^A - \sigma^C) e_\theta = 0 \quad (4.11)$$

along the interface. The unknowns of the laminate at small strains were originally the components of the symmetric tensors ϵ^A and ϵ^C . Definitions of these unknowns that trivially satisfy (4.3) and also the kinematical compatibility on the interface are provided in terms of the macroscopic strain and the components of the jump vector. Therefore, the number of unknowns is reduced from twelve to three without losing generality. The stress response is obtained from the volume average of the stress fields of the phases

$$\bar{\sigma} = \nu^A \sigma^A + \nu^C \sigma^C.$$

4.2 Homogenization of the Laminates in Cylindrical Coordinates Using the Taylor Approach

Let us consider an arbitrary number, say N^I , of rank-1 laminates oriented in space as in Figure 4.2 are homogenized using the Taylor approach. Then, the strain tensor of each laminate ϵ^I is equal to the macroscopic strain tensor $\bar{\epsilon}$ and macroscopic stress $\bar{\sigma}$ is the volume average of the stress tensors of the laminates $\sigma^I = \nu^A \sigma_A^I + \nu^C \sigma_C^I$

$$\epsilon^I = \bar{\epsilon}, \quad \forall I \in \{1, \dots, N^I\} \quad \text{and} \quad \bar{\sigma} = \frac{1}{N^I} \sum_{I=1}^{N^I} \sigma^I \quad (4.12)$$

where σ_A^I and σ_C^I are the stress tensors of the amorphous and crystalline phases of laminate I and we assume that the values of the volume fractions ν^A and ν^C are the same in all laminates and the volumes of the laminates are equal. According to (4.12), (4.10) could be written for each laminate as

$$\begin{aligned} \epsilon_A^I &= \bar{\epsilon} + \frac{\alpha^I}{\nu^A} e_\theta \otimes e_\theta + \text{sym} \left(\frac{\gamma^I}{\nu^A} e_r \otimes e_\theta + \frac{\beta^I}{\nu^A} e_3 \otimes e_\theta \right) \\ \epsilon_C^I &= \bar{\epsilon} - \frac{\alpha^I}{\nu^C} e_\theta \otimes e_\theta - \text{sym} \left(\frac{\gamma^I}{\nu^C} e_r \otimes e_\theta + \frac{\beta^I}{\nu^C} e_3 \otimes e_\theta \right) \quad \forall I \in \{1, \dots, N^I\}. \end{aligned} \quad (4.13)$$

According to (4.13), for a given macroscopic strain tensor $\bar{\epsilon}$, the only unknowns are α^I , γ^I and β^I . Therefore, there are $3N^I$ unknowns in the model. These unknowns

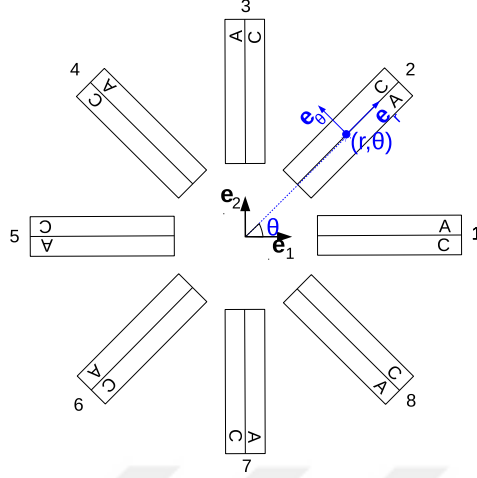


Figure 4.2: Schematic view of the radially oriented laminates spanning a complete circle. The interface normals are along e_θ

are solved to satisfy the traction equilibrium on the interfaces

$$(\sigma_A^I - \sigma_C^I) e_\theta = \mathbf{0}, \quad \forall I \in \{1, \dots, N^I\}$$

which provides $3N^I$ equations.

Notice that, although the same strain is employed in all laminates with the Taylor approach, the distribution of the strain in the amorphous and crystalline regions is not necessarily uniform. In other words, although the relationship

$$\nu^A \epsilon_A^I + \nu^C \epsilon_C^I = \nu^A \epsilon_A^J + \nu^C \epsilon_C^J \quad \forall I, J \in \{1, \dots, N^I\} \quad (4.14)$$

exists between the strain fields of the arbitrary laminates I and J , the strain fields in the amorphous and crystalline regions of arbitrary laminates are not necessarily identical

$$\epsilon_A^I \neq \epsilon_A^J, \quad \epsilon_C^I \neq \epsilon_C^J,$$

and in fact would vary considerably among the laminates, in particular, when the constitutive models employed in the laminates are strongly anisotropic.

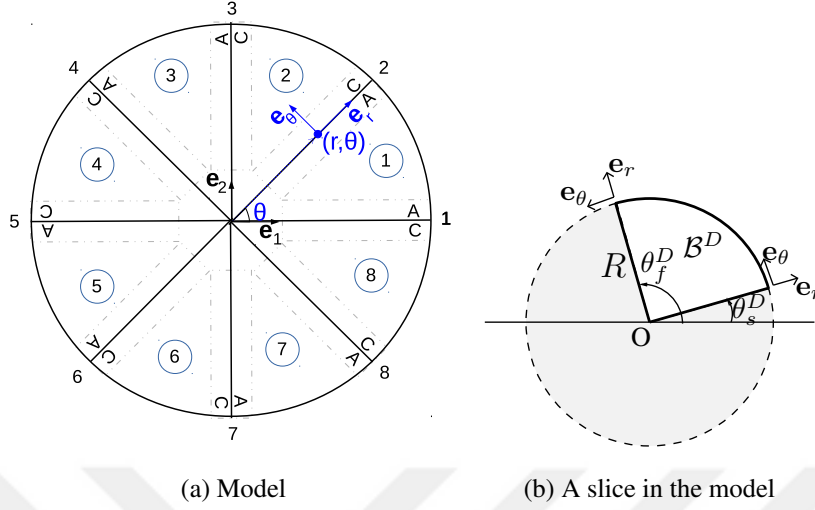


Figure 4.3: Schematic view of the complete model and a slice of the model

4.3 Proposed Model

In Figure 4.3, we join the interfaces of the laminates presented in Figure 4.2 at the origin, and set the outer boundary as a circle with radius R . Each region thus formed between the interfaces of the adjacent laminates is called a slice. There are eight slices in Figure 4.3, in addition to the eight interfaces that are also present in Figure 4.2. The model supports arbitrary number N^S of slices. In Figure 4.3 odd and even numbered slices are considered as amorphous and crystalline, respectively.

In Section 4.2, (4.13) is written for the interface with number I . In this section, we will propose a non-uniform, compatible strain field in the slices presented in Figure 4.3. To this end, first, we need to re-write (4.13) for a slice D . While (4.13) is for the two sides of an interface I ,

$$\begin{aligned}
 \epsilon^D(\theta_s^D) &= \bar{\epsilon} + \frac{\alpha_s^D}{\nu^D} \mathbf{e}_\theta \otimes \mathbf{e}_\theta + \text{sym} \left(\frac{\gamma_s^D}{\nu^D} \mathbf{e}_r \otimes \mathbf{e}_\theta + \frac{\beta_s^D}{\nu^D} \mathbf{e}_3 \otimes \mathbf{e}_\theta \right) \\
 \epsilon^D(\theta_f^D) &= \bar{\epsilon} + \frac{\alpha_f^D}{\nu^D} \mathbf{e}_\theta \otimes \mathbf{e}_\theta + \text{sym} \left(\frac{\gamma_f^D}{\nu^D} \mathbf{e}_r \otimes \mathbf{e}_\theta + \frac{\beta_f^D}{\nu^D} \mathbf{e}_3 \otimes \mathbf{e}_\theta \right)
 \end{aligned} \tag{4.15}$$

is for the inner sides of the start and end interfaces of a slice D , which are at $\theta = \theta_s^D$ and $\theta = \theta_f^D$ polar coordinates as shown in Figure 4.3b. The subscripts s and f denote

the start and finish of a slice. For convenience, the number of unknowns are increased in (4.15) compared to (4.13), with the following additional relationships

$$\begin{aligned}\alpha_f^I + \alpha_s^J &= 0 \\ \gamma_f^I + \gamma_s^J &= 0 \\ \beta_f^I + \beta_s^J &= 0; \forall I \in \{1, 2, \dots, N^S\}\end{aligned}\tag{4.16}$$

$$\text{where } J = \begin{cases} 1 & I = N^S \\ I + 1 & \text{otherwise} \end{cases}.$$

The strain tensors in (4.15) are defined at $2N^I$ locations and are identical to (4.13), when employed with (4.16) and ν^D is either equal to ν^A or ν^C , depending on which phase the slice belongs to. Together (4.15) and (4.16) merely provide a representation for the strain fields of the rank-1 laminates in Figure 4.2 homogenized with Taylor approach. The employed notation is based on the cylindrical coordinate system also presented in the figure. Now we will extend (4.15), by providing a definition for any θ between θ_s^D and θ_f^D :

$$\begin{aligned}\boldsymbol{\varepsilon}^D(r, \theta) &= \bar{\boldsymbol{\varepsilon}} + \frac{N_s^D(\theta)}{\nu^D} \left[\alpha_s^D \mathbf{e}_\theta \otimes \mathbf{e}_\theta + \frac{1}{r} \text{sym}(\gamma_s^D \mathbf{e}_r \otimes \mathbf{e}_\theta + \beta_s^D \mathbf{e}_3 \otimes \mathbf{e}_\theta) \right] \\ &+ \frac{N_f^D(\theta)}{\nu^D} \left[\alpha_f^D \mathbf{e}_\theta \otimes \mathbf{e}_\theta + \frac{1}{r} \text{sym}(\gamma_f^D \mathbf{e}_r \otimes \mathbf{e}_\theta + \beta_f^D \mathbf{e}_3 \otimes \mathbf{e}_\theta) \right]\end{aligned}\tag{4.17}$$

where $N_s^D(\theta) = \frac{\theta - \theta_f^D}{\theta_s^D - \theta_f^D}$ and $N_f^D(\theta) = \frac{\theta_s^D - \theta}{\theta_s^D - \theta_f^D}$ are interpolation functions, that are linear in terms of the polar coordinate θ . Additionally the definition of ν^D is changed to represent the volume fraction of each slice D , $\nu^D = \frac{\theta_f^D - \theta_s^D}{2\pi}$. Interpolation functions $N_s^D(\theta)$ and $N_f^D(\theta)$ have the following properties

$$\begin{aligned}N_s^D(\theta) + N_f^D(\theta) &= 1, \text{ for } \theta_s^D \leq \theta \leq \theta_f^D \\ N_s^D(\theta_s) &= N_f^D(\theta_f) = 1 \\ N_s^D(\theta_f) &= N_f^D(\theta_s) = 0\end{aligned}\tag{4.18}$$

similar to the conventional one-dimensional shape functions employed in finite elements.

In Section 4.1, we have selected the fluctuation field in such a way that it trivially satisfies the equality between the macroscopic strain and the volume average of the

strain field in (4.3). This condition could be written for the complete model as

$$\frac{1}{\pi R^2} \sum_{D=1}^{N^S} \int_{\mathcal{B}^D} \epsilon^D dV - \bar{\epsilon} = \mathbf{0} \quad (4.19)$$

and is not exactly satisfied with (4.17), since the polar basis vectors e_r and e_θ rotate with the polar coordinate θ . In order to satisfy (4.19), the domain of each slice should be sufficiently small, such that the rotation of the polar basis vectors with the change of the polar coordinate θ is negligible within a slice. In other words, in order to satisfy (4.19) in an approximate manner, the number of the slices of the model should be high.

In Sections 4.1 and 4.2 the unknowns α^I , γ^I and β^I are solved to satisfy the traction equilibrium on the interface between the phases of a laminate, where the stress field is uniform in each phase. On the other hand, in (4.17) the strain field is a function of the polar coordinate r as well, which is introduced for the strain compatibility in a slice. Therefore, the stress and strain fields are not uniform on the interfaces between regions. Nevertheless, considering e_r and e_3 basis vectors span the interfaces of the laminates, and according to (4.17) non-zero $e_r \otimes e_r$, $e_r \otimes e_3$, $e_3 \otimes e_3$ components could only be present in the macroscopic strain $\bar{\epsilon}$ that is common in all slices, the kinematical compatibility is satisfied at all points on the interfaces. In order not to deviate much from the formulation presented in Sections 4.1 and 4.2, the traction equilibrium is enforced at a single point at $r = R/2$

$$\left[\sigma^I \left(\frac{R}{2}, \theta_f^I \right) - \sigma^J \left(\frac{R}{2}, \theta_s^J \right) \right] e_\theta(\theta_s^J) = \mathbf{0} \quad \forall I \in \{1, \dots, N^S\} \quad (4.20)$$

$$\text{where } J = \begin{cases} 1 & I = N^S \\ I + 1 & \text{otherwise} \end{cases}.$$

where $\theta_f^I = \theta_s^J$ according to Figure 4.3. The stress of the model is the volume average of the stress field

$$\bar{\sigma} = \frac{1}{\pi R^2} \sum_{D=1}^{N^S} \int_{\mathcal{B}^D} \sigma^D dA. \quad (4.21)$$

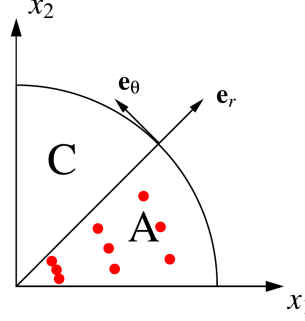


Figure 4.4: Gauss quadrature integration points of slice A

4.4 Results of the Model

Isotropic linear elasticity with bulk modulus $\kappa = 2000$ MPa and shear modulus $\mu = 35$ MPa is employed in the amorphous slices of the model. In the crystalline slices, crystal viscoplasticity with orthotropic elasticity, the algorithmic box of which is presented in Table 4.1, is employed. The material parameters presented in Section 2.2 are used in the crystalline slices. Simple tension deformation mode is studied. Orientation of the crystalline lattice is similar to that employed in Section 2.3. The same crystalline lattice orientation is employed in all integration points of a slice. Tension is applied along the x direction by increasing $\bar{\varepsilon}_{11}$ component of the macroscopic strain. Employed driver function requires normal stress components $\bar{\sigma}_{22}$ and $\bar{\sigma}_{33}$ to be to zero by varying $\bar{\varepsilon}_{22}$ and $\bar{\varepsilon}_{33}$ components of the macroscopic strain. The macroscopic stress is evaluated numerically via the Gauss quadrature with nine integration points in each slice as shown in Figure 4.4.

Variation of the model response with the number of slices for 50 % crystallinity ratio is presented in Figure 4.5. The model response converges as the number slices increases. Figure 4.6 presents the evolution of the error with respect to the satisfaction of (4.19) with strain. While the error is large in (4.19) for the model with eight slices, as the number of slices increases the error decreases as expected.

In Figure 4.7, distribution of the ε_{11} and ε_{rr} strain components in models with different number of slices are presented. Similar to the stress-strain curves, the strain distributions converge with increasing number of slices. According to (4.17) fluctuation is zero in the ε_{rr} strain component. Therefore, the spatial variation of this

Table 4.1: Small strain crystal plasticity with forward Euler integration of ϵ^p

1. Database $\{\epsilon_{n+1}, \Gamma_n^\alpha, \sigma_n\}$ given. Compute the plastic strain of the previous load increment

$$\epsilon_n^p = \sum_{\alpha} \Gamma_n^\alpha \text{sym}(\mathbf{s}^\alpha \otimes \mathbf{m}^\alpha)$$

2. Update the plastic strain with the slip increments $\Delta\gamma_{n+1}^\alpha$ that depend on the Schmid shear stresses τ_n^α

$$\tau_n^\alpha = \sigma_n : \text{sym}(\mathbf{s}^\alpha \otimes \mathbf{m}^\alpha) \quad , \quad \Delta\gamma_{n+1}^\alpha = \Delta\gamma_0 \text{sign}(\tau_n^\alpha) \left| \frac{\tau_n^\alpha}{\tau_y^\alpha} \right|^N$$

$$\epsilon_{n+1}^p = \epsilon_n^p + \sum_{\alpha} \Delta\gamma_{n+1}^\alpha \text{sym}(\mathbf{s}^\alpha \otimes \mathbf{m}^\alpha)$$

3. Compute the stress σ_{n+1} according to the updated plastic strain ϵ_{n+1}^p

$$\epsilon_{n+1}^e = \epsilon_{n+1} - \epsilon_{n+1}^p \quad , \quad \sigma_{n+1} = \mathbb{C} : \epsilon_{n+1}^e$$

4. Update the history variables and return the stress tensor σ_{n+1}

$$\Gamma_n^\alpha = \Gamma_n^\alpha + \Delta\gamma_{n+1}^\alpha \quad , \quad \sigma_n = \sigma_{n+1}$$

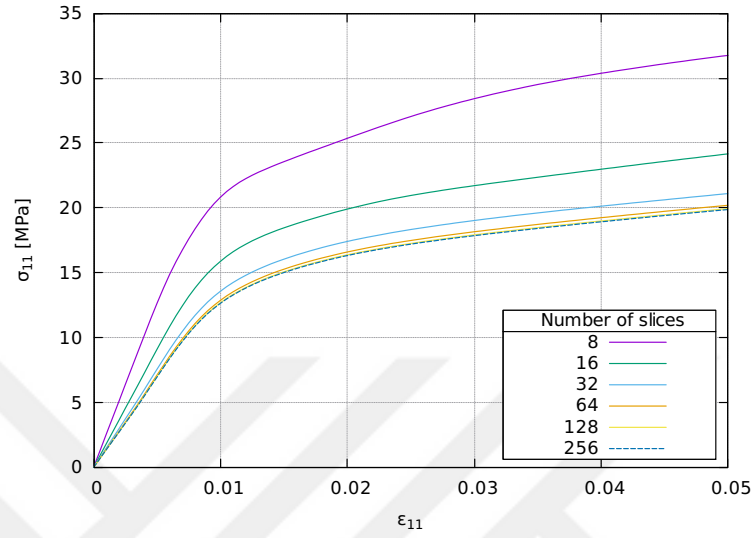
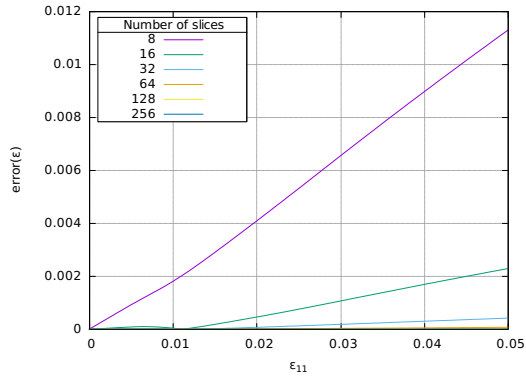
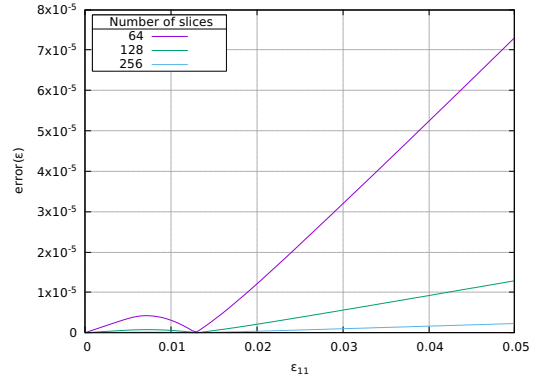


Figure 4.5: Variation of the stress-strain response with the number of slices under simple tension



(a) For all number of slices



(b) For slice numbers greater than 32

Figure 4.6: Norm of the error of the condition (4.19)

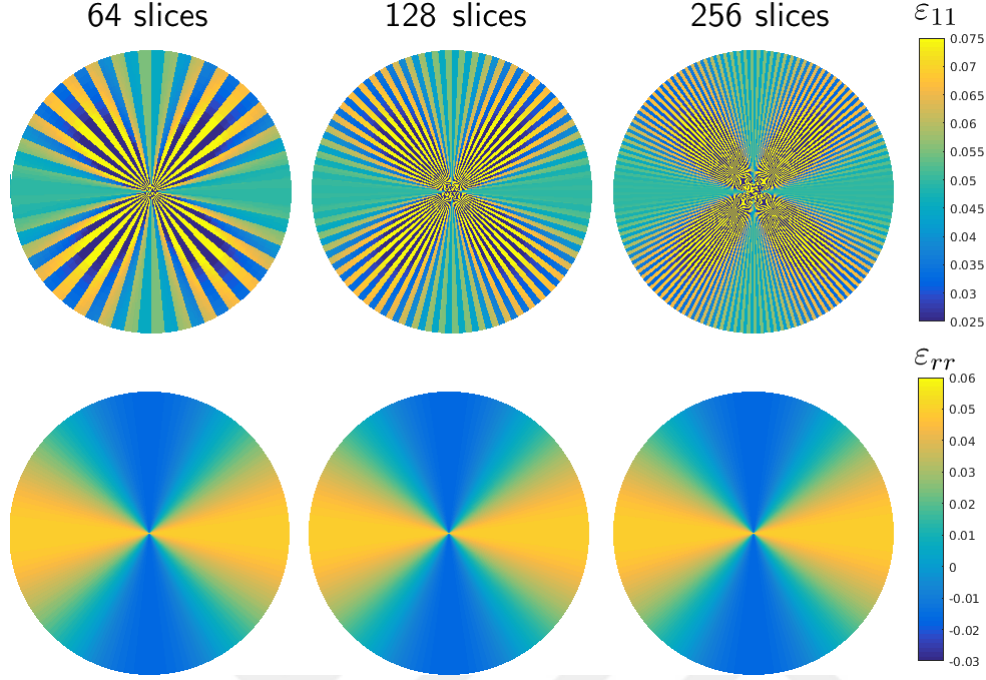


Figure 4.7: Distribution of ε_{11} and ε_{rr} strain components with the number of slices at $\varepsilon_{11} = 0.05$ under simple tension

component is completely the result of the rotation of the basis vectors with the polar coordinate θ . As a consequence, the variation of this strain does not depend on the polar coordinate r in Figure 4.7. Since the loading is along the horizontal, ε_{rr} is positive along the horizontal and negative in the vertical direction, as expected [64]. The strain component ε_{11} , which is along the loading direction, is maximum at the nucleus (center) of the spherulite. Strain along the loading direction is, in general, higher in the soft amorphous slices. One observation that is not in agreement with the observations on spherulites is that ε_{11} is not maximum in the equatorial region in Figure 4.7. It could be argued that this is a result of the Taylor-type homogenization inherently employed in the model.

