MULTISCALE MODELING OF THE STRUCTURE–PROPERTY RELATIONSHIP FOR SEMICRYSTALLINE MATERIALS

J. A. W. van Dommelen, W. A. M. Brekelmans, and F. P. T. Baaijens
Dutch Polymer Institute, Eindhoven University of Technology,
Department of Mechanical Engineering,
PO Box 513, 5600 MB Eindhoven, The Netherlands

Summary A multiscale numerical model is developed to describe the structure–property relationship for semicrystalline polymers. A polycrystalline model is used to represent the mechanical behavior of the semicrystalline microstructure. The basic element in this model is a layered two-phase composite inclusion, comprising both a crystalline and an amorphous domain. An aggregate of composite inclusions is used in macroscopic finite element models of unfilled and particle-modified semicrystalline materials.

INTRODUCTION

The mechanical performance of semicrystalline polymers is strongly dependent on the underlying microstructure, consisting of crystallographic lamellae and amorphous layers. In this work, a micromechanically-based numerical model for the elasto-viscoplastic deformation and texture evolution of semicrystalline materials is presented.

METHODS

A distinction between three different scales is made, as is schematically depicted in figure 1. The constitutive properties of the material are identified at the microscopic scale for the individual crystallographic and amorphous components. At the mesoscopic scale, an aggregate of individual phases is formed, with either a random or a preferential orientation distribution. To bridge between those scales, an elasto-viscoplastic two-phase composite inclusion model is formulated [1–3]. Each composite inclusion consists of a crystalline lamella which is assumed to plastically deform by rate dependent crystallographic slip, and an amorphous layer which is modeled as elasto-viscoplastic with strain hardening resulting from molecular orientation. The local inclusion-averaged deformation and stress fields are related to the mesoscopic fields of the aggregate by a hybrid interaction law, which compromises between local compatibility and local equilibrium. A full micro-meso-macrolevel bridge is obtained by using an aggregate of composite inclusions in each integration point of a macroscopic finite element (FE) model.

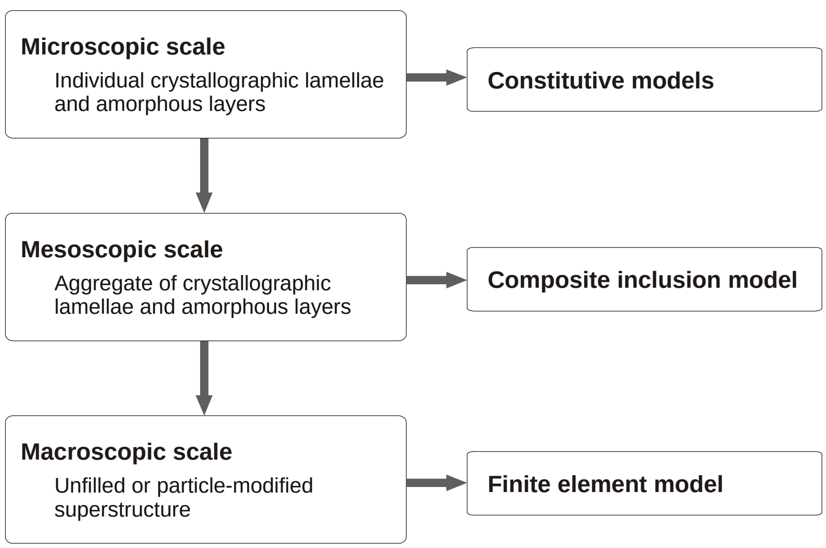

Figure 1: Different scales which can be identified in particle-modified semicrystalline polymeric systems.

APPLICATIONS

The multiscale model is employed to study the mechanics of intraspherulitic deformation for unfilled polyethylene [4]. The spherulitic macrostructure is described by FE models of representative volume elements (RVE). The macroscopic stress-strain response resembles that of a random polycrystalline model; however, the RVE model includes the geometrical effect of the anisotropic structure within a spherulite, causing strain concentrations in the centers. The deformations are linked to microstructural processes as interlamellar deformation and intralamellar crystallographic slip.

The stacked lamellar morphology commonly observed in extruded semicrystalline materials gives rise to a strong influence of the direction of flow with respect to the loading direction on the stability and localization phenomena in tensile experiments. The multiscale numerical model is used to simulate the effect of a stacked lamellar microstructure on the macroscopic behavior [5]. The selection of orientations is based on WAXS experiments on extruded material. The averaged fields of an aggregate, having preferential orientations, constitute the mechanical behavior of extruded material. A macroscopic tensile bar is described by a finite element model. The microstructure-induced deformation hardening in the extrusion direction is found to stabilize the macrostructure, when loaded in the flow direction, whereas when loaded perpendicular to the extrusion direction, a neck is formed.

