M. Incorporating Higher-Order Tensors in the Computation of Polymer Composite Mechanical Properties

Principal Investigator (PI): Douglas E. Smith
University of Missouri at Columbia
E2411 Lafferre Hall / MAE Dept., Columbia, MO 65211
(573) 884-6552; fax: (573) 884-5090; e-mail: smithdoug@missouri.edu

Technology Area Development Manager: Joseph A. Carpenter
(202) 586-1022; fax: (202) 586-1600; e-mail: joseph.carpenter@ee.doe.gov

Participants: David A. Jack and Elijah C. Caselman, University of Missouri at Columbia

---

Contractor: University of Missouri at Columbia
Contract No.: NSF Grant DMI-0522694

Objective

- Develop a predictive capability that incorporates higher-order orientation tensors to evaluate mechanical properties of short- and long-fiber-reinforced polymer composites.

Approach

- Development of a model for predicting mechanical properties from higher-order orientation tensors derived from flow simulations or measurements that are not limited by the material symmetry requirements imposed by current models.
- Development of an automated three-dimensional (3D), voxel-based, finite-element modeling technique that may be used to predict the effective mechanical properties of short- and long-fiber suspensions.
- Integration of micro-computerized-tomography (micro-CT)-derived fiber reconstructions to illustrate the applicability of the proposed approach on production hardware to be obtained from industry or DOE.
- Statistical assessment of mechanical property calculations to address sampling issues associated with using the proposed averaging techniques.
- Demonstration of the proposed methodology on an industrially-relevant polymer-composite product where flow simulation software is to be provided by Moldflow Corporation.

Accomplishments

- Derived an analytical relationship between elastic mechanical properties and fiber-orientation tensors for statistically-independent short fibers, including both the property's mean and variance.
- Evaluated mechanical properties using existing property models from simple melt-flow simulations that employ 4^{th}- and 6^{th}- order-tensor closures.
- Developed Monte Carlo method for computing mean and variance of elastic properties of short-fiber composites from fiber-orientation-distribution functions.
- Evaluated mechanical properties of short-fiber composites via Monte Carlo sampling procedure, validating analytical approach developed as part of this project.

- Developed a 3D, voxel-based, finite-element model using a representative volume element (RVE) and related periodic boundary conditions for predicting elastic material properties of short- and long-fiber polymer composites.

Future Direction

- Continue verification work for 3D, voxel-based, finite-element procedure for fiber-reinforced polymer composites.
- Continue develop an automated procedure for defining 3D finite-element models of a representative cell that may be used with analytically-derived fiber-orientation-distribution functions, or fiber-orientation states obtained from micro-CT measurements.
- Compute elastic mechanical properties from 3D, voxel-based, finite-element model for short- and long- fiber composites.

Introduction

The purpose of this project is to develop a predictive capability that incorporates higher-order orientation tensors to evaluate mechanical properties of short- and long-fiber-reinforced polymer composites. To realize this goal, the project will develop a computational methodology to predict elastic mechanical properties from orientation tensors of second- and fourth-order which are computed during melt-flow simulations. The micromechanics of both short- and long-fiber suspensions will be simulated with a voxel-based finite-element modeling approach, so that the mechanical properties of randomly-oriented fiber samples can be computed. Simulated fiber- orientation states, as well as those obtained from micro-CT scans will serve as input. Finally, fiber- orientation states from industrially-relevant products will be evaluated with the new methodology to assess its applicability for complex geometries.

This report provides a brief description of the three-year project, a predictive method for mean and variance of elastic properties using higher-order tensors, Monte Carlo validation, and new results from our 3D, voxel-based, finite-element approach.

Project Deliverables

The primary deliverable of this research will be a relationship between higher-order (i.e., fourth) orientation tensors and the elastic mechanical properties for short- and long-fiber suspensions. An assessment of the variability of mechanical properties among fiber-orientation states will be provided, along with a computational procedure for evaluating the same properties for a specific orientation state. A final report will be generated to communicate the functional form of the mechanical properties and the relevant model parameters for specific examples studied.

Background

Orientation tensors are used extensively to represent the stochastic nature of polymer- composite fiber suspensions in a form that is suitable for large-scale melt-flow simulations. Orientation-tensor research over the past two decades has advanced the state-of-the-art related to polymer composites; however, the use of tensors today is still basically the same as it was when these methods were first developed nearly twenty years ago. In addition, the application of orientation tensors to long-fiber composites is nearly non-existent.