Figure 4.8 presents the distribution of polar strain components $\varepsilon_{\theta\theta}$, $\varepsilon_{r\theta}$, $\varepsilon_{\theta 3}$ in the model. Considering the amorphous-crystalline interface normals lie along the e_θ basis vectors, $\varepsilon_{\theta\theta}$ indicates the interlamellar separation deformation mode while $\varepsilon_{r\theta}$ and $\varepsilon_{\theta 3}$ indicate the interlamellar shear deformation mode of the amorphous phase. As also discussed in Section 2.4.3.1, interlamellar separation is expected in equatorial regions and interlamellar shear is expected in inclined regions of the spherulites [80] shown

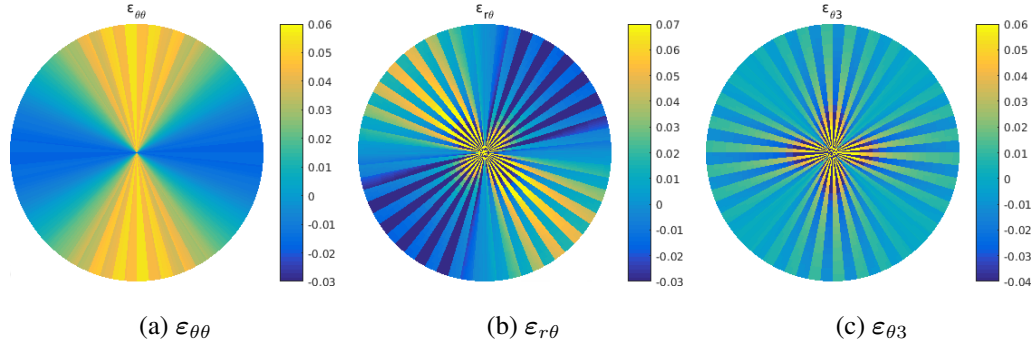


Figure 4.8: Spatial variation of the polar strain components on the model with 64 slices at $\varepsilon_{11} = 0.05$ under simple tension

in Figure 2.2. Moreover, negative interlamellar separation is observed in the polar regions, in the numerical study of Van Dommelen et al. [108]. Therefore, Figures 4.8a and 4.8b are in a very good agreement with the literature on the locations of the interlamellar separation and interlamellar shear deformation modes of the amorphous phase in a spherulite. On the other hand, in Figure 4.8c, $\varepsilon_{\theta 3}$ is not high in the inclined regions. Significant difference between the distributions of the $\varepsilon_{r\theta}$ and $\varepsilon_{\theta 3}$ strain components, while both indicate the interlamellar shear, is expected for the modeled disk-like spherulite geometry. According to Figure 4.8c there is a significant amount of out of plane deformation in the model.

In Figure 4.9, the variation of the response with the crystallinity ratio is presented. The distribution of the amorphous and crystalline phases of the models with different crystallinity ratios are presented in Figure 4.9 as well. The stiffness increases with the degree of crystallinity, as expected. In Figure 4.10, the initial Young's modulus and the yield stresses of the curves in Figure 4.9 are compared with the experimental results of Crist et al. [25], disk-like and three dimensional spherulite FEM models in Chapter 2. Crist et al. [25] presents the yield stress as the stress at the point of intersection of the tangents to the engineering stress-strain curves at approximately 0 % and 20 % engineering strains. The strain range presented in Figure 4.9 is narrower than the range studied by Crist et al. [25]. Therefore, the tangent is drawn at 5 % instead of 20 % strain, to compute the yield stress. Results of the quenched samples of Crist et al. [25] are not plotted due to the lack of time for the growth of spherulite

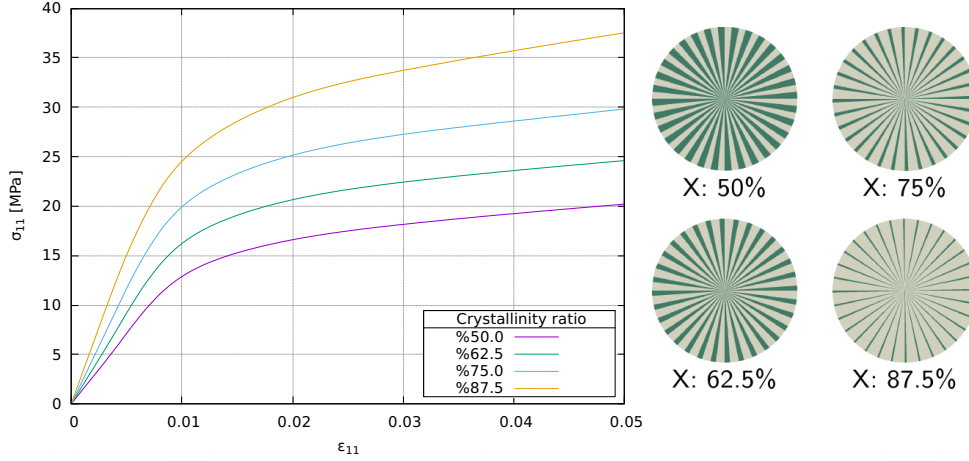


Figure 4.9: Variation of the uniaxial stress-strain response with crystallinity under simple tension and the views of the amorphous (dark colored) and crystalline slices (light colored) of the models with different crystallinity

morphology on those samples. While the slope of the yield stresses of the FE models and this study are similar in Figure 4.10a, yield stresses of this study are closer to the results of the experimental study. The model response is stiffer than the experiments. Especially the Young's modulus is an order of magnitude greater than the experimental measurements. The high Young's modulus is expected since the Taylor-type homogenization is upper bound of homogenization, and is inherently employed in the model.

Slip system activities at $\epsilon_{11} = 0.05$ for the model with 50 % crystallinity ratio are reported in this paragraph. In Table 4.2 relative activity of each slip system, defined in (2.10), is presented in the equatorial, polar and inclined regions of the model shown in Figure 2.2. $(100)[001]$ and $\{110\}\langle 1\bar{1}0 \rangle$ are the dominant slip systems of the equatorial and polar regions, respectively. $\{110\}\langle 1\bar{1}0 \rangle$, $(100)[001]$, $(100)[010]$ systems are among the most active slip systems of the entire model. In Figure 4.11, evolution of the slip system activity with the macroscopic strain ϵ_{11} is presented. In Figure 4.11a, evolution of the accumulated slip in the entire model is presented for each slip system, separately. In literature, $(100)[001]$ is reported as the most active slip system

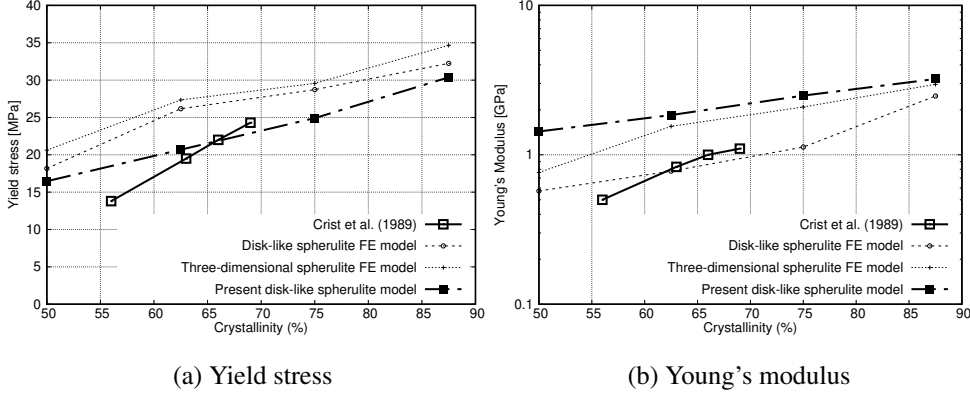


Figure 4.10: Comparison of the model prediction of variation of Young's modulus and yield strength with crystallinity ratio with the experiments of Crist et al. (1989)

of PE. Moreover, (100)[001] is expected to be the first slip system that is activated. Although (100)[001] is the most active slip system in the majority of the loading history in Figure 4.11a, the slip system that activates first is (100)[010]. This condition is related with the low ε_{11} strains in equatorial regions in Figure 4.7, where the discrepancy was associated with the Taylor approach employed in the model. In Figure 4.11b evolution of the total accumulated slip, defined in Section 2.4.3.1, is presented for the regions of the model and the entire model. Figure 4.11b presents the same slip system activity measure with the Figure 2.34a of the three dimensional spherulite FEM model. The volume fraction of the inclined region lamellae of this study is less than that of the three dimensional model. Therefore, inclined region activity is less in Figure 4.11b than Figure 2.34a.

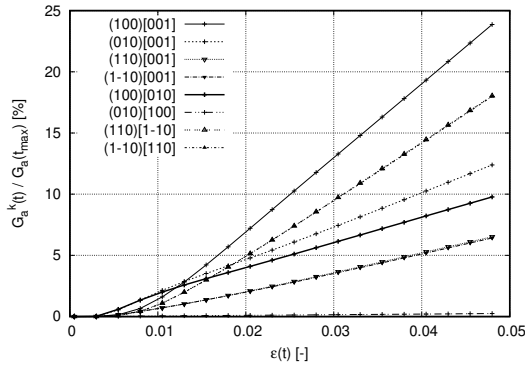
When a single integration point is employed in the radial direction at $r = R/2$, it is numerically found that the Hill's energy criterion is approximately satisfied

$$\int \bar{\sigma} : \dot{\bar{\epsilon}} dt \approx \frac{1}{\pi R^2} \sum_{I=1}^{N^S} \int \int_{\mathcal{B}^I} \sigma^I : \dot{\epsilon}^I dV dt$$

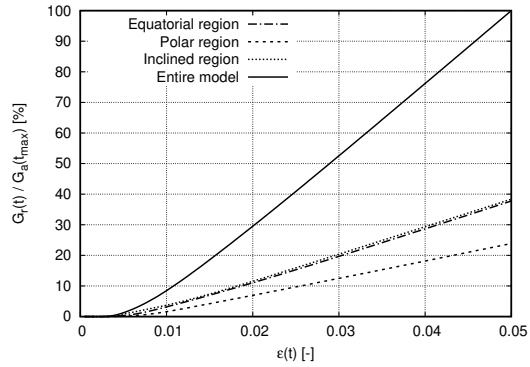
with higher accuracies for increasing number of slices. Notice that $r = R/2$ is the radial coordinate employed for the traction equilibrium of the interfaces in (4.20). Comparison of the response of the models with 256 slices, with different number

Table 4.2: Relative activities of the slip systems, in terms of the accumulated slip, in the regions of the model and the entire model at $\varepsilon_{11} = 0.05$ under simple tension

	$(100)[001]$	$(010)[001]$	$(110)[001]$	$(1\bar{1}0)[001]$	$(100)[010]$	$(010)[100]$	$(110)[1\bar{1}0]$	$(1\bar{1}0)[110]$
Equatorial (%)	48.1	7.9	7.7	6.1	8.2	0.2	11.2	10.6
Polar (%)	6.3	6.6	4.8	3.2	0.9	0.0	40.8	37.5
Inclined (%)	14.1	21.9	7.3	9.6	17.9	0.5	13.1	15.6
Entire model (%)	25.1	13.0	6.8	6.7	10.2	0.3	19.0	18.9



(a)



(b)

Figure 4.11: Slip system activity in the regions of the model

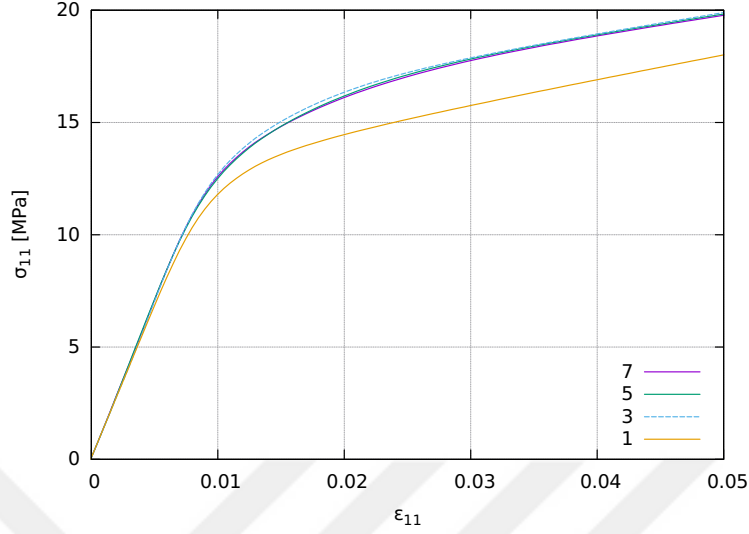


Figure 4.12: Influence of number of integration points in radial direction for models with 256 slices

of integration points in the radial direction is presented in Figure 4.12. According to (4.17) the strain field of the model does not vary linearly in the radial direction. Therefore, the stress field does not vary linearly in the radial direction as well and a single integration point along the polar coordinate r provides a different stress-strain response in Figure 4.12 due to loss of accuracy of the numerical integration. Since the stress-strain response of the model depends on the volume average of the stress and strain fields, the response does not depend on the value of the radius R . Therefore, in order to obtain a reasonable accuracy with the single integration point case, which approximately satisfies the Hill's energy criterion, instead of a whole disk, a disk with a circular concentric hole could be modeled.

4.5 Extension of the Strain Field for Different Interaction Laws

In (4.17), components of the fluctuation field of a slice D , $\tilde{\epsilon}^D = \epsilon^D - \bar{\epsilon}$, in the $e_r \otimes e_r$, $e_r \otimes e_3$ and $e_3 \otimes e_3$ bases are zero. When e_θ is the interface normal, the kinematical compatibility on the interface of a laminate demands these strain com-

ponents to be the same in all phases. Therefore, when Taylor approach is employed these fluctuation field components are trivially zero, as presented in (4.13). However, these fluctuation field components are necessary when an interaction law other than the Taylor approach is employed. In that case, while these fluctuation field components should be the same on both sides of an interface, their values are not necessarily the same for all interfaces of the model. These components are introduced to (4.17) as follows

$$\begin{aligned}
\epsilon^D(r, \theta) = & \bar{\epsilon} \\
& + \frac{\theta_f^D - \theta}{\theta_f^D - \theta_s^D} [\alpha_s^D \mathbf{e}_\theta \otimes \mathbf{e}_\theta + \delta_s^D \mathbf{e}_r \otimes \mathbf{e}_r] \\
& + \frac{\theta_f^D - \theta}{\theta_f^D - \theta_s^D} \text{sym} \left(\frac{\gamma_s^D}{r} \mathbf{e}_r \otimes \mathbf{e}_\theta + \frac{\beta_s^D}{r} \mathbf{e}_\theta \otimes \mathbf{e}_3 + r \phi_s^D \mathbf{e}_r \otimes \mathbf{e}_3 \right) \\
& + \frac{\sin(\theta_f^D - \theta)}{\sin(\theta_f^D - \theta_s^D)} r \eta_s^D \mathbf{e}_3 \otimes \mathbf{e}_3 \\
& + \frac{\theta - \theta_s^D}{\theta_f^D - \theta_s^D} [\alpha_f^D \mathbf{e}_\theta \otimes \mathbf{e}_\theta + \delta_f^D \mathbf{e}_r \otimes \mathbf{e}_r] \\
& + \frac{\theta - \theta_s^D}{\theta_f^D - \theta_s^D} \text{sym} \left(\frac{\gamma_f^D}{r} \mathbf{e}_r \otimes \mathbf{e}_\theta + \frac{\beta_f^D}{r} \mathbf{e}_\theta \otimes \mathbf{e}_3 + r \phi_f^D \mathbf{e}_r \otimes \mathbf{e}_3 \right) \\
& + \frac{\sin(\theta - \theta_s^D)}{\sin(\theta_f^D - \theta_s^D)} r \eta_f^D \mathbf{e}_3 \otimes \mathbf{e}_3; \quad \forall D \in \{1, 2, \dots, N^S\}. \quad (4.22)
\end{aligned}$$

Note that (4.22) reduces to (4.17) when the values of δ_s^D , δ_f^D , ϕ_s^D , ϕ_f^D , η_s^D , η_f^D are zero. These terms determine the values of the strain fluctuation components in $\mathbf{e}_r \otimes \mathbf{e}_r$, $\mathbf{e}_r \otimes \mathbf{e}_3$ and $\mathbf{e}_3 \otimes \mathbf{e}_3$ bases. The expression in (4.22) is also a kinematically compatible strain field for a slice, and can be employed to provide a kinematically compatible strain field between laminates. In order to satisfy the kinematical compatibility on the interfaces with (4.22), following equalities should be satisfied

$$\begin{aligned}
\delta_f^I - \delta_s^J &= 0 \\
\phi_f^I - \phi_s^J &= 0 \\
\eta_f^I - \eta_s^J &= 0; \quad \forall I \in \{1, 2, \dots, N^S\} \quad (4.23)
\end{aligned}$$

$$\text{where } J = \begin{cases} 1 & I = N^S \\ I + 1 & \text{otherwise} \end{cases}.$$

It should be noted that (4.16) is not necessary for the kinematical compatibility of (4.22). With (4.16) the fluctuation fields on opposite sides of an interface have opposite signs and therefore (4.19) is satisfied when the Taylor approach is employed. In (4.22), there are twelve unknowns for each slice. With the three equations in (4.23), the number of unknowns drops to nine and therefore the total number of unknowns of all slices is $9N^S$. In this section, $6N^S$ additional degrees of freedom are introduced to the model presented in Section 4.3. The additional degrees of freedom are necessary for the six components of $\varepsilon^I - \bar{\varepsilon}$ tensor, which are zero when the Taylor approach is employed, where ε^I is the mesoscopic strain employed in laminate I .

4.6 Conclusion

A compatible strain field that is proposed in the cylindrical coordinate system is employed between the interfaces of neighboring laminates homogenized with the Taylor-type homogenization. Laminates that are oriented similarly with the disk-like spherulite model in Section 2.3 are considered. Although the strain field is compatible throughout the model, the stress field is not statically admissible since the traction equilibrium is enforced only at a single point on each interface. The response of the model is obtained by numerically integrating the stress field with the Gauss quadrature, while the average strain of the model is computed algebraically. The average strain is approximately equal to the macroscopic strain when the number of slices is sufficiently high. The response of the model converges with increasing number of slices. The strain field and the slip system activities of the model are in reasonable agreement with the expectations for a spherulite. The Hill's energy criterion is not satisfied when the integration points employed in the radial direction is higher than one and is not at the same radial coordinate where the traction equilibrium is satisfied on the interfaces.

When the average stress of the the model is computed only with the integration points

where the traction equilibrium is satisfied, the model would reduce to the homogenization of the rank-1 laminates with the Taylor approach. Nevertheless, the proposed strain fields in Sections 4.3 and 4.5 could be employed to calculate the spatial derivatives of the strain fields, which are required when non-local constitutive models are to be employed with the rank-1 laminates. Since the strain fields are uniform in the phases of the original rank-1 laminates, the derivatives of the stress and strain fields with respect to position are zero.



CHAPTER 5

A HOMOGENIZATION-BASED CONSTITUTIVE MODEL FOR SEMICRYSTALLINE POLYMERS WITH SPHERULITE MORPHOLOGY

In this section, a homogenization-based constitutive model is proposed for semicrystalline polymers, and its application to the high density polyethylene (HDPE) is presented. The model is composed of N^R number of randomly oriented realizations of the disk-like spherulite model, as schematically presented in Figure 5.1. Each realization I is solved with the statically equilibrated formulation with bonded end interfaces as described in Section 3.4.2. As the interaction law, the Taylor approach

$$\mathbf{F}^I = \bar{\mathbf{F}} \quad \forall I \in \{1, \dots, N^R\} \quad \text{and} \quad \bar{\mathbf{P}} = \frac{1}{N^R} \sum_{i=1}^{N^R} \mathbf{P}^i$$

is employed, where $\bar{\mathbf{F}}$ and $\bar{\mathbf{P}}$ are the macroscopic deformation gradient and the macroscopic first Piola-Kirchhoff stress tensor, respectively, while $\mathbf{F}^I$ and $\mathbf{P}^I$ are the mesoscopic deformation gradient and the first Piola Kirchhoff stress tensor of the realization I .



Figure 5.1: Randomly oriented realizations of the disk-like spherulite model. Dark and light regions are amorphous and crystalline, respectively.

Perhaps the most widely studied semicrystalline polymer is the high density polyethylene (HDPE), which often has a spherulitic microstructure. Van Dommelen et al. [109], Nikolov et al. [78], Agoras and Castaneda [2] are among the recent homogenization-based constitutive models for semicrystalline polymers that also study the deformation of the HDPE. In Section 5.3, mainly the results that are comparable to that of the Van Dommelen et al. [109] are produced with the constitutive models presented in Section 5.1 and the loading procedure in Section 5.2.

5.1 Constitutive Models and Material Parameters

The model of crystal viscoplasticity and the associated material parameters in Section 2.2 are employed in the crystalline phase. For the amorphous phase, elasto-viscoplastic model of Van Dommelen et al. [109] is adopted. For the sake of completeness, this model is described in Section 5.1.1.

The employed constitutive models and material parameters are very similar to that of Van Dommelen et al. [109]. The only difference is in the computation of the Schmid stress in (2.7) of the crystal plasticity, where Van Dommelen et al. [109] considers the influence of the stretch for transforming the stress to the intermediate configuration. Since the critical yield stress of the crystalline phase, which is taken as 20 MPa at most, is well below the elastic moduli, which is at least 1600 MPa, the elastic distortion of the lattice is quite low.