To enhance the toughness of semicrystalline polymeric materials, it is common practice to blend these materials with rubber particles. The present-day notion of the toughening-mechanism in semicrystalline polymers is expressed in a criterion, which states that toughened material behavior occurs when the average interparticle matrix ligament thickness is smaller than a critical value [6], which is attributed to thin layers of transcrystallized material appearing in the microstructural morphology [7–9]. Multiscale calculations are performed on particle-modified HDPE [10, 11]. The effect of a transcrystallized structure of matrix material versus randomly oriented material is examined. A limited effect of dispersed shear yielding for transcrystallized orientations is observed. Moreover, a relocation of maximum triaxial stresses in the polar area is found. Further improved properties in a specific loading direction are obtained for a hypothesized, partly flow-induced, microstructure.

CONCLUSIONS

A multiscale model has been developed to investigate the micromechanically-induced behavior of unfilled and filled semicrystalline material. The macrostructure is described by RVE models. In each integration point of the finite element models, an aggregate of composite inclusions is used as a representative structural element that provides the constitutive behavior of the material at the mesoscopic level. Material properties are assigned at the microstructural level to both the amorphous and the crystalline phase. Besides these properties, the mesoscopic constitutive behavior is formed by the crystallographic and lamellar orientations of the composite inclusions. The multiscale model is employed to study the mechanics of intraspherulitic deformation for polyethylene and the behavior of material with a lamellar row structure. Finally, multiscale calculations are performed on particle-modified HDPE. The effect of a transcrystallized structure of matrix material versus randomly oriented material is examined. A limited effect of the preferential orientations is observed. Further improved properties in a specific loading direction are obtained for a hypothesized, partly flow-induced, microstructure.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge Prof. D. M. Parks and Prof. M. C. Boyce of the Massachusetts Institute of Technology for their stimulating and helpful discussions concerning the modeling of particle-modified semicrystalline material.

References

[1] Lee B. J., Parks D. M., Ahzi S.: Micromechanical modeling of large plastic deformation and texture evolution in semicrystalline polymers. *J. Mech. Phys. Solids* **41:**1651–1687, 1993.

[2] Lee B. J., Argon A. S., Parks D. M., Ahzi S., Bartczak Z.: Simulation of large strain plastic deformation and texture evolution in high density polyethylene. *Polymer* **34**:3555–3575, 1993.

[3] Van Dommelen J. A. W., Parks D. M., Boyce M. C., Brekelmans W. A. M., Baaijens F. P. T.: Micromechanical modeling of the elasto-viscoplastic behavior of semi-crystalline polymers. *J. Mech. Phys. Solids* **51:**519–541, 2003.

[4] Van Dommelen J. A. W., Parks D. M., Boyce M. C., Brekelmans W. A. M., Baaijens F. P. T.: Micromechanical modeling of intraspherulitic deformation of semicrystalline polymers. *Polymer* **44**:6079-6101, 2003.

[5] Van Dommelen, J. A. W., Schrauwen, B. A. G., Van Breemen, L. C. A., Govaert, L. E.: Micromechanical modeling of the tensile behavior of oriented polyethylene, *J. Polym. Sci.*, submitted.

[6] Wu S.: Phase structure and adhesion in polymer blends: a criterion for rubber toughening. *Polymer* **26**:1855–1863, 1985.

[7] Muratoglu O. K., Argon A. S., Cohen R. E.: Crystalline morphology of polyamide-6 near planar surfaces. *Polymer* **36**:2143–2152, 1995.

[8] Muratoglu O. K., Argon A. S., Cohen R. E.: Toughening mechanism of rubber-modified polyamides. *Polymer* **36**:921–930, 1995.

[9] Bartczak Z., Argon A. S., Cohen R. E., Kowalewski T.: The morphology and orientation of polyethylene in films of sub-micron thickness crystallized in contact with calcite and rubber substrates. *Polymer* **40**:2367–2380, 1999.

[10] Van Dommelen J. A. W., Brekelmans W. A. M., Baaijens F. P. T.: Micromechanical modeling of particle-toughening of polymers by locally induced anisotropy. *Mech. Mater.* **35**:845–863, 2003.

[11] Van Dommelen J. A. W., Brekelmans W. A. M., Baaijens F. P. T.: Multiscale modeling of particle-modified polyethylene, *J. Mater. Sci.* **38**: 4393–4405, 2003.