The continued advancement of our basic understanding of short- and long-fiber-reinforced polymer composites is critical in numerous industries including consumer products and automotive which rely heavily on the low cost, design flexibility, and superior performance offered by these materials. A major focus of the National Science Foundation / US Department of Energy / American Plastics Council-sponsored workshop[1] co-organized by the PI in June, 2004 was the need for the US automotive industry to incorporate more fiber-reinforced polymer composites in the design of

[1] See http://web.missouri.edu/~smithdoug/nsf/index.htm for workshop information.

future vehicles to reduce the weight, emissions, and fuel consumption.

For example, a specific goal of the 2010 FreedomCAR is to reduce the weight of an automotive structure by 50% for the same cost and durability as seen in today's products. The key to meeting these objectives is a comprehensive predictive engineering approach that is capable of accurately simulating critical attributes associated with the design of both the product and its manufacturing process. In this approach, a critical link that connects the computation of materials processing attributes with the prediction of a composite product's performance is the evaluation of structural mechanical properties from melt flow predictions.

Given the dramatic increases in affordable computing over the past decade, and the anticipated demands on our predictive capabilities for composite materials over the next decade, it is now time to reconsider assumptions that compose today's fiber-orientation models. For example, limiting fiber-orientation predictions to solving for only the second-order orientation tensor components in an effort to reduce computational effort may not be necessary. This project will provide a higher-order approximation to more accurately describe suspension mechanics for complex flow fields such as those where both shear and elongational velocity gradients exist in all three different planes.

Evaluating Elastic Properties from Orientation Tensors

One task of this research is to develop a formulation for computing short- and long-fiber- composite elastic properties from orientation tensors. Here we consider the expected value of the elasticity tensor $\langle C_{ijkl} \rangle$ defined in terms of the orientation distribution function $\psi(\mathbf{p})$ as

$$\langle C_{ijkl} \rangle = \int_S Q_{pi}(\mathbf{p}) Q_{qj}(\mathbf{p}) Q_{rk}(\mathbf{p}) Q_{sl}(\mathbf{p}) \overline{C}_{pqrs} \psi(\mathbf{p}) d\mathbf{p}$$

where $\mathbf{p}$ is a unit vector in the fiber direction as shown in Figure 1. In this calculation, $\overline{C}_{pqrs}$ is the underlying unidirectional compliance tensor for a single fiber (computed using the Mori-Tanaka, Halpin-Tsai, or similar inclusion model) and $Q_{ij}(\mathbf{p})$ is a rotation tensor that rotates the unidirectional components into a selected coordinate system. Elasticity tensor components may be computed in this manner for various orientation-distribution functions. However, when $\psi(\mathbf{p})$ is written in terms of the Fourier basis functions such as those given in Advani and Tucker[2], it can be shown that $\langle C_{ijkl} \rangle$ reduces to the well-known result

$$\begin{aligned}\langle C_{ijkl} \rangle &= B_1(a_{ijkl}) + B_2(a_{ij}\delta_{kl} + a_{kl}\delta_{ij}) \\ &+ B_3(a_{ik}\delta_{jl} + a_{il}\delta_{jk} + a_{jl}\delta_{ik} + a_{jk}\delta_{il}) \\ &+ B_4(\delta_{ij}\delta_{kl}) + B_5(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})\end{aligned}$$

where a_{ij} and a_{ijkl} are the 2nd- and 4th-order fiber-orientation tensors, δ_{ij} is the identity tensor, and the constants $B_1 – B_5$ are computed from $\overline{C}_{pqrs}$.

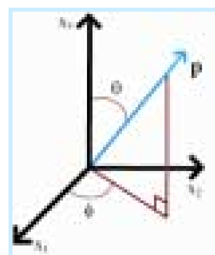

Figure 1. Coordinate system defining fiber direction with the unit vector p(θ,ϕ).

This derivation shows that when the elastic properties are computed as the volume-averaged result of unidirectional fibers weighted by the orientation-distribution function, the highest-order orientation tensor that contributes is the fourth. In this case, sixth- and higher-ordered orientation tensors do not define to the composite's elasticity tensor. What is important to note about this result is that the equations above implicitly assume that each fiber that composes a_{ij} and a_{ijkl} does not interact when stresses are imposed on the fiber-matrix structure. While the effect of fiber interaction has received much attention in melt-flow analyses, little

2 S. Advani and C.L. Tucker, Jn. of Rheology, vol. 31, no. 8, pp. 751-784, 1987.

effort has been devoted to understanding its role on elastic response of these composites. The elastic response of a high volume-fraction short-fiber composite, as well as long-fiber composites, is expected to be influenced by structural fiber-fiber interaction.