5.1.1 Constitutive Model of the Amorphous Phase

Above the glass transition temperature the amorphous phase of semicrystalline polymers, while elastically softer, is plastically stronger than the crystalline phase [64]. Considering the tie molecules, the constraints of the adjacent lamellae are also considered to hinder the plastic deformation of the amorphous phase [13]. Therefore, above the glass transition temperature, constitutive models of rubber elasticity, which take the locking stretch phenomenon into account, are suggested as good estimates for the amorphous phase constitutive model to obtain a macroscopic stress-strain response [13]. However, among the micromechanical constitutive models of semicrystalline

polymers, plasticity of the amorphous phase is widely taken into account especially since the employment of rubber elasticity models such as the 8-chain model [8] results in a less significant yield-plateauing compared to the experiments [107]. In this section, the elasto-viscoplastic constitutive model of Van Dommelen et al. [109] for the amorphous phase is presented.

Multiplicative decomposition of the deformation gradient $\mathbf{F} = \mathbf{F}^e \mathbf{F}^p$ into the elastic $\mathbf{F}^e$ and plastic $\mathbf{F}^p$ components is employed. A generalized neo-Hookean formula

$$\boldsymbol{\sigma} = \frac{G}{J} \left(\tilde{\mathbf{B}}^e - \frac{\text{tr}(\tilde{\mathbf{B}}^e)}{3} \mathbf{1} \right) + \kappa (J - 1) \mathbf{1}$$

is given for the Cauchy stress in terms of the volume change factor $J = \det(\mathbf{F})$ and the left Cauchy-Green tensor $\tilde{\mathbf{B}}^e = \tilde{\mathbf{F}}^e \tilde{\mathbf{F}}^{eT}$ of the volume-preserving part of the elastic component of the deformation gradient $\tilde{\mathbf{F}}^e = J^{-3} \mathbf{F}^e$ considering $\det(\mathbf{F}^p) = 1$. Material parameters κ and G are the bulk and shear moduli, respectively. As the flow rule, a power-law relationship

$$\dot{\gamma} = \dot{\gamma}_0 \left(\frac{\tau}{\tau_0} \right)^n \quad (5.1)$$

is employed where $\dot{\gamma}_0$, τ_0 , n are the reference shear rate, reference strength and the rate sensitivity parameter, respectively. The stress

$$\tau = \sqrt{\frac{1}{2} \boldsymbol{\sigma}_* : \boldsymbol{\sigma}_*}$$

that drives the plastic flow is defined in terms of

$$\boldsymbol{\sigma}_* = \mathbf{R}^{eT} \boldsymbol{\sigma} \mathbf{R}^e - \mathbf{H} \quad (5.2)$$

in the intermediate configuration where kinematical hardening is introduced with the 8-chain rubber elasticity [8]

$$\mathbf{H} = \mu_R \frac{\sqrt{N}}{\lambda_{\text{chain}}} \mathcal{L}^{-1} \left(\frac{\lambda_{\text{chain}}}{\sqrt{N}} \right) (\mathbf{B}^p - \lambda_{\text{chain}}^2 \mathbf{1}) \quad (5.3)$$

that is employed as the back stress tensor. In (5.3), $\lambda_{\text{chain}} = \sqrt{\frac{1}{3} \text{tr}(\mathbf{B}^p)}$ and $\mathcal{L}(\beta) = \coth(\beta) - 1/\beta$ is the Langevin function. $\mathbf{B}^p = \mathbf{F}^p \mathbf{F}^{pT}$ is the left Cauchy-Green tensor of the plastic component $\mathbf{F}^p$ of the deformation gradient. Evolution of the plastic part of the deformation gradient is given by the plastic velocity gradient defined as

$$\dot{\mathbf{F}}^p \mathbf{F}^{p-1} = \frac{\dot{\gamma}}{\tau} \boldsymbol{\sigma}_*,$$

Table 5.1: Material parameters employed in the amorphous phase

κ	G	$\dot{\gamma}_0$	τ_0	n	μ_R	N
[MPa]	[MPa]	[s ⁻¹]	[MPa]	[-]	[MPa]	[-]
2000	35	1×10^{-3}	1.2×8	9	1.6	49

where $\boldsymbol{\sigma}'_*$ is the deviatoric part of $\boldsymbol{\sigma}_*$. The values of the material parameters are presented in Table 5.1, and are also adopted from Van Dommelen et al. [109].

5.2 Load Application Procedure

Similar to Van Dommelen et al. [109], a uniaxial loading is applied with the constant true strain rate $\dot{\varepsilon}$, such that the macroscopic uniaxial true strain at time t is

$$\varepsilon(t) = \eta \dot{\varepsilon} t,$$

where $\eta = 1$ for tension and $\eta = -1$ for compression. The strain is applied by defining

$$U_{11} = \exp(\eta \dot{\varepsilon} t)$$

component of the right stretch tensor $\boldsymbol{U}$, and rotation part of the deformation gradient

$$\boldsymbol{F} = \boldsymbol{R}\boldsymbol{U} \quad (5.4)$$

is $\boldsymbol{R} = \mathbf{1}$. The remaining components of the symmetric right stretch tensor $\boldsymbol{U}$ are solved for a uniaxial stress state

$$\sigma_{22} = \sigma_{33} = \sigma_{12} = \sigma_{23} = \sigma_{13} = 0, \quad (5.5)$$

where $\boldsymbol{\sigma}$ is the macroscopic Cauchy stress tensor. An equivalent stress tensor is defined as

$$\sigma^{eq}(\boldsymbol{\sigma}) = \sqrt{\frac{3}{2} \boldsymbol{\sigma}' : \boldsymbol{\sigma}'} \quad (5.6)$$

where $\boldsymbol{\sigma}' = \boldsymbol{\sigma} - \frac{1}{3} \text{tr}(\boldsymbol{\sigma}) \mathbf{1}$ is the deviatoric part of the stress. Notice that $\sigma^{eq} = \sigma_{11}$ according to (5.5) and (5.6).

Following Van Dommelen et al. [109], shear deformation, that originates from a stress free state with a symmetric deformation gradient $\bar{\boldsymbol{F}}_0$, is also studied. For a shear

strain strain γ_{12} , a right stretch tensor $\bar{U}$ is obtained from the polar decomposition of $(\mathbf{1} + \gamma_{12}\mathbf{e}_2 \otimes \mathbf{e}_1) \bar{\mathbf{F}}_0$. Loading is applied with $\bar{\mathbf{F}} = \bar{U}$, where the $\bar{U}_{12}$, $\bar{U}_{11}$, $\bar{U}_{22}$ components of $\bar{U}$ are taken as constant, while the remaining are solved to obtain a plane stress state

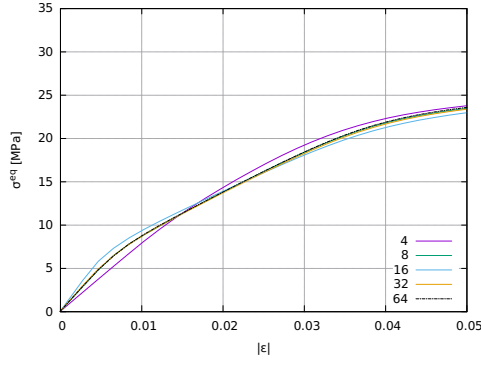
$$\sigma_{33} = \sigma_{23} = \sigma_{13} = 0$$

for $\gamma = \sqrt{3}\dot{\epsilon}\Delta t$, where Δt is the elapsed time of shear deformation.

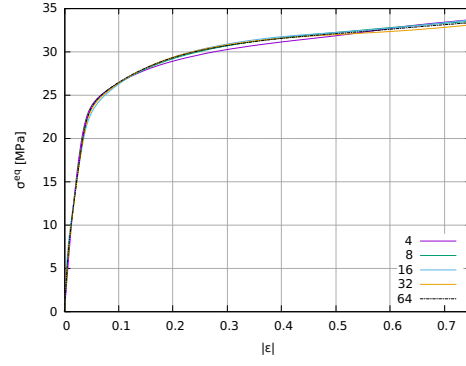
5.3 Results

Simple compression deformation mode under true strain rate $\dot{\epsilon} = 1 \times 10^{-3} \text{ s}^{-1}$ is studied. The crystallinity ratio is taken as 70% as in Van Dommelen et al. [109]. The influence of the number of realizations, i.e. the number of randomly oriented disk-like spherulite models, on the macroscopic stress-strain response is presented in Figure 5.2, where the new randomly oriented realizations are added to the set of the previous size. Influence of the number of realizations on the stress-strain curves is rather limited in Figure 5.2. In Figure 5.3, the influence of the selected different random orientations on the macroscopic stress-strain curves is presented for the models composed of 4 and 32 realizations, respectively. Influence of the selected random set of orientations on the stress-strain response is also low and decreases with increasing number of realizations, as expected.

In Figure 5.4, orientations of the crystallographic axes and the interface normals are presented for the model composed of randomly oriented 32 realizations with 32 slices, where the loading is applied along the x direction that is perpendicular to the y and z axes of Figure 5.4. The corresponding orientations at the end of loading is presented in Figure 5.5. Distribution of the poles in Figure 5.5 are approximately axially symmetric along the x axis due to the symmetry of the simple compression deformation mode. The lattice axes a and c move towards and away from the loading direction, respectively. This indicates the activity of the slip system $(100)[001]$, which has the smallest critical resolved shear stress in Table 2.4. The lamellae normals move towards the loading direction in Figure 5.5d, as a result of the interlamellar shear deformation mode of the amorphous phase. The crystallographic and morphological textures presented in Figure 5.4 are in agreement with the numerical studies in lit-

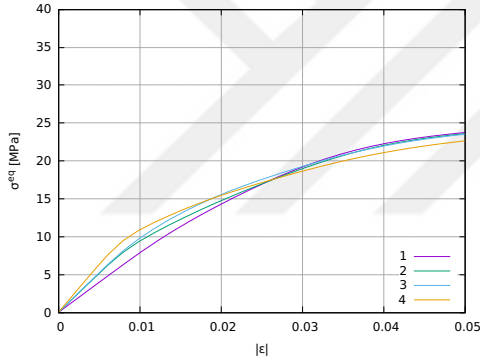


(a) For $|\bar{\epsilon}| \leq 5\%$

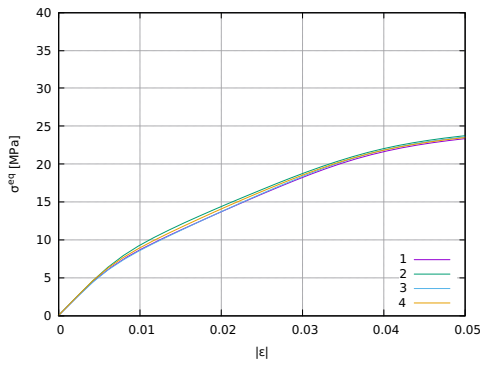
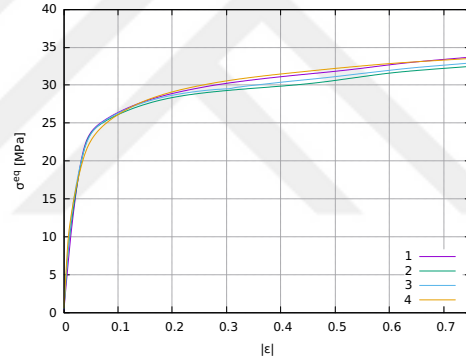


(b) For the complete $|\bar{\epsilon}|$ range

Figure 5.2: Influence of the number of randomly oriented realizations



(a) 4 realizations



(b) 32 realizations

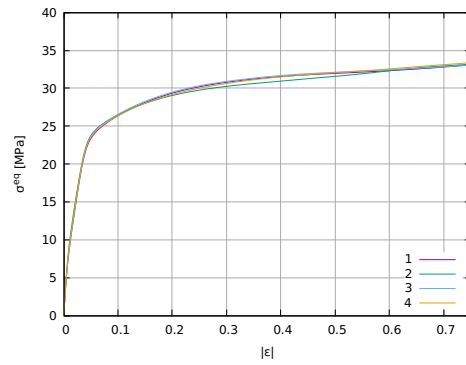


Figure 5.3: Variation of stress-strain response of the models composed of 4 and 32 realizations with different sets of random orientations, for $|\bar{\epsilon}| \leq 5\%$ and the complete strain range

erature [109, 78, 2] and the experimental observation of Bartczak et al. [12], where the evolution of the crystallographic texture of HDPE under uniaxial compression is studied. In order to emphasize that the model is composed of disk-like spherulite models, the initial and final textures of the seed-1 analysis of Figure 5.3a, with only 4 realizations are also presented in Figures 5.6 and 5.7, respectively. The initial texture is composed of circular patterns that represent the spherulite morphology, and it is therefore statistically non-uniform. The obtained texture in Figure 5.7 possesses the general characteristics of the texture evolution with only 64 PE crystals that are distributed into randomly oriented 4 disk-like spherulite models. It could be remarked that, while the initial texture is not statistically uniform at a material point, its variation in different material points of a body that possesses a polyspherulitic morphology is considered as statistically uniform. The results in the figures that follow Figure 5.7 are obtained with the initial orientations presented in Figure 5.4.

The variation of stress-strain response with the rate of loading is presented in Figure 5.8a. In Figure 5.8b, the equivalent stresses of Figure 5.8a, at $|\varepsilon| = 0.1$ and $|\varepsilon| = 0.5$ are presented together with that of Van Dommelen et al. [109], for comparison. The values in the horizontal axis of Figure 5.8b are normalized with the reference flow rate of $\dot{\gamma}_0 = 1 \times 10^{-3}$. As Van Dommelen et al. [109], an approximately log-linear relationship is present in Figure 5.8b, which is a result of the power-law flow rules (2.6) and (5.1) of the constitutive models of the crystalline and amorphous phases, respectively. Our model is slightly less sensitive to the loading rate than Van Dommelen et al. [109].

In Figure 5.9, the variation of the response with the degree of crystallinity is presented. The degree of crystallinity is varied as described in Section 3.6.6. The initial Young's modulus and the yield stress, which are computed as described in Section 2.4.3.3 according to the data points in Figure 5.9, are presented in Figure 5.10. The tangent lines for the yield stress calculation are drawn at 5% strain in order to be compatible with the results of Van Dommelen et al. [109]. The results of the experimental study of Crist et al. [25] are also given in Figure 5.10 in addition to that of Van Dommelen et al. [109] for comparison. As in Figures 2.22, 2.41 and 4.10, the results of the quenched samples of Crist et al. [25] are not plotted, since it is considered that there is not sufficient time for the growth of the spherulites in those samples.

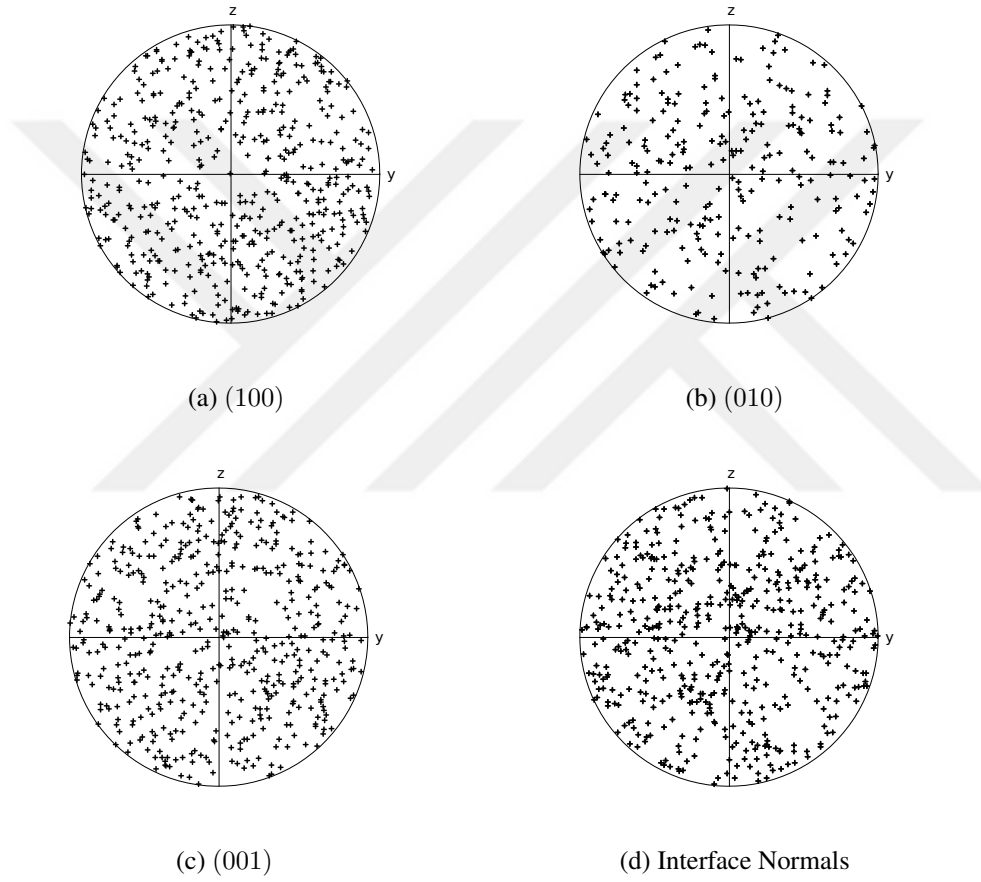


Figure 5.4: Equal area projection pole figures of the crystallographic lattice directions and the interface normals before load application

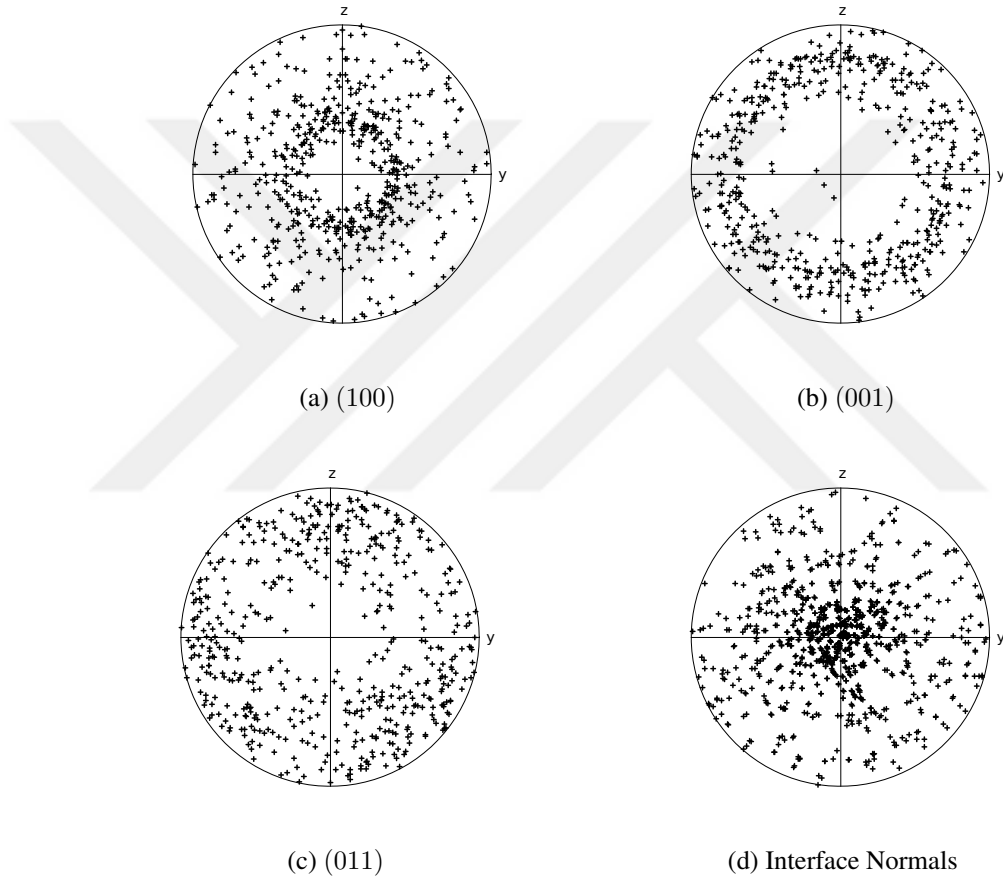


Figure 5.5: Equal area projection pole figures of the crystallographic lattice directions and the interface normals at $|\varepsilon| = 0.75$ in simple compression

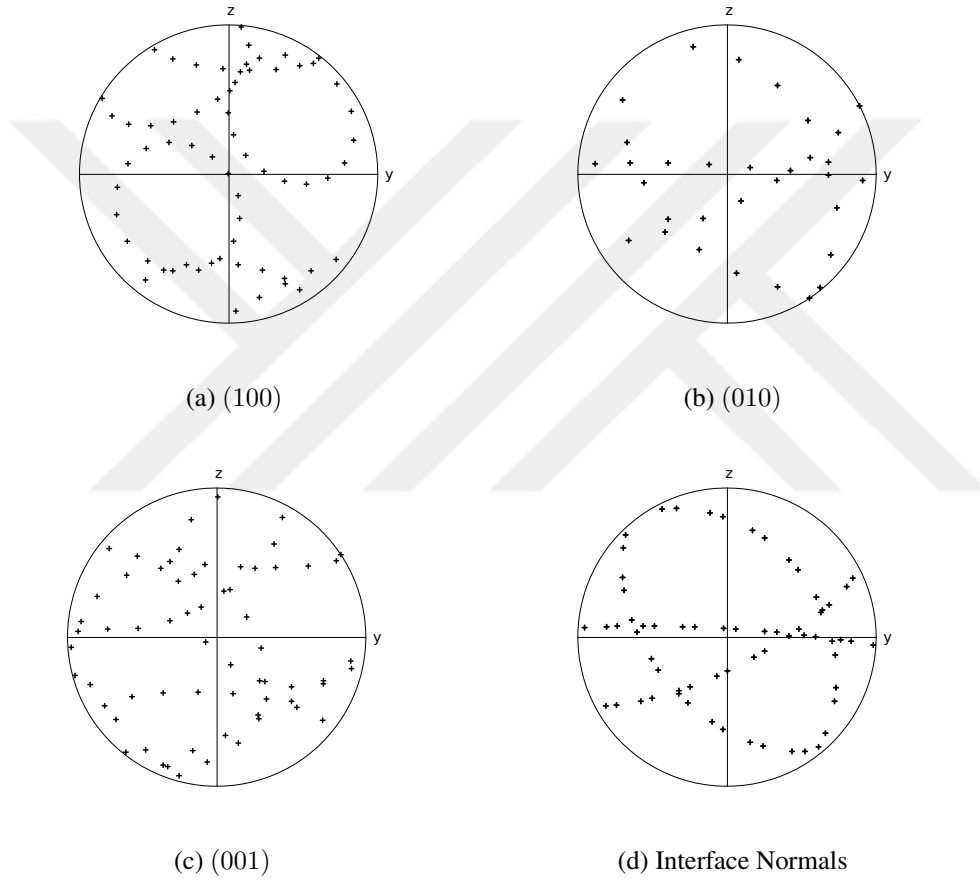


Figure 5.6: Equal area projection pole figures of the crystallographic lattice directions and the interface normals before load application, for 4 realizations

