FROM PROCESS MODELING TO ELASTIC PROPERTY PREDICTION FOR LONG-FIBER INJECTION-MOLDED THERMOPLASTICS[1]

Ba Nghiep Nguyen[2] [(a)], Vlastimil Kunc[(b)], Barbara J. Frame[(b)],
Jay H. Phelps[(c)], Charles L. Tucker[(c)] III, Satish K. Bapanapalli[(a)],
James D. Holbery[(a)], Mark T. Smith[(a)]

(a) Pacific Northwest National Laboratory, P.O. Box 999, Richland, WA 99352, (b) Oak Ridge National Laboratory, P.O. Box 2009, Oak Ridge, TN 37831, (c) University of Illinois at Urbana-Champaign, Department of Mechanical Science and Engineering, Urbana, IL 61801

Abstract

This paper presents an experimental-modeling approach to predict the elastic properties of long-fiber injection-molded thermoplastics (LFTs). The approach accounts for fiber length and orientation distributions in LFTs. LFT samples were injection-molded for the study, and fiber length and orientation distributions were measured at different locations for use in the computation of the composite properties. The current fiber orientation model was assessed to determine its capability to predict fiber orientation in LFTs. Predicted fiber orientations for the studied LFT samples were also used in the calculation of the elastic properties of these samples, and the predicted overall moduli were then compared with the experimental results. The elastic property prediction was based on the Eshelby-Mori-Tanaka method combined with the orientation averaging technique. The predictions agree reasonably well with the experimental LFT data.

Background

Long-fiber injection-molded thermoplastics have generated great interest within the automotive industry since these materials that have already been used for semi-structural applications are now candidate materials for structural applications to reduce vehicle weight. LFTs offer much better mechanical performance than the short-fiber counterparts since long fibers owing to higher fiber aspect ratios considerably increase the composite stiffness and strength, and enhance creep and fatigue endurance [1-2]. Although LFTs have been commercially available for nearly twenty years and have been used in the automotive industry for more than fifteen years [3], there exists little experimental and modeling work on these materials. The objective of our research is to develop computational tools to assist in development of LFTs for automotive applications [4-6]. To use LFTs for structural applications, it is essential to be able to (i) predict the as-formed LFT microstructure as a function of the constituents' properties and

---

[1] This manuscript has been authored by Battelle Memorial Institute, Pacific Northwest Division, under Contract No. DE-AC06-76RL0 1830 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

[2] Corresponding author Tel.: (509) 375-3634; fax: (509) 375-6736. E-mail address: ba.nguyen@pnl.gov.

characteristics as well as processing parameters, (ii) predict their thermoelastic properties and nonlinear responses as a function of the microstructure, and (iii) develop characterization methods to obtain all the key microstructural data for the model validation. This paper presents the state-of-the-art of process-linked-property prediction by applying the current process model to simulate the injection-molding of long glass fiber filled polypropylene and then utilizes the predicted LFT microstructure to compute the effective elastic properties of the final composite [4-6].

In order to predict the orientation state due to an injection molding process, the equations of balance of mass, momentum, and energy must be solved so that a velocity field can be computed. In a *flow/orientation decoupled approach*, the orientation state is determined using the velocity field that is computed as if fibers were absent. The Folgar-Tucker model has been used for more than two decades to simulate the flow-induced fiber orientation in short-fiber systems [7]. This model contains a parameter named C_{I} accounting for the fiber-fiber interaction in non-dilute fiber suspensions. Applying the Folgar-Tucker model to LFTs shows that this model cannot capture fiber orientation in these materials. For instance, it cannot predict the larger core that exists in the skin/shell/core orientation layer structure of LFTs. Recently efforts have been made to improve the Folgar-Tucker model for concentrated suspensions in which intense fiber/fiber interactions slow down fiber rotation. An additional factor was then introduced to slow down fiber rotation, and the improved model is defined as the *reduced strain closure (RSC) model* [8]. In this paper, the RSC model was used to predict the fiber orientation in LFTs.