The variance σ^2_{ijkl} of the elastic properties may be computed in a similar manner by taking the second moment of the rotated unidirectional compliance tensor about the mean given as

$$\sigma^2_{ijkl} = \oint_{S^2} \left(Q_{qi}(\mathbf{p})Q_{rj}(\mathbf{p})Q_{sk}(\mathbf{p})Q_{tl}(\mathbf{p})\overline{C}_{qrst} - \left\langle C_{ijkl}\right\rangle\right)^2 \psi(\mathbf{p})d\mathbf{S}$$

For relatively simple $\psi(\mathbf{p})$, an analytical result for variance may be obtained by integrating the above equation. This has been performed as part of this research for several distribution functions. These results are beyond the scope of this report, but may be found elsewhere[3]. It is important to note here that while the mean value of the elasticity tensor $\left\langle C_{ijkl}\right\rangle$ contained orientation tensors through 4th order, the variance σ^2_{ijkl} is a function of orientation tensors through 8th order. Therefore, to use the result described here, analytical or approximate values of the 2nd-, 4th-, 6th- and 8th-order orientation tensors are required.

Monte Carlo Simulation

A Monte Carlo simulation method was developed to validate analytical results for the mean and variance obtained above for the elastic properties of a short-fiber composite. The Monte Carlo calculations included an Accept-Reject algorithm to obtain a set of fiber orientation angles for any given orientation distribution that is described by an orientation-distribution function or set of orientation tensors. The mean and variance of the compliance matrix are computed in these Monte Carlo simulations using a discrete form of the continuous equations given above for $\left\langle C_{ijkl}\right\rangle$ and σ^2_{ijkl}.

Several example Monte Carlo calculations have been performed to verify the analytical forms described above. Consider the fiber-orientation-distribution function

$$\psi(\theta,\phi) = \tfrac{9}{4\pi}\sin^8\theta\cos^8\phi$$

where θ and ϕ describe the fiber angle as shown in Figure 1. Figure 2 illustrates the form of this distribution function which is highly aligned in the x_1 direction. The figure also shows a representative volume of fibers with a volume fraction of 10% that were generated for the Monte Carlo simulations. Computed results for C_{1111} are shown in Figure 3 for 10^8 Monte Carlo simulations where matrix and fiber properties are taken from Tucker and Liang[4] and the volume fraction is 10%. As shown in the figure, this component of the compliance tensor varies between a predictable lower and upper bound with a linear probability distribution.

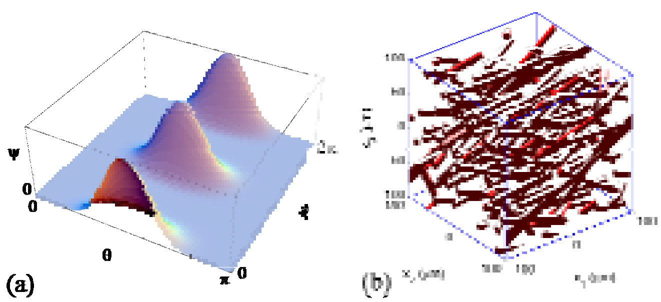

Figure 2. Fiber-orientation-distribution function used in example calculations (a) and a representative sample set of fibers (b).

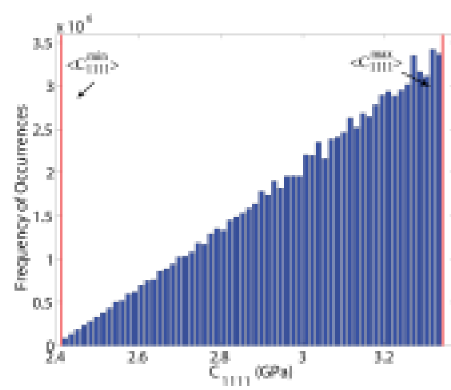

Figure 3. Histogram from Monte Carlo calculations for C_{1111} using 10^8 fiber occurrences.