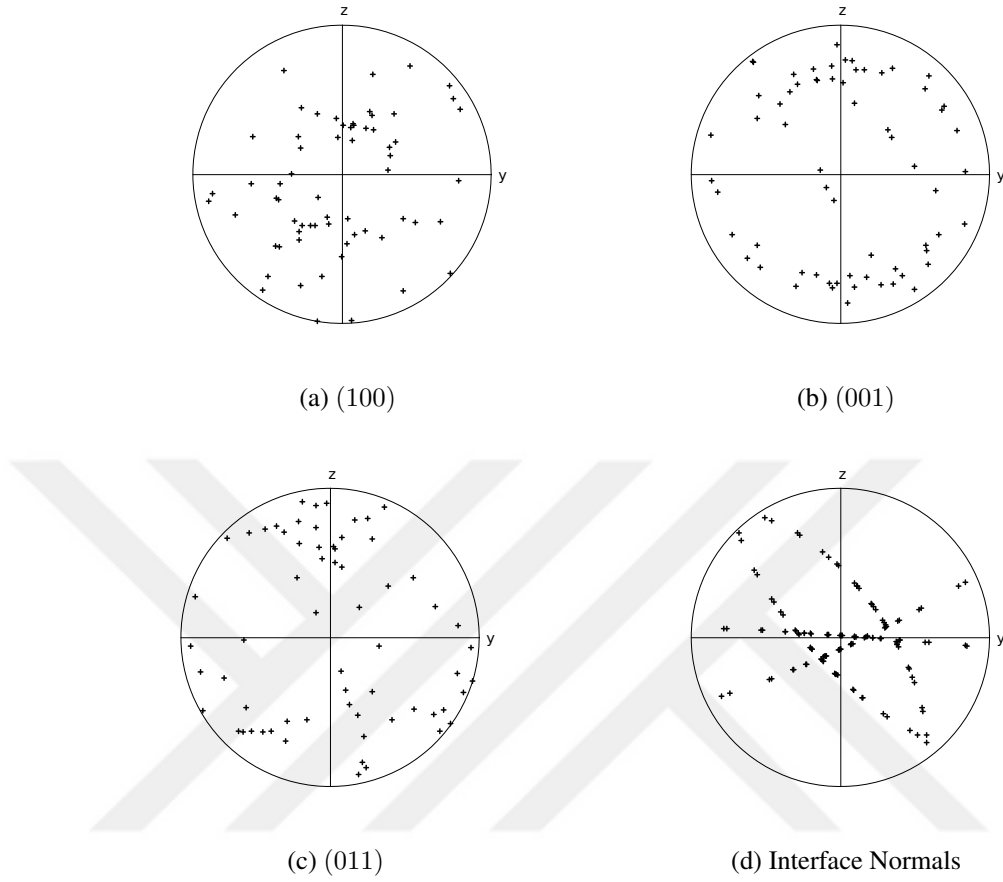


Figure 5.7: Equal area projection pole figures of the crystallographic lattice directions and the interface normals at $|\varepsilon| = 0.75$ in simple compression, for 4 realizations

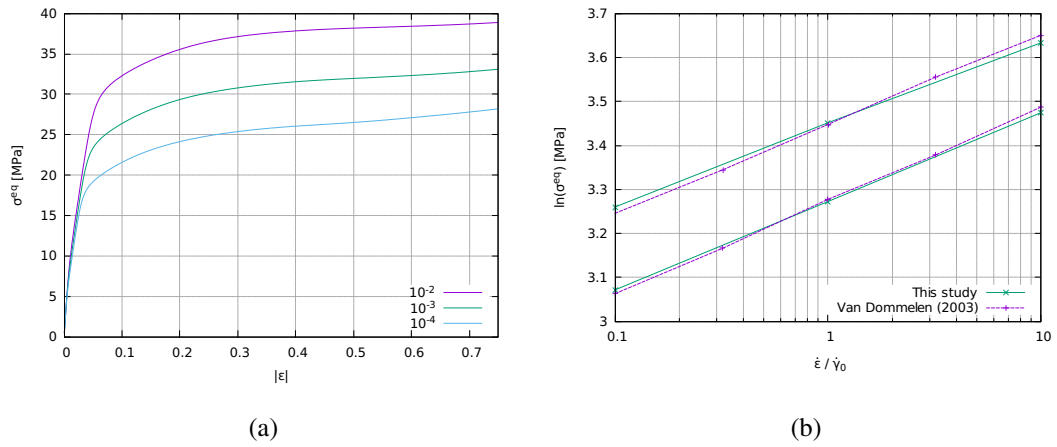


Figure 5.8: Variation of the (a) response (b) equivalent stresses at 10% and 50% equivalent strain with the rate of loading

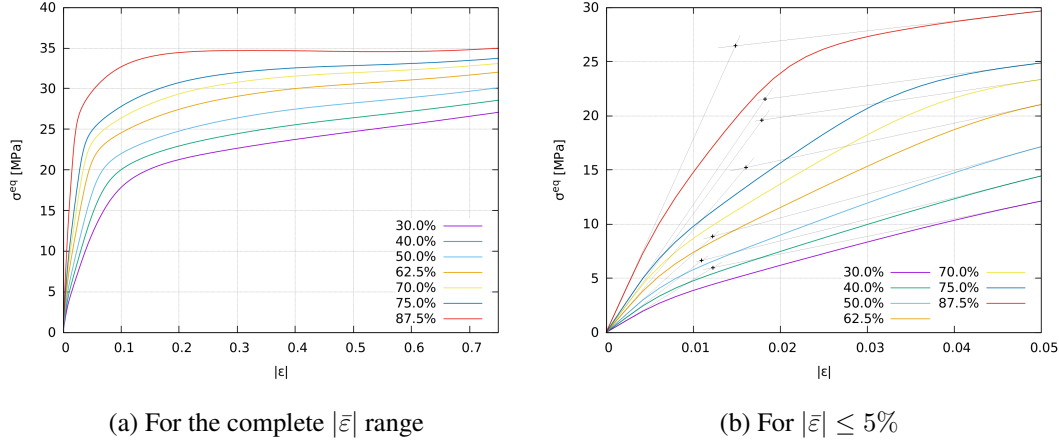


Figure 5.9: Influence of the degree of crystallinity

Both the Young's modulus and the yield stress of the model are slightly closer to the results of the Crist et al. [25], compared to that of the Van Dommelen et al. [109]. In Figure 5.11, variation of the response with the number of slices of the realizations are presented for different crystallinity ratios. Thirty-two slices are considered sufficient for the studied range of crystallinity ratio. In Figure 5.12, at true strains of $\epsilon = 0.10$, $\epsilon = 0.22$ and $\epsilon = 0.36$ the loading is reversed until a zero stress state is achieved and then the loading is applied again. As in Van Dommelen et al. [109], hysteresis loops are observed and the strain recovery decreases with increasing crystallinity ratio, since the crystalline phase mainly deforms plastically. Van Dommelen et al. [109] refers to a number of experimental studies where hysteresis loops are observed for polyethylene samples in a loading and unloading cycle.

In Figure 5.13, a uniaxial compression in x direction up to $|\epsilon| = 0.60$ is followed by an unloading up to a stress-free state. Beginning from the reached stress-free state, separate simple compression and shear analyses are conducted in different directions and the obtained stress-strain responses are presented in Figure 5.14, where both $|\epsilon| = 0$ and $\gamma = 0$ corresponds to the stress-free state reached in Figure 5.13. In Figure 5.14a, the stiffest response is along x , the previous loading direction. As required from the initial isotropy, and symmetry of the loading, very similar responses are obtained in the y and z directions that are transverse to the initial loading direction x . Therefore, similarity of γ_{xy} and γ_{xz} responses in Figure 5.14b are expected as

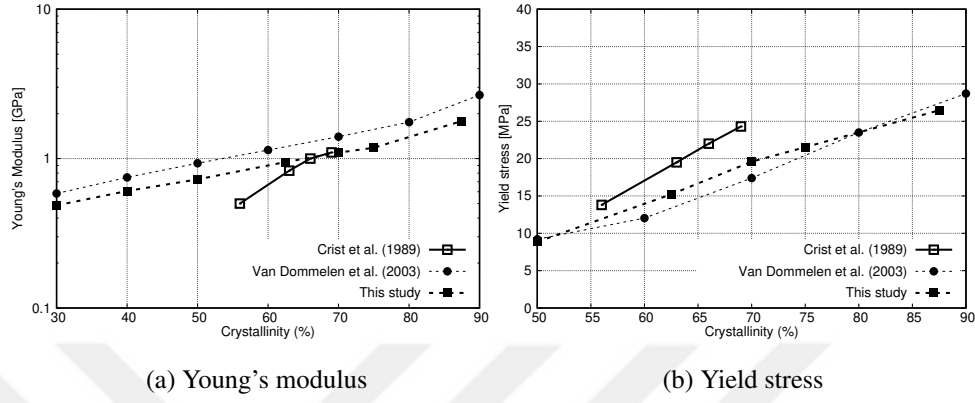


Figure 5.10: Variation of the Young's modulus and yield strength with the crystallinity ratio

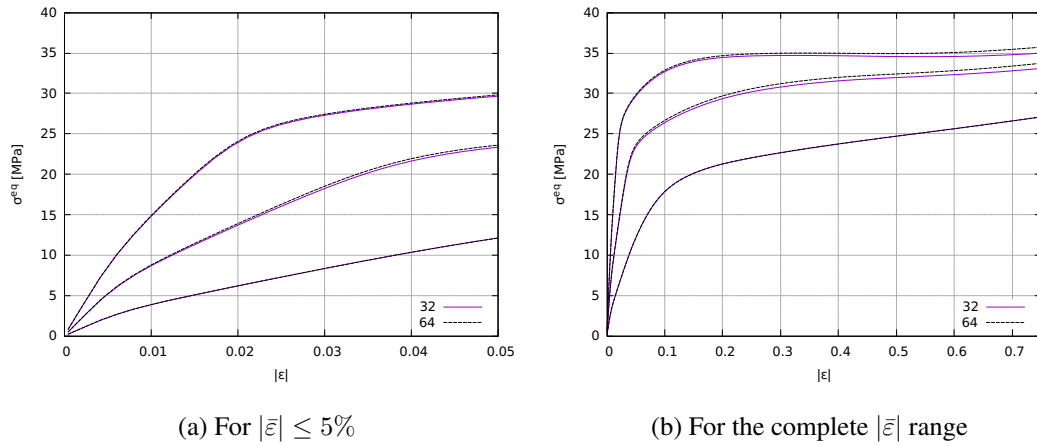


Figure 5.11: Variation of the response with the number of slices of the realizations with 30%, 70% and 87.5% crystallinity ratios

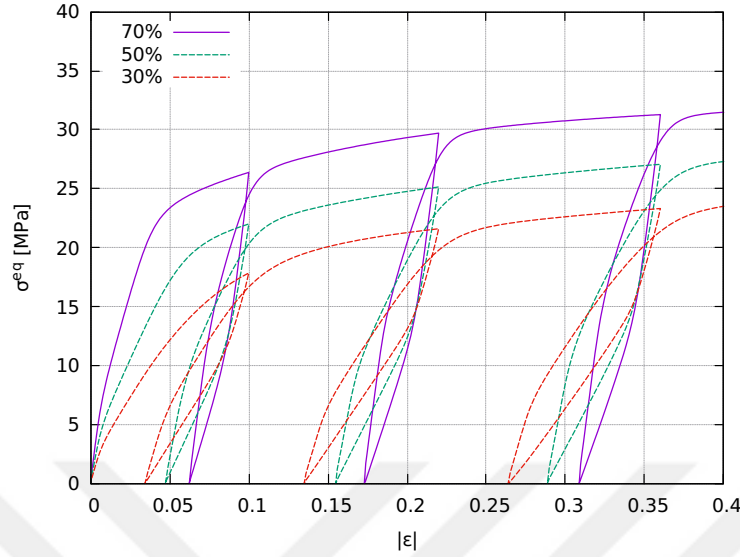


Figure 5.12: Cyclic application of the uniaxial compression for different crystallinity ratios

well. The γ_{yz} response is the stiffest as in Van Dommelen et al. [109]. The presented anisotropic responses are due to the developed crystallographic and morphological texture, in addition to the macromolecular texture evolution that is accounted for by the kinematic hardening of the amorphous phase constitutive model, and are in good agreement with Van Dommelen et al. [109].

In Figure 5.15, uniaxial compression response of the model is compared with the studies in literature including the experiments of Bartczak et al. [12]. Curves of the studies in Figure 5.15b are as presented in Agoras and Castaneda [2]. Constitutive models employed in the amorphous and crystalline phases of our model are adopted from [109], along with their material parameters. The obtained responses are similar to the other models in the literature. The experimental study of Bartczak et al. [12] is an influential study in literature that explains the large deformation of PE with crystallographic deformation mechanisms, and we have also frequently referred to Bartczak et al. [12] in Chapter 2. While their emphasis is not on the stress-strain response, they have provided a typical true-stress strain curve of the samples. Their samples are composed of spherulites with approximately $5\mu\text{m}$ diameter, and the crystallinity ratio is estimated to exceed 70%. They report that at 73% permanent strain, the spherulites are intact but more difficult to recognize. At 82% permanent strain, however, defor-

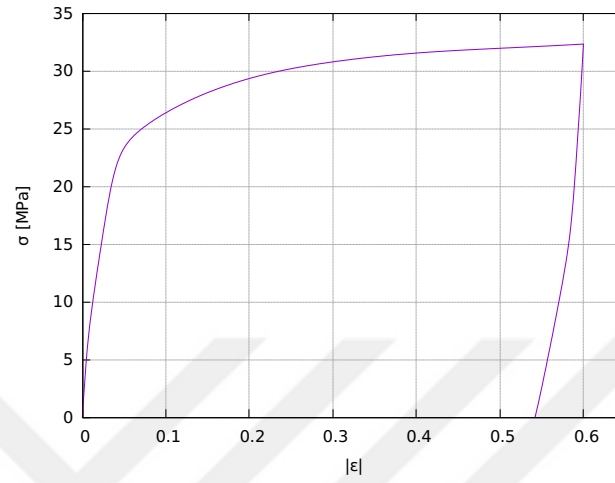
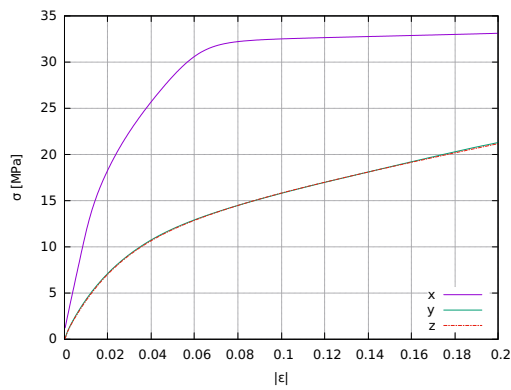
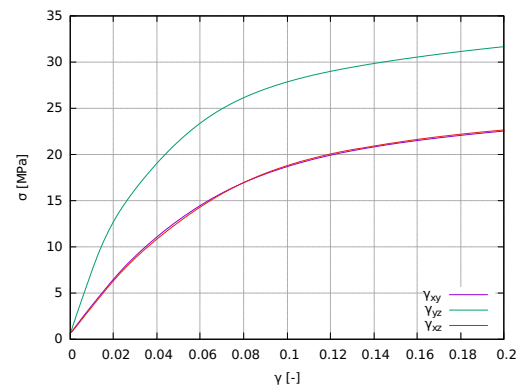


Figure 5.13: Uniaxial compression in x direction and unloading to a zero stress state



(a) Uniaxial compression along different directions



(b) Shear along different directions

Figure 5.14: A demonstration of the strain induced anisotropy of the model, starting from the zero stress state reached in Figure 5.13

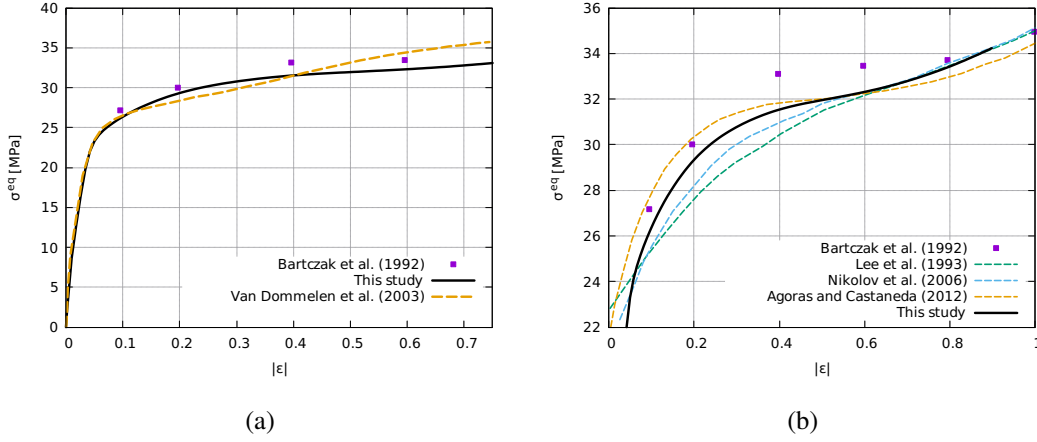


Figure 5.15: Comparison of the response of the model under uniaxial compression with the self-consistent-like models of Van Dommelen et al. [109], Lee et al. [62], and self-consistent models of Nikolov et al. [78], Agoras and Castaneda [2], and the experiment of Bartczak et al. [12]

mation begins to localize into densely packed fine shear bands. The results of the experimental study of Bartczak et al. [12] are in a rather good agreement with our study and also with Agoras and Castaneda [2].

Bartczak et al. [11] conducted simple shear experiments under a constant engineering strain rate of $1 \times 10^{-3} \text{ s}^{-1}$, and presented a representative engineering stress-strain curve and pole figures for the texture evolution. The properties of the employed HDPE are similar to that of their compression study [12]. Both Nikolov et al. [78] and Agoras and Castaneda [2] present comparisons with this study. In order to be consistent with the load application procedure of Bartczak et al. [11], the constant engineering strain rate $1 \times 10^{-3} \text{ s}^{-1}$ is employed. Considering the initial pole figures in Figure 5.4, shear deformation is assigned to the $\bar{F}_{zy}$ component while the values of the $\bar{F}_{xx}$, $\bar{F}_{yx}$, $\bar{F}_{zx}$ components of the macroscopic deformation gradient are solved to satisfy a plane-stress state, i.e. $\bar{\mathbf{P}}\mathbf{e}_x = \mathbf{0}$, where $\mathbf{e}_x$ is the unit vector along the x axis that is perpendicular to the y and z axes of Figure 5.4. The obtained engineering stress-strain curve is presented in Figure 5.16 with that of the studies in literature [11, 2, 61, 78]. The softening that begins at approximately 50% engineering strain in the experiment is not present in any model. Response of our model is quite comparable

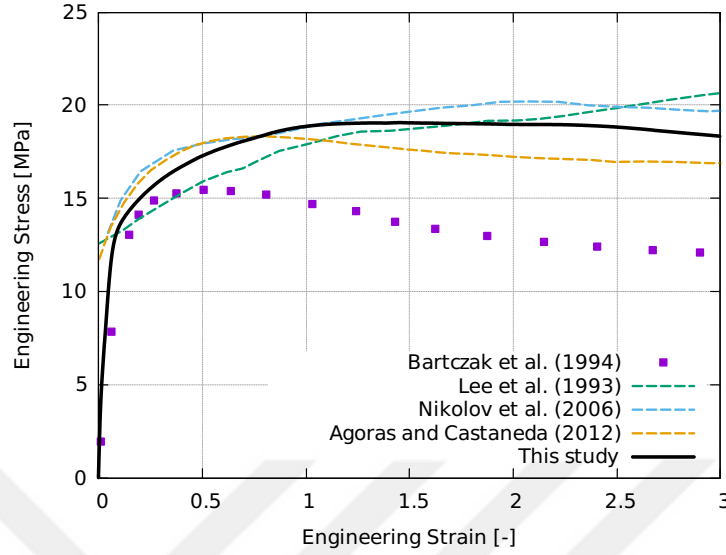


Figure 5.16: Comparison of the response of the model under simple shear with Lee et al. [61], Nikolov et al. [78], Agoras and Castaneda [2], and the experiment of Bartczak et al. [11]

with that of the other models, and especially in the elastic range it is more similar to that of the experiment. In the plastic range, Agoras and Castaneda [2] is the study that has the strongest qualitative similarity with the experiment, while Nikolov et al. [78] considers that the discrepancy may be associated with the difficulty to obtain uniform fields in the experiment.

Starting from the undeformed state in Figure 5.4, the evolution of the morphological texture is presented in Figure 5.17 with pole figures. According to the figure, the interface normals of the model align with the normal of the macroscopic shear plane, i.e. y axis, as expected. According to Figure 5.17, the rate of change of the interface orientations with the strain seems to be more similar to that of the experimental study, and slower than that of the other models [2, 61].

In Figure 5.18, crystallographic texture evolution is presented with pole figures. After 1.4 engineering strain Bartczak et al. [11] expresses that their pole figures clearly show a bi-modal distribution and therefore they explain the texture evolution by grouping crystals into two clusters, according to how their orientation changes. In the first cluster, (001) poles tend to orient at 5° away from the z direction, (010) poles

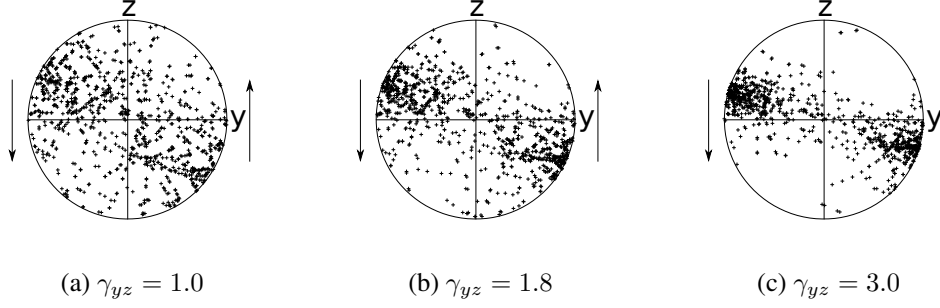


Figure 5.17: For simple shear, equal area projection pole figures of the interface normals that represent the evolution of the morphological texture, i.e. the orientation change of the crystalline lamellae. Arrows indicate the direction of shear.

spread around the x direction, and (100) poles are approximately along y axis. The second cluster also presents a similar orientation change, however less strongly and the maximum intensity of (001) poles orient at approximately 40° away from z direction at 1.4 engineering strain and continues approaching z direction at higher strains. As the strain increases, the first cluster grows, and at engineering strain 4.3 all crystals are considered to belong to the first cluster. Before comparing our results with the expressed observations of Bartczak et al. [11], it should be mentioned that our pole figures are obtained under load, following the other models [2, 61, 78], while the measurements of Bartczak et al. [11] are on the unloaded samples. The following observations from Figure 5.18 are particularly in line with the experimental observation. Notice the two maxima of (001) poles that are vaguely identifiable at 1.0 engineering strain, which are at z axis and at 45° degrees away from z axis on the $y - z$ plane, where the circular boundary of the pole figures correspond to the $y - z$ plane. As the strain increases, the second maximum approaches to the first. Since the a , b and c lattice axes of PE are orthogonal, and (010) poles of the second cluster are also close to the x axis as the first cluster, the second maximum present in (100) poles at 45° from y axis is also expected. Bartczak et al. [11] explains that while the maxima of (100) poles appears as a large and single block in their pole figures, their wide angle X-ray scattering patterns indicate that this is misleading. These blocks are actually composed of the superposition of two separate maxima, and their appearance as a single maximum is an experimental artifact.

We also find a third cluster that introduces a spurious texture not observed in the experiment, which is originally reported by Agoras and Castaneda [2]. In this cluster, (001) poles are along the x axis, and the (110) poles are aligned with the direction of maximum stretch 45° away from y axis in $y - z$ plane. Agoras and Castaneda [2] speculated that this cluster may be an artifact that is introduced with their approach of attempting to take the Coulomb-type hardening of the slip systems into account with a strain hardening law that depends on the accumulated slip rather than the pressure. In the following we will express that the reasoning of this speculation is rather similar to our initial considerations for the unexpected activity of $\{110\} \langle 1\bar{1}0 \rangle$ system in Sections 2.3.4.3 and 2.3.4.4, and also in Oktay and Gürses [79], before the three-dimensional spherulite model of Section 2.4 was studied. Coulomb-type hardening is an experimentally measured phenomenon in PE [10] and it is the increase of the resistance to slip with the increasing pressure on a slip plane. Among the models discussed here, including our model, only Agoras and Castaneda [2] attempted to take this into account. Speculation of Agoras and Castaneda [2] is according to the pole figures of Nikolov et al. [78], where the third cluster is not clearly visible. We consider that this is because Nikolov et al. [78] provides contour plots instead of the actual poles, and we see vague indications of the third cluster especially in (110) poles. Moreover, the third cluster may also be present in Lee et al. [61] according to the distribution of their (001) poles, while (110) poles are not presented. In Chapter 2, we have associated the unexpected activity of $\{110\} \langle 1\bar{1}0 \rangle$ slip activity with the neglected crystallographic deformation modes twinning and stress-induced martensitic transformation. We repeat here that, while the strain accommodated by the neglected deformation modes can be negligible, they result in drastic orientation changes when activated [13], and are considered to be more easily activated than the $\{110\} \langle 1\bar{1}0 \rangle$ slip system [10]. Therefore, these deformation modes may provide texture evolutions that are closer to the experiments, and the otherwise obtained spurious textures may also be associated with this. However, it should also be considered that among the plastic deformation modes of crystalline phase Bartczak et al. [11] only detected evidence on the activity of (100) [001], (100) [010] and (010) [001] slip systems. Moreover they have found an evidence that suggests the martensitic transformations can be excluded from the important deformation modes of HDPE for simple shear.

According to the pole figures of the compared studies [11, 2, 61, 78], the crystallographic texture evolution in our model progresses more slowly than the theoretical studies and at a more similar rate to that of the experimental study. According to this observation, the disk-like spherulite models homogenized with the statically equilibrated formulation, that has almost compatible amorphous-crystalline interfaces in static equilibrium, can also desirably slow down the texture evolution in the Taylor approach, which is known otherwise to provide too sharp textures [119].

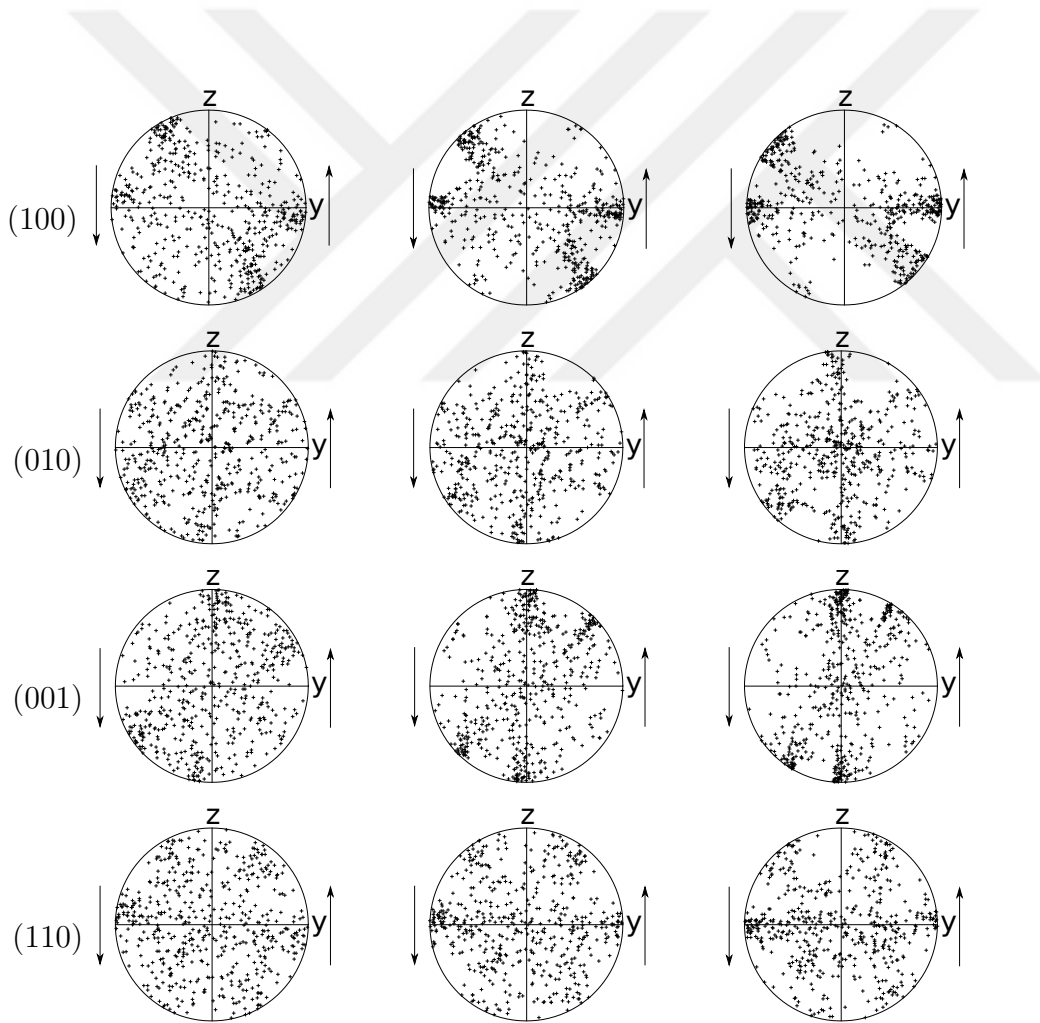


Figure 5.18: For simple shear, equal area projection pole figures of the lattice planes with normals along a , b , c and approximately $(0.5a + 0.8b)$ lattice axes, i.e. (100), (010), (001) and (110) poles, respectively, and for engineering strains 1.0, 1.8 and 3.0 that increase from left to right. Arrows indicate the direction of shear.

5.4 Conclusion

A homogenization-based constitutive model for semicrystalline polymers and its application to the HDPE are presented. The model is the Taylor-type homogenization of the randomly oriented disk-like spherulite models that are solved with the statically equilibrated formulation with bonded end interfaces in Section 3.4.2. As in Section 3.4.2, the interaction between the adjacent phases are considered with traction equilibrium on the interfaces where the kinematical compatibility is approximately satisfied. The neighborhood information is obtained from the PE spherulite morphology. The obtained results are in good agreement with the theoretical and experimental studies in the literature.

In simple shear, the interface normals align with the normal of the macroscopic slip plane with increasing strain. Therefore, as the deformation progresses, a greater percentage of the macroscopic shear force is in the interlamellar shear direction, and if the resistance to interlamellar shear remains constant, deformation by the interlamellar shear would require a lower macroscopic shear force. Agoras and Castaneda [2] points out to the favorable alignment of the lamellae for interlamellar shear and that it would result in a macroscopic softening in large strains, since the interlamellar shear is the most easily activated deformation mode of the semicrystalline polymers. However, recall that the macroscopic softening that begins approximately at 50% engineering strain is not captured by any model in literature including the present study. Considering the favorable alignment of the lamellae for interlamellar shear is present in all models, it could be stated for the models that the rate of increase of the resistance to interlamellar shear overcomes the expected softening. Agoras and Castaneda [2] criticizes the widely employed approach of taking the critical resolved shear stresses as constant in crystalline phase and taking the locking stretch of the amorphous phase as the primary factor for the macroscopic hardening in large strains, and argue that the strong macroscopic hardening in large strains under simple compression is primarily due to the hardening of the crystalline phase. Therefore, taking the Coulomb-type hardening of the crystalline phase into account in the models may lead to an amorphous phase constitutive model with a less strong strain hardening response, and this might result in the desired softening in simple shear. It should also be stated that the

experimental simple shear stress-strain curve of HDPE provided by G'Sell et al. [36] presents a less strong softening. Since the aim of Bartczak et al. [11] is to investigate the active plastic deformation processes, the dimensions of their samples were subjected to the requirements of the X-ray measurements, and Bartczak et al. [11] could not follow the dimensional guidelines suggested by G'Sell et al. [36] in the sample the stress-strain response of which is presented. These guidelines are proposed to minimize the influence of the inhomogeneous deformation near the ends of the sample, and Nikolov et al. [78] considers that the difficulty to obtain uniform fields in the simple shear experiments may have a part in the discrepancy between the experimental results and model predictions.

According to Section 3.6.7, in simple shear, there were inconsistencies on the slip system activities between the statically equilibrated disk-like spherulite models and the expectations of the experimental study of Bartczak et al. [11]. Since we employ the same disk-like models in this section, but with a different degree of crystallinity and another amorphous phase material model, we have investigated the texture evolution of the constitutive model in greater detail under simple shear. The obtained textures are in a good agreement with Bartczak et al. [11] and also with the models in literature [2, 61, 78]. Therefore, the inconsistency could be considered to disappear under a more general loading state. The relatively soft response and the slip system activities that are localized into only a few slices in Section 3.6.7 reinforces the idea that, the certain slip systems of crystal plasticity constitutive models are favorably aligned to provide easy plastic deformations in certain slices of the disk-like model in the ideal simple shear deformation state. The uniform strain assumption especially along the radius of the model could not prevent this condition. On the other hand, it seems logical to think that the strong kinematical requirements of the Taylor approach on the randomly oriented models can prevent such mechanisms to escalate, and provides the essential response expected from the disk-like spherulite models.

In simple shear, the similarity between the pole figures of our study and Agoras and Castaneda [2] does not support their speculation on the reason of the spurious texture they obtain. They express that their employment of the Voce-type strain hardening law instead of the actual Coulomb-type hardening of slip systems may be the reason of the spurious texture. Voce-type strain hardening employed by Agoras and Cas-

taneda [2] depends on the amount of accumulated slip and not on the pressure on the slip plane, while the plastic hardening of the semicrystalline polymer crystals, that is expressed by a Coulomb-type hardening law, actually depends on the pressure instead of the accumulated slip. Agoras and Castaneda [2] has taken this approach since their model requires an associated flow rule. Nevertheless, we obtain a similar spurious texture without considering any hardening of the critical resolved shear stresses of the crystalline phase and we consider that the spurious texture may also be present in other models referred to in this chapter. In our opinion, the implications of taking crystallographic slip as the only plastic deformation mode of the crystalline phase are not widely known among the modeling community. Although Bartczak et al. [11] reports that they only detected the activity of $(100) [001]$, $(100) [010]$ and $(010) [001]$ slip systems among the deformation modes of the crystalline phase, we think that incorporation of the neglected deformation modes of the crystalline phase in the models may provide more consistent texture evolutions with the experiments. The importance of the other deformation modes, twinning and stress-induced martensitic phase transformation, have long been appreciated by the experimental studies [10], and the drastic orientation change with their activation [13] is widely employed to explain the texture evolution. Therefore, in order to narrow down the gap between the experimental and modeling studies on this subject, more models that point out the drawbacks of this simplification should be published, including our three-dimensional spherulite model in Section 2.4.



CHAPTER 6

UHMWPE COMPONENTS EMPLOYED IN KNEE IMPLANTS

Ultra high molecular weight polyethylene (UHMWPE) is also composed of chains of the same monomer as HDPE, ethylene. Compositionally, the only difference between the HDPE and UHMWPE is the length of the polymer chains, which are considerably longer than that of the HDPE. Molecular weight of the chains varies in ranges $0.05\text{-}0.25 \times 10^6$ g/mol and $2\text{-}6 \times 10^6$ g/mol in HDPE and UHMWPE, respectively. While spherulitic morphology is widely observed in HDPE, the characteristic morphology of UHMWPE is randomly oriented lamellar stacks [97], since the molecular mobility of the long chains of UHMWPE is not generally sufficient to form the complex spherulite morphology [115]. Similarly, the degree of crystallinity of UHMWPE is lower than HDPE, and varies in the range 39-75%, while the crystallinity ratio of HDPE is in 60-80% range. In this section, employment of the UHMWPE in total knee replacement designs is reviewed and a FEM study on the surface texture of UHMWPE to decrease the friction coefficient is presented. The analyses employ Bergstrom Bischoff [16] as the constitutive model of the UHMWPE. Additionally, an attempt to adapt the micromechanical constitutive model presented in Chapter 5 to the UHMWPE is also presented.

6.1 UHMWPE Components in Knee Implants

Knee is one of the most complex joints in the body, which enables complex rotation and sliding between the joint surfaces up to 140° , and extends 5 mm while not under load. The number of degrees of freedom required to model all rotation and sliding movements of the knee is six. UHMWPE has been employed in knee implants

since the end of 1960s. Before employment of UHMWPE, knee implants employed monolithic designs, where the femoral and tibial components are connected with a pin that only enables the rotation of the knee around a single axis. Restriction of the natural movement of the knee in these designs lead to high stress concentrations on the bones. Therefore, the kinematically restricted designs, with their short lifetime and high complication rates, are not preferred today.

Implant designs rapidly advanced in 1970s, with the employment of UHMWPE. In modern designs, parts of the femoral and tibial bones that face towards the knee are removed, instead cobalt-chromium femoral and tibial components are attached with bone cement. In between these two components a UHMWPE tibial insert is placed. The tibial insert is generally bonded to the surface the tibial component, however there are also designs that do not employ that bond. There is no bonding between the tibial insert and the femoral component. The three components of the knee implant, that may not be attached, works under the confinement of the surrounding natural tissues. These designs, do not considerably restrain the movement of the knee in daily use, and the placement errors during surgeries are tolerated with the soft tibial insert that could also wear. On the other hand, for a long service life, the tibial insert should provide high resistance against impacts and abrasion. UHMWPE has a high toughness, a low friction coefficient, and a high resistance to chemical reactions. Therefore, the tibial insert is resistant against impact loads and wear, and is bio-compatible. Thus, the UHMWPE is the golden standard in modern knee implants. In Figure 6.1, radiographs of a knee with a modern implant are presented, where the tibial and femoral components are clearly visible, while the tibial insert is not radio-opaque.

Total knee replacement and hip implants are among the most frequently employed joint replacement operations. These are generally successful operations, and 90% of the patients could use the implants for more than 10 years. The average lifetime is 15 years, which is limited by the wear and fatigue damage on the UHMWPE. Due to the size of the knee, the thickness of the tibial insert is limited, and while the wear rate of UHMWPE is low, it would eventually wear out. Moreover, because of the small size of wear particles, the particles interact with the bones, and osteolysis, the weakening of the bone density occurs [48]. Osteolysis loosens the connection between implant

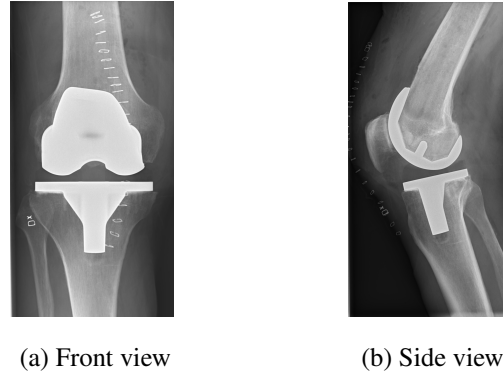


Figure 6.1: Radiographs of a knee with a modern implant adopted from [38] and [39], respectively

and the bone. In order for the tibial insert not to become too thin through wearing and the amount of wear particles to be lower than the threshold of side effects, the wear resistance of the UHMWPE should be increased.

6.1.1 Major Surface Damage Modes of the Tibial Insert

Types of damages observed in the retrieved tibial inserts from patients are grouped into seven modes [46]. i) Pitting is the formation of the pits with 2-3 mm width and 1-2 mm depth, which is a result of fatigue damage. The wear particles of pitting are too large to interact with bones. Therefore, this damage mode is relatively harmless. ii) A bone or metal particles could embed into the tibial insert and cause embedded debris type damage. These particles increase the wear of the cobalt-chromium components of the implant. In turn, the worn components increase the wear rate of the tibial insert. iii) Scratching is a wear type damage mode, where the irregularities of the femoral component scratches the surface of the UHMWPE component. iv) Delamination is a fatigue failure and a large body of UHMWPE surface layer is removed. If the tibial insert is thick it may remain functional, if it is thin or became rigid due to oxidation then it becomes inoperable. v) Plastic surface deformations belong to the surface deformation damage mode. vi) Burnishing is a damage mode associated with wear and abrasion, and distinguished as a polished surface due to wear. The wear-particles of this damage mode is very small and can interact with the bones. vii) Abrasion is a

wear damage mode, where the surface is locally worn out.

6.1.2 Approaches for Improving the Performance of Total Joint Replacements

Cross-linking, optimization of the design geometry, and surface texturing are among the techniques that are considered to improve the performance of UHMWPE in total joint replacements.

6.1.2.1 Cross-linking

When the UHMWPE is subjected to radiation, the radiation energy forms strong covalent bonds between the nearby chains of the amorphous phase, which is found to significantly increase the wear resistance. For instance, according to hip simulator experiments [54], the wear of the UHMWPE components in the hip replacements are decreased by 90% when a radiation dose of 100 kGy is employed. However, the UHMWPE becomes more brittle with this method. Therefore, cross-linking is not rapidly adopted in knee implants where the impact resistance is important as it is employed in hip implants where the contact area is wider and the impact resistance has a lower priority. Another complication of the cross-linking is that it forms free-radicals in UHMWPE, which increase the rate of reaction of UHMWPE with oxygen, and decreases the durability. This phenomenon is called post-irradiation aging. Research on cross-linking is an ongoing effort and there are commercial products as well. Heat treatment by melting or annealing is proposed against the free radicals and also the decrease in ductility. Additionally, bio-compatible stabilizers such as vitamin-E are incorporated into UHMWPE to consume the free-radicals. In a network of over 100 hospitals in United States, 57% of the implanted knee implants had a cross-linked UHMWPE in 2003 [53].

6.1.2.2 Shape Optimizations

The next approach to decrease the wear of the tibial insert is the optimizations with the changes in the implant shapes. The shape of the tibial insert could be adjusted to

increase the contact area with the femoral component. The femoral component both slides and rotates on the tibial insert. Additionally, it could twist on the axis along the normal component of the tibial insert surface. Moreover, sliding and rotation directions vary throughout the gait. Patten et al. [84] experimentally measured the wear amount of UHMWPE, on which a semi-sphere is slid and rolled repeatedly in certain combinations. The study reports that application of different movement types together has a significant effect on the wear rate, and the major effect is due to sliding. There are different positions that could be optimized for a higher conformation, and an increase in the conformity for certain position could negatively affect the conformity of the others. Additionally, the tibial insert could be fixed to the tibial component (fixed bearing) or not (mobile bearing).