---

[3] D.A. Jack and D.E. Smith, Jn. of Composite Materials, Accepted for Publication, 2007.

[4] C.L. Tucker and E. Liang, *Comp. Science and Technology*, vol. 59, pp.655-671, 1999.

Of more practical use is the predicted distribution and related statistical properties for multiple sets of fibers since we are typically interested in the elastic properties of a material point which can vary from part to part. To this end, we apply the Central Limit Theorem to compute the statistical nature of multiple sets of fibers such as that shown in Figure 2b. Figure 4 shows the results of 10^6 sets of fibers where each set contains 101 fibers. As predicted by the Central Limit Theorem, the results become normally distributed. Also shown is the analytically-evaluated normal distribution function obtained by applying the Central Limit Theorem to the analytical mean and variance equations given above. The figure clearly shows that the analytical form that we have derived for the statistical properties of fiber suspensions is verified through our Monte Carlo simulations.

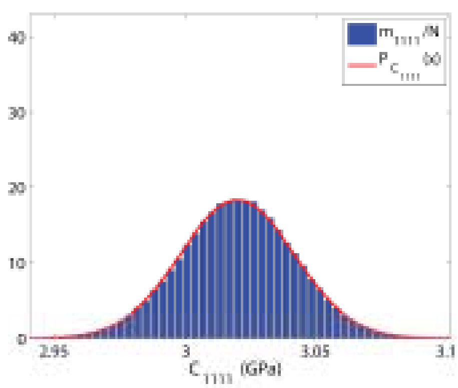

Figure 4. Histogram and normal probability distribution function for C_{1111} using 10^6 fiber sets.

Monte Carlo simulations have also been performed to predict the statistical behavior of the complete compliance tensor for both simple analytical fiber-orientation distributions and for distributions that arise during polymer melt flow. The latter are evaluated from numerical simulation of the fiber-orientation-distribution function evolution. In all cases, our analytical results are validated with Monte Carlo-simulation results. This work is the first to consider the statistical nature of short-fiber composite elastic properties at this level of detail.

Three-dimensional Finite-Element Analysis for Evaluating Mechanical Properties

All of the calculations described above are restricted to short-fiber composites, and explicitly assume that the stress state around each fiber is not influenced by its neighbors. To address these shortfalls, a 3D finite-element modeling technique is being developed that will be used to gain a better understanding of the effect of fiber-fiber interaction in the elastic solid. Our approach is to use a voxel-based finite-element model to represent a characteristic elastic unit within the composite structure. Properties of each point within the model will be based on fiber distributions obtained analytically, or through experimentally-derived procedures, making it possible to include short- and long-fiber composites in the analysis. To reduce the number of degrees-of-freedom in these 3D finite-element simulations, a custom element has been developed that can incorporate a large number of sampling (or Gauss) points within its domain, and thus provide a more refined spatial definition without increasing the number of degrees of freedom.

This modeling approach is being implemented in the general-purpose finite-element program ABAQUS using a user-defined element. RVEs (see e.g., Figure 2b) are defined by a uniform grid of 3D elements where the elastic properties of each Gauss point are defined by its spatial location in relationship to the position of the fibers. The number of Gauss points is increased within each element to provide the required spatial refinement. Unique elemental shape functions have been created to increase the complexity of the displacement field in each element so that elements which contain both matrix and fiber are not over-constrained by the more traditional bi-linear response. In addition, periodic boundary conditions are implemented so that both axial and shear loads may be accurately applied to the RVE. A fitting procedure has also been developed to compute elastic properties from multiple RVE runs where constraints are imposed to enforce a desired material symmetry.

Figure 5 shows a finite-element model of a single fiber embedded in a polymer matrix that has been used to validate our calculation procedure. The dimensions and material properties for this model are taken from Xia, et al.[5] who provide both simulation and experimental results for comparison. Figure 6 shows a voxel-based finite- element model of the same geometry where contours are provided to indicate the fiber's location. Note that this plot simply shows element- by-element values, and thus does not provide insight into the spatial refinement with each element that is included with our multiple-Gauss- point approach. Both of these models were loaded in axial tension in each of the coordinate directions in addition to various shear-loading conditions.