It is theoretically clear that, as the contact forces decrease the wear rate decreases as well. According to the size and geometry of the knee that enable complex motions, the contact area is significantly smaller than that of the hip. It is also known that the maximum von Mises stresses on the tibial insert are higher than the yield stress of UHMWPE. Nevertheless, clinical findings indicate that the long term success does not only depend on the amount of contact stress on the tibial insert. For example both Miller-Gallante (MG) and Insall-Burstein (IB) are considered as clinically successful tibial insert designs [15, 103], while the von Mises stresses in MG are 20% higher than the IB design [55]. MG is relatively flat, and therefore enables the movement of femoral component relatively freely on the tibial insert. IB, however, has curves and conforms well with the femoral component and therefore restricts the movement. Therefore, implant loosening is relatively less observed in MG due to the free movement [15], and the wear of the tibial insert, with serious damage modes such as delamination is less frequently observed in IB [103] due to the lower contact stresses. In order to mitigate the higher constraints of IB design, mobile bearings are proposed. Improvements on the long term performance of knee implants with the geometrical enhancements are difficult to assess with simulations only, according to the described complex knee movement, wear and loosening mechanisms.

6.1.2.3 Surface Texturing

The third technique that could be employed to decrease the wear is texturing the surface of UHMWPE. Application of micro textures composed of small pits or grooves on the contact surfaces of machine elements is known to decrease the friction coefficient and wear rates [52]. Surface textures are present in natural and man-made objects, and are known to be beneficial. For example, linear grooves on the shark skin decrease the water resistance, and pits on golf balls increase the aerodynamic performance. Surface textures on UHMWPE are also investigated in literature to decrease the friction coefficient. Applicability of this approach with implant designs is a subject of ongoing study. Application of the surface texture on the metal components or the tibial insert, and their influences on the friction coefficient and wear rate are investigated in literature under dry or wet contact conditions. Cervantes et al. [66] indicates that pits on tibial insert could significantly decrease the friction coefficient of UHMWPE for an individual who weights 70 kg, based on their experimental study with oil lubrication. Zhang et al. [120] studied whether the surface texture should be on the metal components. It is found that when the texture is on the UHMWPE, the wear rate and friction coefficient decreases more under water lubrication. Ripoll et al. [88] modeled the dry contact between a textured UHMWPE and titanium with a FEM model. They found that surface texture significantly affects the friction, and based on plastic strains obtained with textured surfaces, they concluded that textured surfaces would have shorter lifetimes. Therefore, they suggested filling the pits with materials that have similar elastic properties with UHMWPE. Kustandi et al. [57] studied the dependence of the friction coefficient between UHMWPE and a silicone-nitrate sphere on trench shaped textures on UHMWPE, experimentally under dry contact conditions. They reported improvement on the wear resistance and friction coefficient. Sawano et al. [93] experimentally studied the friction coefficient between UHMWPE and a textured metal surface under water lubrication. They report that the texture decreases wear rate, and associate the improvement with micropits on metal surface traps the worn out metal particles, which may have otherwise deeply scratch the UHMWPE surface.

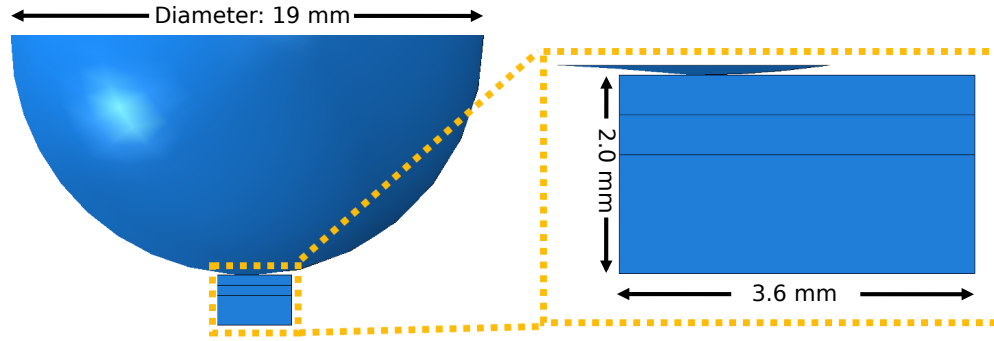


Figure 6.2: The rigid sphere, the UHMWPE part and their dimensions

6.2 The Effect of Surface Texture on the Friction Coefficient of UHMWPE

Since the femoral component that slides and rotates on the tibial insert has a certain curvature, in this study the contact of a curved surface with UHMWPE is considered. This is motivated by the experimental study of Cervantes et al. [66]. A rigid sphere of 19 mm diameter is placed on a 2 mm thick UHMWPE part as presented in Figure 6.2. In the FE analyses, the three—network model of Bergsrom and Bischoff [16] is employed as the constitutive model of UHMWPE with the associated material parameters for their sterilized sample from GUR 1050 resin, and an attempt to adapt the constitutive model developed in Chapter 5 to UHMWPE is presented in Section 6.3.

The interaction between the sphere and the UHMWPE is solved with contact pairs of Abaqus FEM software, with a hard-pressure overclosure relationship and the Coulomb friction law for sliding. The friction coefficient is taken as 0.1 [22, 86]. The analysis is composed of two steps that are schematically presented in Figure 6.3. In the first step, a 17 N vertical load is applied on the centroid of the sphere in 1 s. In the second step, the sphere is horizontally moved with a 50 mm/s speed with the applied constant vertical force. The diameter of the sphere and the magnitude of the vertical force are based on the experiments of Cervantes et al. [66].

A square control region with 1.2 mm edge-length is considered on the UHMWPE surface as presented in Figure 6.4. The first point of contact between the sphere and the UHMWPE part in Step 1 is shown in Figure 6.4 as the starting point, and the location

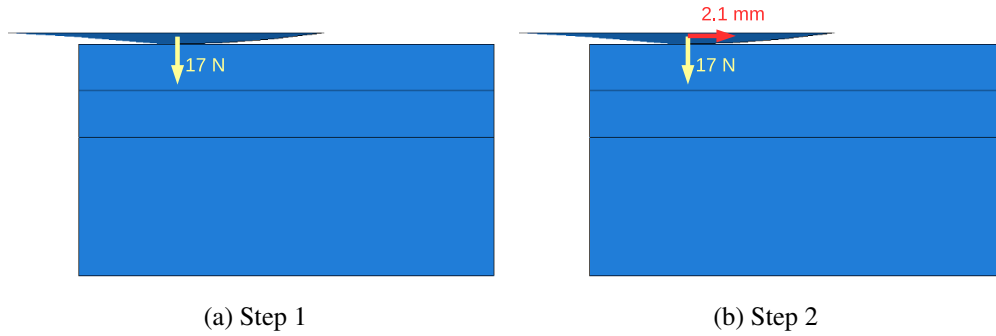


Figure 6.3: Steps of the load application the analyses

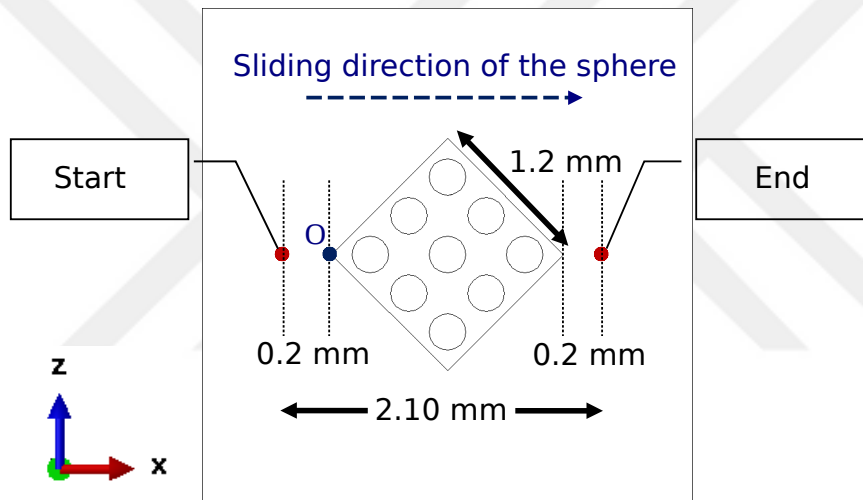


Figure 6.4: The control surface on the UHMWPE surface. The initial point of contact of the sphere is marked as the start point

of the centroid of the sphere at the end of the second step are also presented. Surface textures are obtained by considering empty cylindrical dimples with 0.4 mm depth. Dimples are distributed into the control region with the patterns in Figure 6.5. In each dimple pattern the uniformly distributed dimples cover the 34.9% of the surface area of the control surface. The diameter of the dimples and the shortest distance between them are given in Table 6.1, where the dimple depth is constant among the patterns and equal to 0.4 mm.

The boundary conditions of the UHMWPE part are presented in Figure 6.6. FE nodes are fixed at the bottom surface, while the displacement components along the surface normals are zero on the side surfaces.

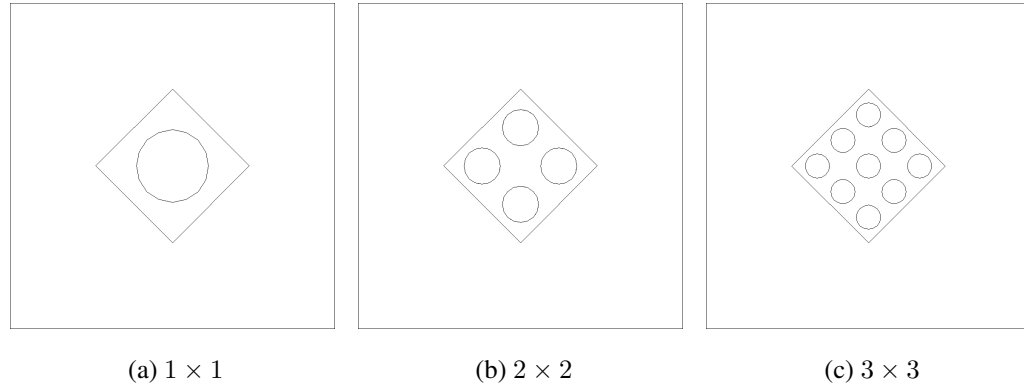


Figure 6.5: Dimple patterns

Table 6.1: Diameter of the dimples and the shortest distance in-between in mm for the studied dimple patterns

Pattern	Dimple diameter (mm)	Shortest distance (mm)
3x3	0.266	0.133
2x2	0.4	0.2
1x1	0.8	0.4

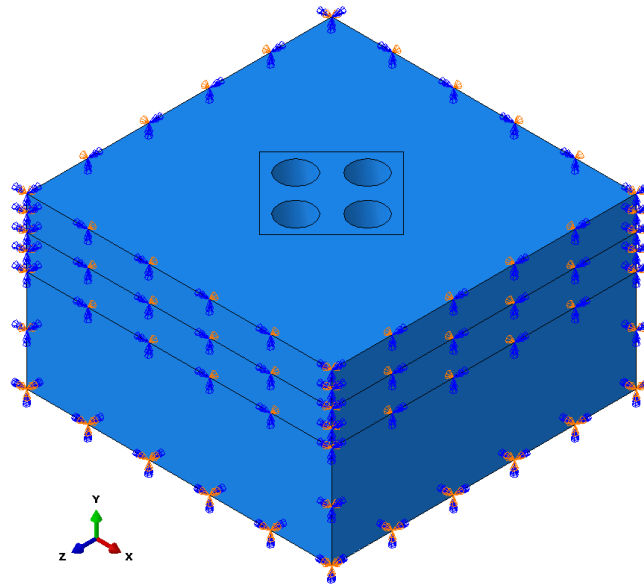


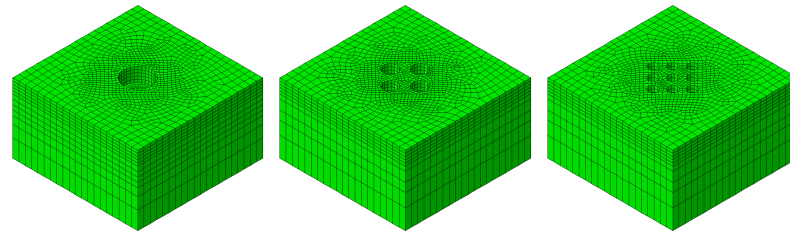
Figure 6.6: Boundary conditions of the UHMWPE part, where the presented conditions are enforced to all FE nodes on the indicated surfaces

6.2.1 Mesh Sensitivity

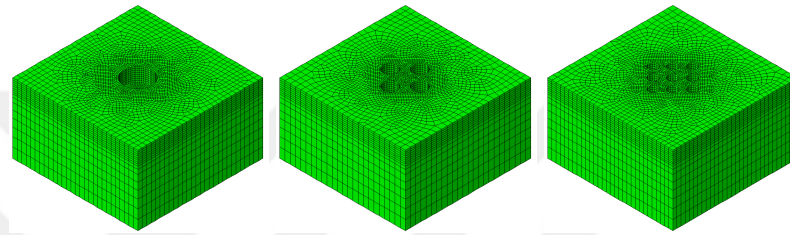
A mesh sensitivity study is conducted for the dimple patterns with the meshes presented in Figure 6.7, and the results are presented in Figure 6.8 in terms of the friction coefficient along the sliding path of the sphere. The friction coefficient is found by dividing the horizontal force by the 17 N vertical force on the sphere. Normal and fine meshes are composed of approximately 13,000 and 40,000 C3D8IH elements, respectively from the element library of Abaqus. In the fine meshes, the edge-lengths of the elements on the surface near the dimples are approximately a half of that of the normal meshes. C3D8IH is a 8-noded fully-integrated linear brick element that has 8 integration points, where the letter 'I' in stands for the incompatibility mode, which means that the element is less vulnerable to shear locking numerical problem common among the fully-integrated linear elements. Finally, 'H' stands for the hybrid formulation, that avoids problems when approximately incompressible constitutive models are employed. According to the results, the normal meshes are sufficiently fine. Origin of the horizontal axis in Figure 6.8 and the similar figures that follow corresponds to the origin of the control surface marked as point O in Figure 6.4.

For each dimple pattern, another set of FE meshes are obtained by filling the dimples of the meshes in Figure 6.7 with additional finite elements. The obtained meshes are employed to model the smooth UHMWPE and the cases that consider the dimples are filled with another material. The obtained results with these meshes, where the UHMWPE constitutive model are also employed in the new elements, are presented in Figure 6.9. In the figure, the arithmetic average of the curves are also presented, which is taken as the representative friction coefficient curve of the the smooth i.e. texture-free UHMWPE surface.

An average friction coefficient is defined for the control surface. The centroid of the sphere is on the control surface for the $[0, 1.7]$ mm range in Figure 6.9. The average friction coefficient is the area under a friction coefficient curve in this range, divided by the length of the sliding path in the control surface, 1.7 mm. The value of the average friction coefficient is 0.1062 for the smooth surface and is also presented in Figure 6.9.



(a) Normal



(b) Fine

Figure 6.7: FEM meshes of the dimple patterns

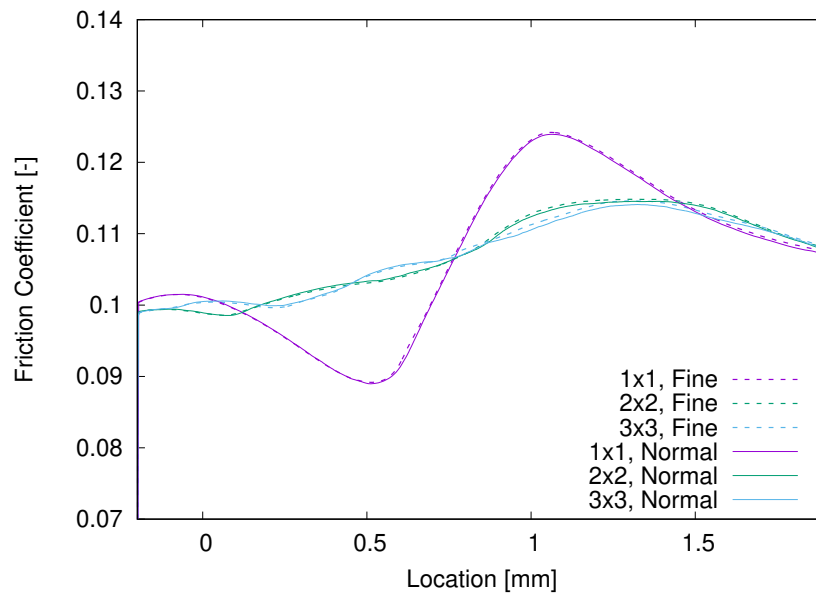


Figure 6.8: Variation of the friction coefficient along the sliding path for different dimple patterns and meshes

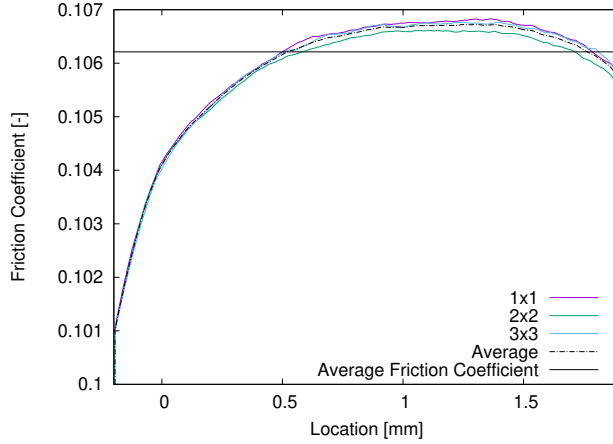


Figure 6.9: Variation of the friction coefficient along the sliding path for the smooth, i.e. texture-free, surface with different meshes of normal fineness

6.2.2 Surface Textures Composed of Empty Dimples

Variation of the friction coefficient of 1×1 dimple pattern along the sliding path is compared with that of the smooth surface, and beginning, centroid and end points of the dimple are marked in Figure 6.10, which are at positions 0.45, 0.85, and 1.25 mm, respectively. The friction coefficient decreases while approaching the dimple, and then increases steadily almost until the end of the dimple reached. Finally, when leaving the control surface, the friction coefficient approaches to that of the texture-free surface. The magnitude of the increase in the friction coefficient is higher than the obtained reduction.

The reason of the undulation in Figure 6.10 could be explained as follows. In Figure 6.11, schematic views of an elastic solid that is deformed with the contact of a rigid sphere is presented. The forces acting on the sphere are the vertical force F , and the normal components of the contact forces, where the tangential contact forces are neglected. The elastic solid is divided into two imaginary half-spaces by a vertical plane passing through the centroid of the sphere. If both half spaces are identical as in Figure 6.11a, the sphere would be in equilibrium and would not move. If region B is elastically softer than the region A , the sphere would move towards the region B since the forces in region B are smaller than the reaction of the region A that pushes the sphere towards region B . The region B could also be considered as a void that

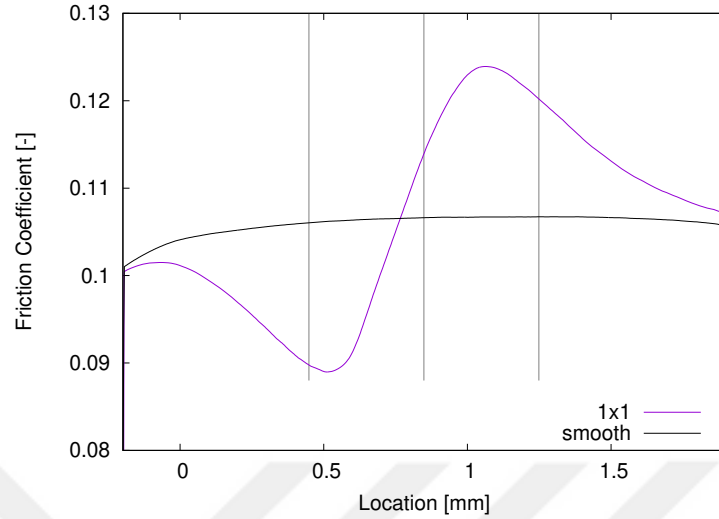


Figure 6.10: Variation of the friction coefficient along the sliding path for the 1×1 dimple pattern

does not apply a force on the sphere, and this explains the decrease of the friction coefficient when approaching the dimple. The increase in the friction coefficient that follows the decrease is simply the opposite, moving from a soft region towards a stiff region.

The contact forces could be decomposed into the tangential and normal forces with respect to the normal vector of the infinitesimal surface of contact. In the FE analyses, while the normal components are governed by a pressure-overclosure relationship, the tangential components are governed by a pressure-friction relationship. Normal contact forces on the surface has non-zero components along the sliding direction. In Figure 6.12, the influence of the tangential and normal contact forces on the friction coefficient are presented separately for 1×1 dimple pattern, where the friction coefficient is the sliding direction component of the net force divided by the vertical force as presented in the previous figures. According to 0.1 value of the friction coefficient of the Coulomb friction law, and the employed constitutive model of UHMWPE, the influence of the normal contact forces on the friction coefficient is in the order of 20%, which changes sign along the sliding path to push the sphere towards the empty dimple. The results in Figure 6.12 are in line with the explanation with the normal contact forces, given above.