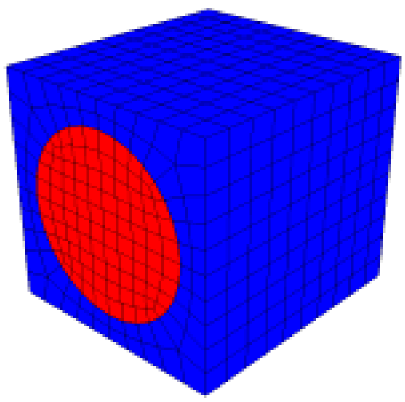

Figure 5. Two-material RVE finite-element model as defined by Xia et al.[5].

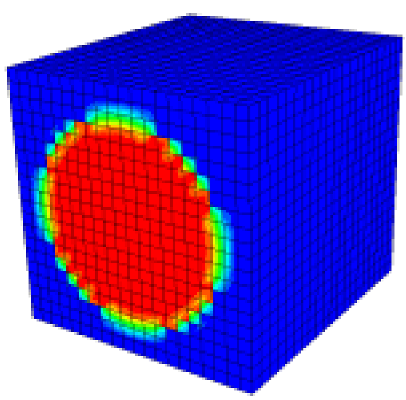

Figure 6. Voxel-based finite-element model of RVE with contours illustrating elements that contain fiber, matrix, or both.

[5] Z. Xia, Y. Zhang, and F. Ellyin, *Solids and Structures*, vol. 40, pp. 1907-1921, 2003.

Table 1 compares engineering elastic constants obtained from the models in Figures 5 and 6 with values provided by Xia et al.[5]. It can be seen that our two-material model (see e.g., Figure 5) provides results that are nearly identical to Xia's results which compare well with experimental data as reported in their paper. Table 1 also shows that results obtained with our voxel finite-element model agree quite well with the two-element models. These latter results are computed with ten elements in each of the *x*, *y*, and *z* directions where the elements have six Gauss points in each direction.

In other simulations, we have been able to obtain similar agreement with results provided by Tucker and Liang[4] who model a more complicated short-fiber composite sample. Other runs have been performed to establish limits on the number of elements and Gauss points that will be needed for modeling RVEs with smaller fibers (such as that shown in Figure 2b).

Table 1. Elastic properties computed from two-material and voxel-based finite-element models (moduli in GPa).

Elastic Constants	Xia et al.[5]	Two-mat'l model (Fig. 5)	Voxel-based model (Fig. 6)
E11	214	214.4	216.1
E22	143	143.7	147
E33	143	143.7	147
G12	54.2	53.9	56.4
G23	45.7	45.4	45.6
G13	54.2	53.9	56.4
v12	0.195	0.195	0.194
v23	0.253	0.253	0.244
v13	0.195	0.195	0.194

Conclusions

This research project focuses on the prediction of elastic mechanical properties from higher-order fiber-orientation tensors. As part of this work, the expression for computing elastic properties form fiber-orientation tensors is verified and shown to ignore fiber-fiber interaction. In addition, the role of higher-order tensors on mechanical property prediction when using this approach is explored. Finally, results of a 3D, voxel-based finite-element modeling procedure have been shown to be in good agreement with more conventional FE approaches.

Presentations/Publications/Patents

1. D.A. Jack and D.E. Smith, 'Elastic Properties of Short-Fiber Polymer Composites, Derivation and Demonstration of Analytical Forms for Expectation and Variance from Orientation Tensors', Accepted for publication in *Jn. of Composite Materials*, 2007.

2. D.A. Jack and D.E. Smith, 'A Statistical Method to Obtain Material Properties from the Orientation Distribution Function for Short-fiber Polymer Composites', IMECE2005-81263, Proceedings of ASME IMECE'05, Orlando, Florida, Nov. 5-11, 2005.

3. D.A. Jack and D.E. Smith, 'Mechanical Properties from the INV6 Closure for Short-Fiber Suspensions', SPE ANTEC 2006, Charlotte, North Carolina, May 7-11, 2006.

4. Jack, D.A. *Advanced Analysis of Short-Fiber Polymer Composite Material Behavior with Higher-order Orientation Tensor Closure Methods*. PhD thesis, University of Missouri - Columbia, December 2006.

5. D.E. Smith, M.T. Grimshaw, and D.A. Jack, 'Incorporating Higher Order Tensors in the Computation of Polymer Composite Mechanical Properties', Poster presentation at the *2006 NSF Design, Service and Manufacturing Grantees and Research Conference,* St. Louis, Missouri, July 24-27, 2006.