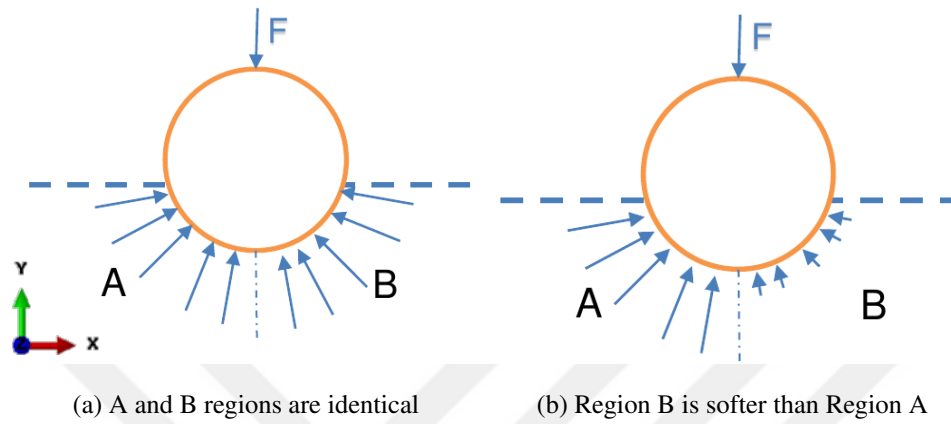


Figure 6.11: Schematic view of a solid, deformed by a sphere

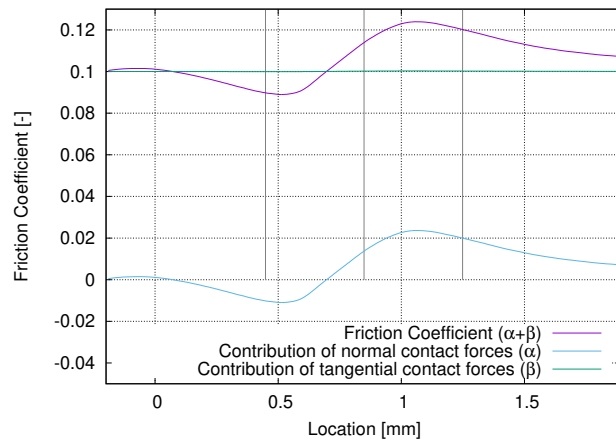


Figure 6.12: Contribution of the tangential and normal contact forces on the computed friction coefficient for 1×1 dimple pattern

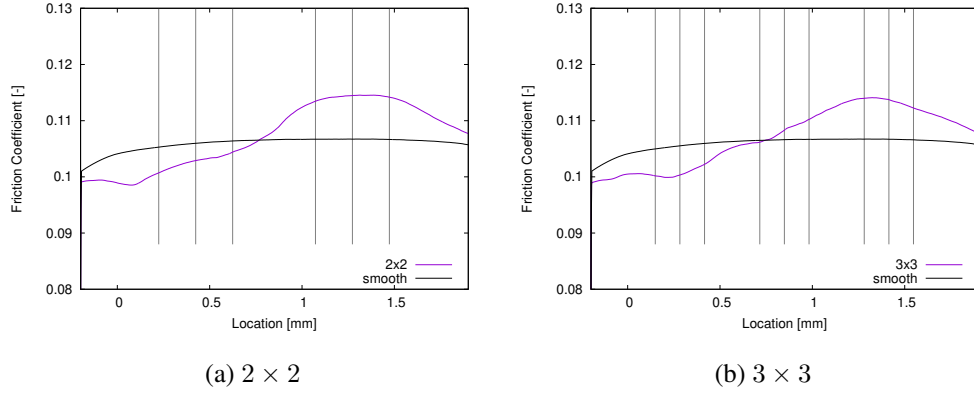


Figure 6.13: Variation of the friction coefficient along sliding path

In Figure 6.13, the friction coefficient along the sliding path is presented for 2×2 and 3×3 dimple patterns, where results of the smooth surface are also present for comparison. The start, middle and end points of the dimples are also indicated in the figures. While the curves in Figure 6.13 undulate as well, it is difficult to identify the contribution of the individual as in the experiments of Meine et al. [70] where multiple dimples are in contact with the sphere.

In Figure 6.14a, variations of the vertical displacement at the centroid of the sphere along the sliding path are presented for the studied dimple patterns. Recall that the total surface area of the dimples are the same among all dimple patterns. As the number of dimples increases the vertical displacements decreases. In Figure 6.14b, average friction coefficients of the control surface for different dimple patterns are compared with that of the smooth case, and the smooth case has the lowest average friction coefficient.

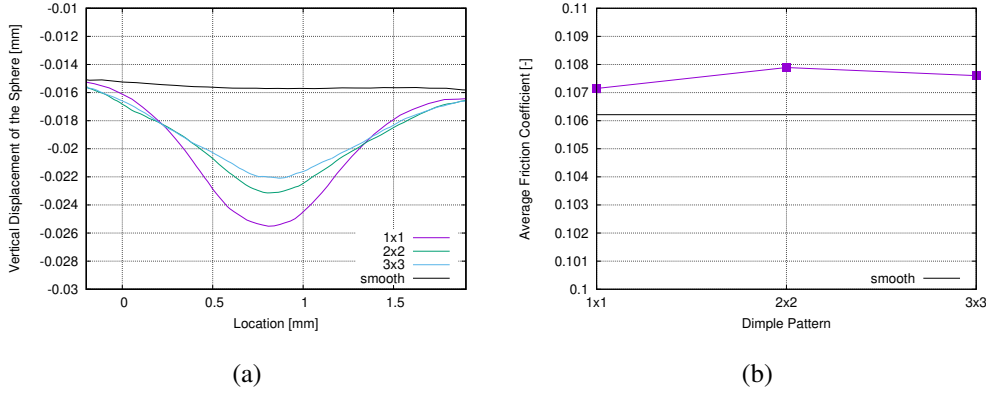


Figure 6.14: For the studied dimple patterns, (a) vertical displacement of the centroid of the sphere along the sliding path, (b) average friction coefficient of the control surface

6.2.3 Influence of the Aspect Ratio of the Dimples and Sliding Speed

As presented in Figure 6.15, dimples with different aspect ratios are studied by changing the depth of the dimples. Friction coefficients for different dimple patterns and diameter to depth (D/d) aspect ratios are presented in Figure 6.16. The aspect ratio does not substantially change the shape of friction coefficient curve along the sliding path. The average friction coefficient approaches to that of the smooth case when the dimple depth is decreased. Response of the UHMWPE constitutive model of Bergsrom and Bischoff [16] is rate dependent. Influence of the sliding speed on the friction coefficient is found as negligible according to Figure 6.17. This is expected according to the results presented in Bergsrom and Bischoff [16].

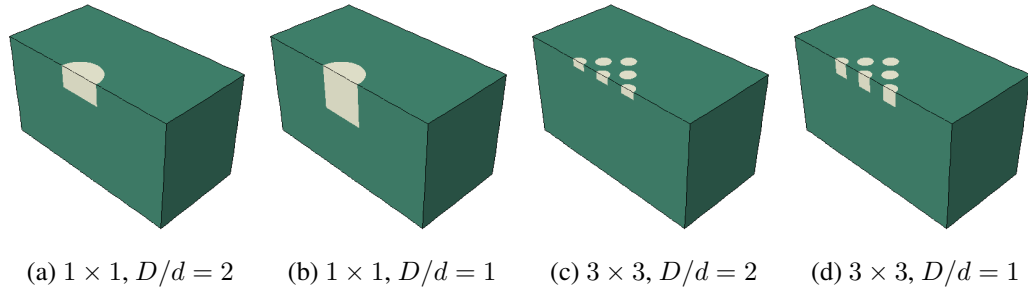


Figure 6.15: Views of the cross-sections of the model with different dimple patterns and aspect ratios. Empty dimples are drawn in a different color for visualization.

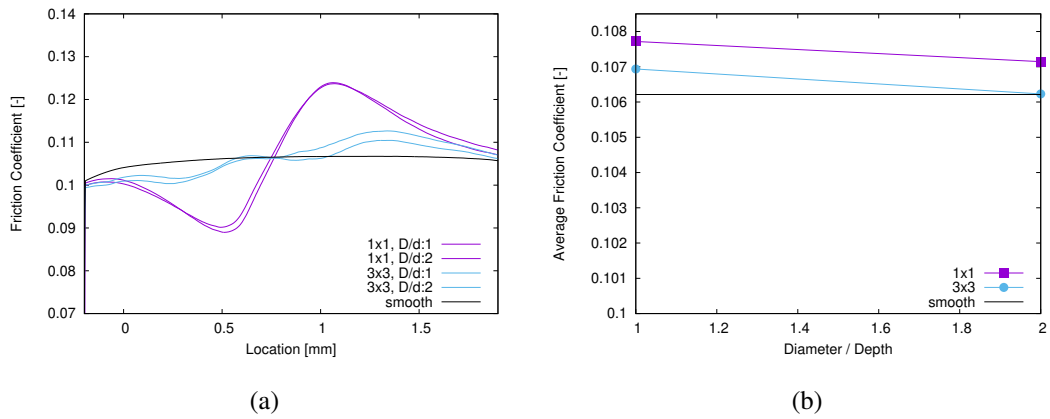


Figure 6.16: For the different dimple patterns and aspect ratios (a) the variation of the friction coefficient along the sliding path (b) the average friction coefficient in the control surface

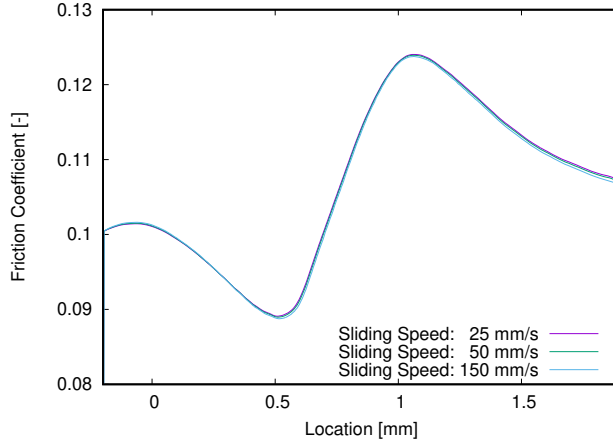
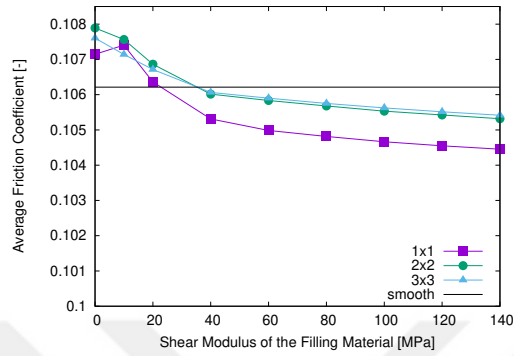


Figure 6.17: The influence of the sliding speed of the sphere on the friction coefficient along the sliding path

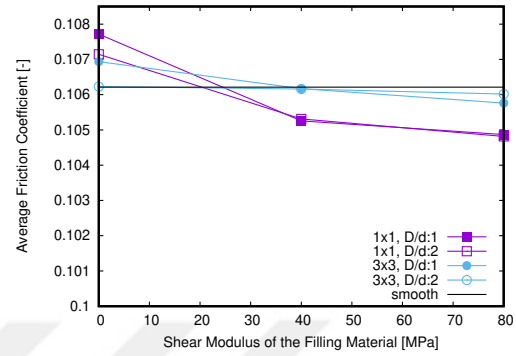
6.2.4 Surface Texture Composed of Dimples Filled with a Neo-Hookean Solid

Dimples are filled with new elements that employ the compressible Neo-Hookean hyperelasticity from Abaqus constitutive model library. The constitutive model of the other elements of the mesh are not altered. The bulk modulus of the filling material is taken as 2200 MPa. The friction coefficient is uniformly taken as 0.1 throughout the model surface. Variation of the average friction coefficient with the shear modulus of the filling material is presented in Figure 6.18a, where the zero value in x axis corresponds to the empty dimples. According to the figure, for all dimple patterns, a smaller average friction coefficient could be obtained compared to the uniform case, where the smallest is obtained with 1×1 dimple pattern.

Variations of the average friction coefficient with the shear modulus of the filling material are presented for different dimple aspect ratios and patterns in Figure 6.18b. The dimple aspect ratio is as defined in Section 6.2.3, and the zero value of the x axis corresponds to the empty dimples. For the filled dimples, influence of the aspect ratio is low, and 1×1 dimple pattern still provides the lowest average friction coefficient. Variation of the friction coefficient along the sliding path with the dimple aspect ratio is presented in Figure 6.19 for 1×1 and 3×3 dimple patterns, where the shear and bulk moduli of the filling material are 40 MPa and 2200 MPa, respectively. As the aspect ratio increases the undulations diminish, desirably.

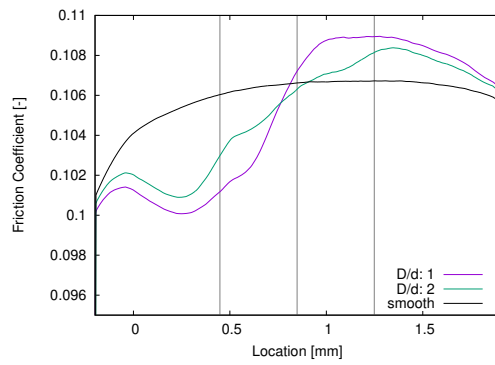


(a) For a 0.4 mm dimple depth

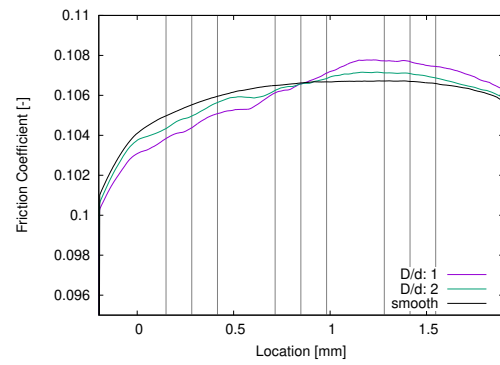


(b) For different D/d dimple aspect ratios

Figure 6.18: Variation of average friction coefficient with shear modulus of the filling material

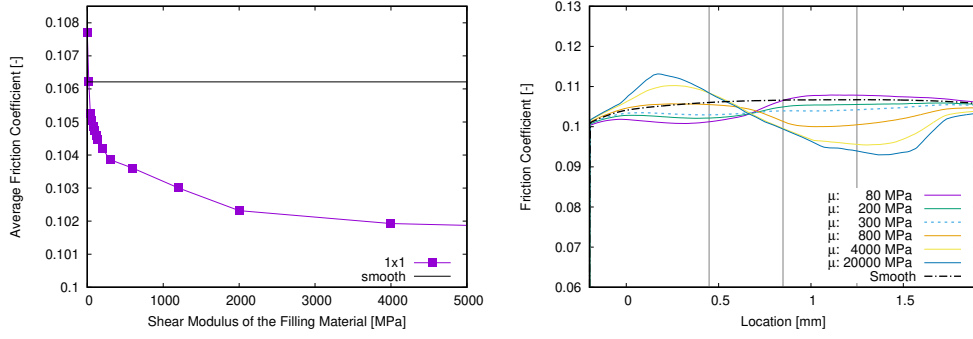


(a) 1×1



(b) 3×3

Figure 6.19: Variation of the friction coefficient along the sliding path



(a) Average friction coefficient (b) Variation of friction coefficient along the sliding path

Figure 6.20: Variation of friction coefficients with the shear modulus of the filling material

In the following, only 1×1 dimple pattern is studied, where the diameter to thickness ratio is taken as one. The variation of the friction coefficients with the shear modulus of the filling material are presented in Figure 6.20, where the bulk modulus of the filling material is taken as 2200 MPa. The average friction coefficient decreases with the increasing shear modulus, and an approximately 5% decrease is obtained compared to the friction coefficient of the smooth surface. The shear modulus of 150 MPa is a turning point for the response. With this value, the undulations of the friction coefficient curve diminishes, and the friction coefficient is below that of the smooth surface at all locations. Beyond 150 MPa, dimples behave as the stiff regions, and probably due to the geometry of the dimples, the elastic potential energy stored in the dimples begins to push the sphere towards the sliding direction even before the middle of the dimple is reached. Therefore, the shear modulus of the filling material may play an important role to decrease the friction coefficient.

The variation of friction coefficients with the bulk modulus of the filling material is presented in Figure 6.21, where the shear modulus of the filling material is taken as 40 MPa. The influence of the bulk modulus on the average friction coefficient is found as negligible, and the undulations of the friction coefficient along the sliding path decreases with increasing bulk modulus.

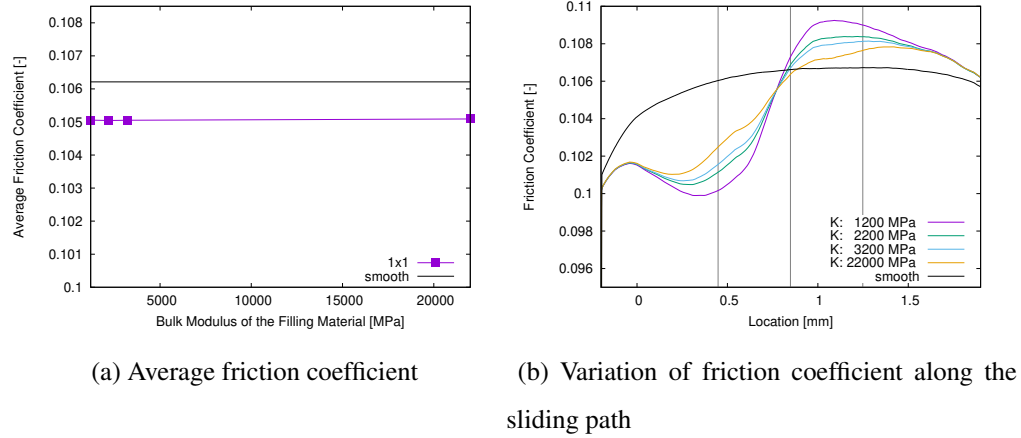
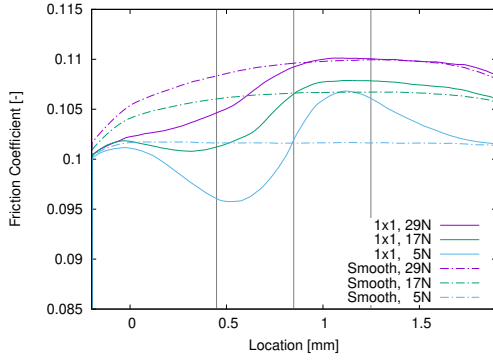


Figure 6.21: Variation of friction coefficients with the bulk modulus of the filling material

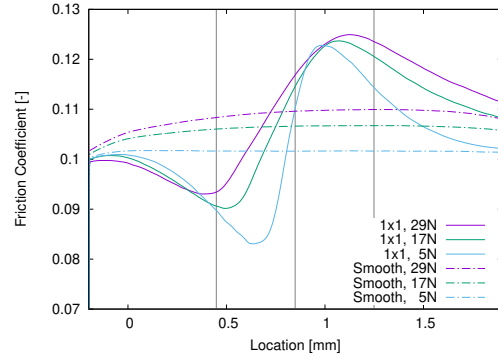
6.2.5 Influence of the Vertical Force

Variation of the friction coefficient with the vertical force acting on the sphere is presented in Figure 6.22 both for the filled and empty dimples, where only 1×1 dimple pattern with $D/d = 1$ is employed. The bulk and shear moduli of the Neo-Hookean filling material are taken as 2200 MPa and 40 MPa, respectively. The corresponding curves of the smooth surface are also presented in Figure 6.22. As the vertical force decreases, the friction coefficient of the smooth surfaces approaches to the value of 0.1, which indicates that the contribution of the normal contact forces on the friction coefficient decreases with the decreasing vertical force, as expected. As the vertical force increases, the sphere moves more easily on the control surfaces with filled dimples, compared to the smooth case. Therefore, the improvement of the dimples on the average friction coefficient would increase with increasing vertical force. For the empty dimples, considerable undulation of the friction coefficient persists with the different values of the vertical force.

The variation of the vertical displacement along the sliding path is presented in Figure 6.23 for different vertical forces on the sphere. For the filled dimples, as the vertical force increases, the variation of the vertical displacement along the sliding path decreases and the location of the highest displacement shifts from center to the beginning of the dimple. As for the empty dimples, the amount of vertical displace-



(a) Dimples filled with the Neo-Hookean solid



(b) Empty dimples

Figure 6.22: The influence of the vertical force on the sphere on the friction coefficient along the sliding path

ment almost linearly increases with the vertical force.

The variation of the average friction coefficient with the vertical force is presented in Figure 6.24. The average friction coefficient increases non-linearly with the vertical force, and the filled dimples could decrease the friction coefficient approximately 1% compared to the smooth surface.

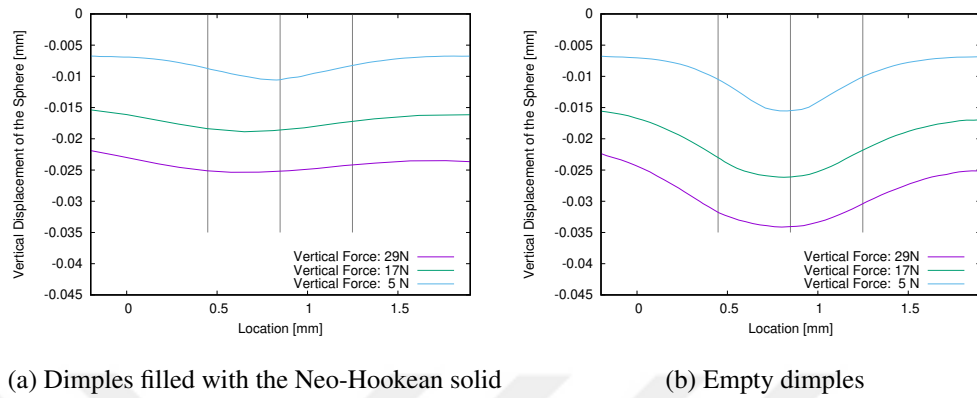


Figure 6.23: The influence of the vertical force on the vertical displacement of the centroid of the sphere along the sliding path

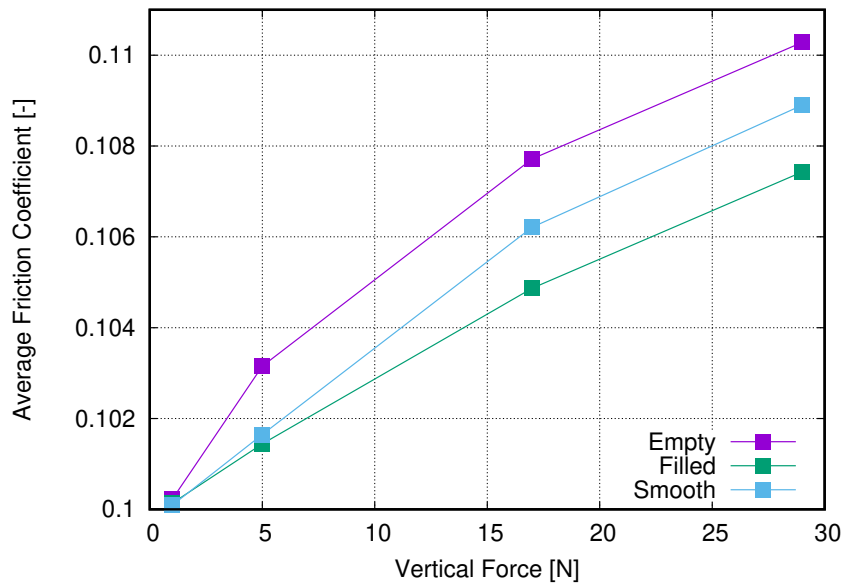


Figure 6.24: Variation of the average friction coefficient of the control surface with the vertical force for the empty and filled dimples and the smooth surface

6.3 A Micromechanical Constitutive Modeling Attempt for UHMWPE

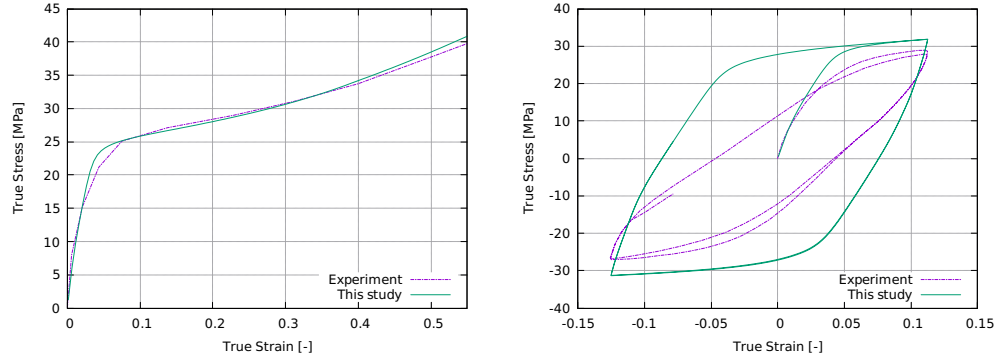
The FE models in literature [34, 40, 47] of the implants employ simple constitutive models. A recent study [50] suggests that FE simulations should employ multi-scale constitutive models, which consider the nano-, micro- and meso- structure of UHMWPE. The UHMWPE models in literature are more limited compared to that of the HDPE and most of them are phenomenological. Moreover, they do not include phenomena such as texture evolution, that is reported to be observed in UHMWPE knee inserts that are retrieved from patients [56]. Material properties of the HDPE constitutive model is adjusted in this section in order to obtain a constitutive model for UHMWPE. HDPE constitutive model of Chapter 5 employs statistical information according to the spherulites. However, the spherulitic morphology is associated with PE of slightly lower molecular weight than UHMWPE. In UHMWPE, spherulites are observed only in Basell 1900 resins, that are not produced anymore but large stocks were available [54]. The information of the spherulite morphology is also employed in this section, since it provides transversely symmetric clusters that are suitable for a Taylor type homogenization. Alternatively, when the required information on the spatial distribution of the crystals is not available or described simply as random, the interface normals of the model and the lattice orientations of the crystalline phase may be selected as random, similar to the ALAMEL [112] model.

Above the glass transition temperature, rubber elasticity is considered to provide a good representation of the behavior of the amorphous phase of semicrystalline polymers [13]. Since the lamellae of PE are thin, dislocations are not trapped in the crystalline phase, and consequently a strain hardening is not observed in the crystalline phase of semicrystalline polymers [80]. Therefore, the overall true stress-strain curve is considered as the sum of the almost constant stress of the plastic response of the crystalline phase and the increasing stress of the rubber elasticity of the amorphous phase. We consider that the characteristics of the crystalline phase is identical with the HDPE, while the long chains in the amorphous regions are more entangled in the UHMWPE, by also taking into account that the UHMWPE is tougher than HDPE. In micromechanical models of semicrystalline polymers, generally the 8-chain rubber elasticity formulation is incorporated as a back-stress tensor of a viscoplastic consti-

tutive model of the amorphous phase, following Lee et al. [61]. In rubber elasticity, the chain length parameter N is a material parameter and determines the value of the locking stretch, where the stresses approach infinity. The locking stretch is the result of extension of the covalently bonded strong polymer chains along the loading direction, and occurs earlier with decreasing values of the chain length parameter N . In order to obtain a good fit with the simple tension curve of the experiments of Bergstrom et al. [18], the following material parameters of the amorphous phase are changed compared to their values in Table 5.1. The chain length parameter N is decreased to 3, and the parameter μ_R that scales the magnitude of kinematical hardening described by the 8-chain elasticity formulation is increased to 2.5 and the reference shear strength τ_0 of the amorphous phase is decreased to 4 MPa. Finally, since the initial slope of UHMWPE response is higher, the shear modulus G is increased to 70 MPa. The response of the model composed of 32 realizations of disk-like spherulite instances with 32 slices are presented Figure 6.25, with the results of the experiments of Bergstrom et al. [18]. According to the figure, in the experiments, UHMWPE unloads with a slope that results in a lower plastic dissipation than that of our model. Therefore, the predictions of our model for the unloading and the subsequent loading stages are quite different than the experiment. The microsphere rubber elasticity model [73] is also employed as the amorphous phase constitutive model, without viscoplasticity, and a good fit for the simple compression curve in Figure 6.25a could not be obtained in that case. It should be mentioned that although it is known that the amorphous phase is more resistant than the crystalline phase to plastic deformation [64], in order to obtain a good fit with the experimental stress-strain curve in Figure 6.25a, yield stress of the amorphous phase is taken as half of the critical resolved shear stress of the easiest slip system of the crystalline phase in the present model. Therefore, the higher plastic dissipation of the model compared to the experiment is probably associated with the amorphous phase constitutive model. It could be noted that the response of our model in Figure 6.25b is similar to that of an earlier UHMWPE constitutive model, Bergstrom et al. [17], as presented in Bergstrom et al. [18].

While we do not find the response of the obtained micromechanical model as representative of the UHMWPE response, applicability of the model in FEM analyses of

Section 6.2 is investigated, for the smooth surface case, composed of 12817 C3D8IH elements with 8 integration points. Four realizations of the disk-like spherulite models with 32 slices are employed with identical orientations in all integration points. When the crystal plasticity is neglected and crystalline phase is simply taken as orthotropic elastic with St. Venant hyperelasticity and the amorphous phase is modeled with 8-chain rubber elasticity, the analysis completes in 25 hours with 12 threads and an Intel CPU. The vertical loading of the sphere completed in 16 load increments, where the total number of the severe discontinuity contact iterations and equilibrium iterations are 32 and 42, respectively. The sliding of the sphere is solved in 47 displacement increments, with 96 contact and 126 equilibrium iterations. The constitutive model does not require hybrid formulation and performs equally well with C3D8I elements as well. We were able to solve the analysis with 20-noded C3D20R elements that has quadratic shape functions and employs a reduced-integration scheme, which are also employed in Chapter 2. However, the obtained stress fields were not as smooth as the ones obtained with C3D8I or C3D8IH. The same problem is also observed when the three—network model of Bergsrom and Bischoff [16] is employed with C3D20RH elements. As for the case when the neglected crystal plasticity is also taken into account in all elements of the mesh, the vertical loading step and approximately 30% of the sliding path completed in approximately 10 days, and in the following days the Abaqus crashed with an illegal memory reference error, that we have considered as a hardware problem. The vertical loading completed in 389 load increments, where the total number of the contact and equilibrium iterations are 170 and 748, respectively including the ones in the increment attempts that were not solved. The 30% of the sliding path is solved in 266 displacement increments, composed of 251 contact and 548 equilibrium iterations, respectively including the ones in the increment attempts that were not solved. The numerical difficulties in crystal plasticity constitutive model is a major reason of the obtained small increment sizes. Therefore, a single crystalline slice in a disk-like model in an integration point of an element limits the increment size of the complete model. Van Dommelen [107], employs a sub-incrementation scheme in their constitutive model for this purpose. In that scheme, when a numerical problem occurs at an integration point, the aimed deformation state at that integration point is solved in a number of sub-increments, instead of decreasing the size of the FE load increment. This approach should also be employed in our model. On the



(a) Uniaxial tension at approximately 0.7% true strain rate (b) Cyclic loading at approximately 5% true strain rate

Figure 6.25: Comparisons with the experimental results of Bergstrom et al. [18] for the sterilized GUR 1050 resin

other hand, we consider that the incremental application of a general load increment is rather arbitrary. Additionally, the associated time interval of each sub-increment is not known. Therefore, the influence of sub-incrementation on the results should be studied, considering the potential risk of employing this scheme in the majority of the elements by choosing a large increment size.

6.4 Discussion and Conclusions

Since a decrease in the friction coefficient is considered to decrease the wear rate, the variation of the friction coefficient with the cylindrical dimples on UHMWPE surface is studied with FEM. The diameter, thickness, sliding speed, dimple filling material and vertical force are the considered parameters. According to the results, the empty dimples increase the average friction coefficient. On the other hand, filled dimples could decrease the average friction coefficient by 5% compared to the smooth UHMWPE surface for the studied problem. It is attempted to obtain a micromechanical constitutive model for UHMWPE, by adjusting the material parameters of the amorphous phase of the HDPE model of Chapter 5. The predicted cyclic response is not in good agreement with the experiments since the dissipation predicted by the model is higher than that of the experiment. The amorphous phase constitutive model

is probably the reason of the high plastic dissipation. In that case, it should be extended to limit the observed excessive plastic dissipation in order to obtain more realistic results.



CHAPTER 7

CONCLUSION AND RECOMMENDATIONS

This study is concerned with the development of a micromechanical constitutive model for semicrystalline polymers where an interaction among the adjacent regions is considered. The neighborhood information is obtained from the well characterized spherulite morphology of polyethylene. Therefore, the thesis starts with the study of the deformation of the spherulite morphology with a finite element method model that has similarities with the considered modeling approach. The model discretizes a disk-like spherulite into amorphous and crystalline slices, separated by planar interfaces. An important aspect of this model is the consistent deformation of the amorphous and the crystalline phases at the spherulite length scale. This feature separates the model from the other spherulite models employing crystal plasticity in literature. Moreover, planar interfaces in the model allow rank-1 laminates, the widely recognized basic structural unit of semicrystalline polymers, to be readily employed in the simplified versions. In order to investigate the reason of a high activity of $\{110\}\langle 1\bar{1}0\rangle$ slip system, a three-dimensional spherulite model is also studied, and it is found that the discrepancy is not an artifact that is unique to our approach. In order to obtain simplified versions of the disk-like spherulite model, a set of methods are developed by extending the rank-1 laminates to support an arbitrary number of lamination directions. The uniform strain field assumption of rank-1 laminates is retained, and consequently the equilibrium and compatibility equations are not satisfied together on the interfaces. The approximation of the relaxed condition improves with finer discretizations. The overall response of the geometries that are discretized with planar interfaces into regions with at least one and up to two neighbors are solved with the methods. The methods are consistent averaging schemes, and satisfy the Hill's energy criterion and the separation of length scales principle. According to these qualities,

the methods could be employed in micromechanical constitutive models. Nevertheless, volume is not conserved in the transition between the length scales, except at small strains where this condition is trivially satisfied. Disk-like spherulite model is solved with the proposed methods and sequential laminates from literature, and the results are compared. Certain versions of the method provide an expected approximately transversely isotropic response. The ribbon-shaped lamellae of spherulites, which are twisted around the radial directions, are also modeled. This demonstrates the applicability of the methods on different geometries, and additionally a desired tension-compression asymmetry is obtained for the uniaxial response of the lamella model. Another simplified version of the disk-like spherulite model, which does not employ the uniform strain assumption is also studied. The model is based on a compatible strain field in cylindrical coordinates at small strains. The proposed strain field could also be employed in regions between the interfaces of non-parallel rank-1 laminates where the axis of intersection is the z direction of the cylindrical coordinate system. Such a strain field may enable employment of constitutive models, that require the derivatives of the strain field, with the rank-1 laminates. The disk-like spherulite models are considered as analogous to the symmetric crystals with many common slip systems, since their response is transversely isotropic and the interlamellar shear deformation mode of the amorphous phase is considered to act as another slip system in the literature. Therefore, the Taylor approach is employed and considered suitable for the homogenization of randomly oriented disk-like models, and the aimed micromechanical constitutive model is obtained. The response and deformation characteristics of the model are quite similar to the other studies since the deformation modes considered in the constitutive models of the amorphous and crystalline phases are very similar. However, the principles of the model are quite different than the homogenization schemes discussed in the introduction. In particular, the separation of the length scales principle is satisfied and the spatial information on the distribution of heterogeneities that is not present in the Taylor approach, is introduced. On the other hand, the model does not depend on the size or shape of the heterogeneities, but the orientations of the interfaces. It should also be remarked that, as the other constitutive models of semicrystalline polymers, this model is also an approximation, especially since the compatibility and equilibrium equations are not satisfied together. Following are the other notable conclusions and the associated

recommendations.

- Employment of the crystallographic slip as the only plastic deformation mode in crystalline phase results in plastically soft polar regions, which contradicts with the experimental observations. The high activity of $\{110\}\langle 1\bar{1}0\rangle$, any activity of which is also questioned, is also a result of this simplification. While this approach is widely employed in the literature, other deformation modes which are more easily activated than $\{110\}\langle 1\bar{1}0\rangle$, should be taken into account to obtain more realistic models. We consider that this simplification may also be responsible for the spurious texture in simple shear that is present in the literature and also in our constitutive model. In the literature, the spurious texture is incorrectly attributed to a different condition, as we have initially attributed the high $\{110\}\langle 1\bar{1}0\rangle$ slip system activity to the disk-like idealization. Therefore, the importance of the neglected twinning and martensitic transformations are not widely appreciated among the modeling community, and to increase the awareness more research should be published on that subject.
- The neighborhood information that is considered in the proposed approaches could potentially lead to model phenomena observed in semicrystalline polymers such as kinking or cavitation. The tension-compression asymmetry of the model of the twisted lamellar microstructure is attractive for modeling the kinking phenomenon. The kinematical incompatibility distribution in the disk-like spherulite models resembles the separations that would be obtained if cavitation was modeled. Additionally, the methods might also be employed to consider the mosaic blocks of the lamellae. However, in order to attribute a meaning to the displacement jumps on the interfaces, possible interpenetrations should be eliminated by limiting the jumps to opening or sliding gaps only.
- The models could also be applied in micromechanical constitutive models of heterogeneous materials other than semicrystalline polymers, when the neighborhood information is available.
- Formulation could be streamlined to automatically discretize a given geometry and to consider more than two neighbors per region, and could be extended to conserve volume across the length scales.

- The strain field that connects rank-1 laminates may be employed with our methods that extend the rank-1 laminates to support arbitrary number of lamination directions.
- Elastic response of the models with kinematical compatibility and static equilibrium lies above and below the response of the sequential laminates, as expected. Similarly, the responses of the weighted hybrid approach are between that of the approaches with kinematical compatibility and static equilibrium.
- The approximately transversely isotropic response obtained with our disk-like models is also attractive for investigating the applicability of the proposed methods in a similar manner with the differential schemes of homogenization.
- While the narrow variation of the results with the number of realizations and among different random orientation sets indicate the expected suitability of the Taylor type-homogenization, the disk-like spherulite models could also be homogenized with more realistic approaches such as the self-consistent scheme.
- Texture evolution under compression could be compared with the experimental studies in literature in more detail, especially with Bartczak et al. [12] by following their load cycles in uniaxial compression to identify the influence of the recoverable part of interlamellar sliding on the obtained textures.
- Coulomb-type hardening of the critical resolved shear stresses could be taken into account by extending the constitutive model of the crystalline phase, and the influence of this phenomenon on the macroscopic response could be studied especially under uniaxial compression.
- The disk-like spherulite models homogenized with the statically equilibrated formulation, that has almost compatible amorphous-crystalline interfaces in static equilibrium, can also desirably slow down the texture evolution in the Taylor approach, which is known otherwise to provide too sharp textures.
- The mechanism that leads to the discrepancy between the UHMWPE constitutive modeling attempt and the experiments should be studied, and introduced to the micromechanical constitutive model.

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APPENDIX A

CONTRIBUTION OF THE SLIP SYSTEMS OF POLYETHYLENE TO THE RATE OF PLASTIC DEFORMATION TENSORS

In this section, the plastic velocity gradient

$$\mathbf{L}^p = \dot{\mathbf{F}}^p (\mathbf{F}^p)^{-1} ,$$

the plastic rate of deformation

$$\mathbf{d}^p = \frac{1}{2} \left(\mathbf{L}^p + (\mathbf{L}^p)^T \right) ,$$

and the plastic spin tensors

$$\mathbf{w}^p = \frac{1}{2} \left(\mathbf{L}^p - (\mathbf{L}^p)^T \right) ,$$

are presented for eight cases, where $\mathbf{F}^p$ is the plastic part of the deformation gradient.

In each case, only one slip system of PE is considered to be active with a slip rate of $\dot{\gamma}$.

(100)[001]:

$$\mathbf{L}^p = \dot{\gamma} \mathbf{c} \otimes \mathbf{a}$$

$$\mathbf{d}^p = \dot{\gamma} \frac{1}{2} (\mathbf{c} \otimes \mathbf{a} + \mathbf{a} \otimes \mathbf{c})$$

$$\mathbf{w}^p = \dot{\gamma} \frac{1}{2} (\mathbf{c} \otimes \mathbf{a} - \mathbf{a} \otimes \mathbf{c})$$

(010)[001]:

$$\mathbf{L}^p = \dot{\gamma} \mathbf{c} \otimes \mathbf{b}$$

$$\mathbf{d}^p = \dot{\gamma} \frac{1}{2} (\mathbf{c} \otimes \mathbf{b} + \mathbf{b} \otimes \mathbf{c})$$

$$\mathbf{w}^p = \dot{\gamma} \frac{1}{2} (\mathbf{c} \otimes \mathbf{b} - \mathbf{b} \otimes \mathbf{c})$$

(110)[001]:

$$\begin{aligned}\mathbf{L}^p &= \dot{\gamma} (0.55\mathbf{c} \otimes \mathbf{a} + 0.83\mathbf{c} \otimes \mathbf{b}) \\ \mathbf{d}^p &= \dot{\gamma} \frac{1}{2} [0.55 (\mathbf{c} \otimes \mathbf{a} + \mathbf{a} \otimes \mathbf{c}) + 0.83 (\mathbf{c} \otimes \mathbf{b} + \mathbf{b} \otimes \mathbf{c})] \\ \mathbf{w}^p &= \dot{\gamma} \frac{1}{2} [0.55 (\mathbf{c} \otimes \mathbf{a} - \mathbf{a} \otimes \mathbf{c}) + 0.83 (\mathbf{c} \otimes \mathbf{b} - \mathbf{b} \otimes \mathbf{c})]\end{aligned}$$

(1 $\bar{1}$ 0)[001]:

$$\begin{aligned}\mathbf{L}^p &= \dot{\gamma} (0.55\mathbf{c} \otimes \mathbf{a} - 0.83\mathbf{c} \otimes \mathbf{b}) \\ \mathbf{d}^p &= \dot{\gamma} \frac{1}{2} [0.55 (\mathbf{c} \otimes \mathbf{a} + \mathbf{a} \otimes \mathbf{c}) - 0.83 (\mathbf{c} \otimes \mathbf{b} + \mathbf{b} \otimes \mathbf{c})] \\ \mathbf{w}^p &= \dot{\gamma} \frac{1}{2} [0.55 (\mathbf{c} \otimes \mathbf{a} - \mathbf{a} \otimes \mathbf{c}) - 0.83 (\mathbf{c} \otimes \mathbf{b} - \mathbf{b} \otimes \mathbf{c})]\end{aligned}$$

(100)[010]:

$$\begin{aligned}\mathbf{L}^p &= \dot{\gamma} \mathbf{b} \otimes \mathbf{a} \\ \mathbf{d}^p &= \dot{\gamma} \frac{1}{2} (\mathbf{b} \otimes \mathbf{a} + \mathbf{a} \otimes \mathbf{b}) \\ \mathbf{w}^p &= \dot{\gamma} \frac{1}{2} (\mathbf{b} \otimes \mathbf{a} - \mathbf{a} \otimes \mathbf{b})\end{aligned}$$

(010)[100]:

$$\begin{aligned}\mathbf{L}^p &= \dot{\gamma} \mathbf{a} \otimes \mathbf{b} \\ \mathbf{d}^p &= \dot{\gamma} \frac{1}{2} (\mathbf{a} \otimes \mathbf{b} + \mathbf{b} \otimes \mathbf{a}) \\ \mathbf{w}^p &= \dot{\gamma} \frac{1}{2} (\mathbf{a} \otimes \mathbf{b} - \mathbf{b} \otimes \mathbf{a})\end{aligned}$$

(110)[1 $\bar{1}$ 0]:

$$\begin{aligned}\mathbf{L}^p &= \dot{\gamma} [0.46 (\mathbf{a} \otimes \mathbf{a} - \mathbf{b} \otimes \mathbf{b}) + 0.69\mathbf{a} \otimes \mathbf{b} - 0.30\mathbf{b} \otimes \mathbf{a}] \\ \mathbf{d}^p &= \dot{\gamma} [0.46 (\mathbf{a} \otimes \mathbf{a} - \mathbf{b} \otimes \mathbf{b}) + 0.19 (\mathbf{a} \otimes \mathbf{b} + \mathbf{b} \otimes \mathbf{a})] \\ \mathbf{w}^p &= \dot{\gamma} \frac{1}{2} (\mathbf{a} \otimes \mathbf{b} - \mathbf{b} \otimes \mathbf{a})\end{aligned}$$

(1 $\bar{1}$ 0)[110]:

$$\begin{aligned}\mathbf{L}^p &= \dot{\gamma} [0.46 (\mathbf{a} \otimes \mathbf{a} - \mathbf{b} \otimes \mathbf{b}) - 0.69\mathbf{a} \otimes \mathbf{b} + 0.30\mathbf{b} \otimes \mathbf{a}] \\ \mathbf{d}^p &= \dot{\gamma} [0.46 (\mathbf{a} \otimes \mathbf{a} - \mathbf{b} \otimes \mathbf{b}) - 0.19 (\mathbf{a} \otimes \mathbf{b} + \mathbf{b} \otimes \mathbf{a})] \\ \mathbf{w}^p &= \dot{\gamma} \frac{-1}{2} (\mathbf{a} \otimes \mathbf{b} - \mathbf{b} \otimes \mathbf{a})\end{aligned}$$

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