The current approach to predict the elastic properties of a discontinuous fiber composite includes two steps. The first step computes the elastic stiffness of a “reference” composite that contains unidirectional (UD) fibers having the same features, volume fraction and length distribution as for the actual composite. In the second step, the stiffness of the actual as-formed composite is obtained from the stiffness of the reference composite that is averaged over all possible orientations using the orientation averaging technique [9-10]. There is a large body of literature addressing the computation of the elastic stiffness for the aligned fiber composite using micromechanical modeling or numerical simulations of a composite representative volume element. To date, the Eshelby’s equivalent inclusion method [11] combined with the Mori-Tanaka model (EMT) [12] is one of the most effective models in terms of accuracy and efficiency of the prediction [13-17]. This paper employs the EMT formulation developed in [16-17] to compute the stiffness of the UD composite that contains a fiber length distribution. The orientation averaging method [9-10] is then applied to compute the stiffness matrix of the actual LFT composite containing a fiber orientation distribution (FOD).

Modeling Approach to LFTs

This section describes the key steps in the computation of the elastic properties of a LFT composite that include a description of fiber length distribution, prediction of fiber orientation, and computation of the elastic properties.

Description of Fiber Length Distribution

Important fiber length attrition occurs during the injection molding process. As a consequence, the remaining average fiber length is much less than the initial length of the thermoplastic compounds used for injection-molding. There is a fiber length distribution (FLD) in the final composite that needs to be determined for property prediction. A common description of the length distribution is the probability density function (PDF) *f*(*l*) defined such that the probability of a fiber having a length between *l* and *l*+*dl* is *f*(*l*)*dl*. This function must be normalized such that

$$\int_0^\infty f(l)\,dl = 1 \tag{1}$$

From the raw FLD data, the discrete approximation of $f(l_i)$ is $\tilde{f}_i$ given by:

$$\tilde{f}_i = \frac{N_i}{\Delta l \sum_i N_i} \tag{2}$$

where Δl is the bin width, and a set of length values (bin centers) l_i such that $l_{i+1} = l_i + \Delta l$. The lengths l_i span the range of the data. N_i is the number of fibers with lengths between $l_i - \Delta l/2$ and $l_i + \Delta l/2$. An alternate FLD representation is a *probability density function for weight*, *w*(*l*) that is related to *f*(*l*) as:

$$w(l) = \frac{f(l)\,l}{\int_0^\infty f(l)l\,dl} \tag{3}$$

Similar to Eq. (2), the discrete approximation to $w(l_i)$ using the experimental/histogram data is

$$\tilde{w}_i = \frac{N_i l_i}{\Delta l \sum_i N_i l_i} \tag{4}$$

The measurement of fiber lengths in LFT samples shows that the actual fiber length distributions are unsymmetrical and exhibit a shape having a sharp peak in the short fiber range and a long “tail” towards the long fiber range (> 1mm). The two-parameter Weibull’s distribution function can be used to represent an experimental FLD. The Weibull’s probability density function is given by:

$$p(l) = \frac{c}{b}\left(\frac{l}{b}\right)^{c-1} e^{-\left(\frac{l}{b}\right)^c} \tag{5}$$

where *b* and *c* are the shape parameters.

Prediction of Fiber Orientation

The flow-induced fiber orientation is predicted by the RSC model described in [8]:

$$\frac{DA_{ij}}{Dt} + \frac{1}{2SRF}(\omega_{ik}A_{kj} - A_{ik}\omega_{kj}) = \frac{1}{SRF}[\frac{1}{2}\kappa(\dot{\gamma}_{ik}A_{kj} + A_{ik}\dot{\gamma}_{kj} - 2\dot{\gamma}_{kl}A_{ijkl}) + 2C_I\dot{\gamma}(\delta_{ij} - 3A_{ij})] \tag{6}$$

where A_{ij} and A_{ijkl} are the second and fourth-order orientation tensors, respectively. ω_{ij}

is the vorticity tensor, and $\dot{\gamma}_{ij}$ is the rate of the deformation tensor whose scalar magnitude is $\dot{\gamma}$. δ_{ij} is the identity tensor. κ and C_I are material constants; κ depends on the fiber aspect ratio, and C_I is called the interaction coefficient. *SRF* is an empirical factor that is introduced to slow down fiber rotation in concentrated fiber suspensions. A closure approximation is necessary to estimate A_{ijkl} from A_{ij}. Equation (6) was established for short-fiber systems and has been a first step to improve the Folgar-Tucker model in order to predict fiber orientation in concentrated suspensions. This paper assesses the applicability of Eq. (6) for fiber orientation prediction for LFTs.

Prediction of Elastic Properties

The stiffness matrix of a unidirectional (UD) fiber composite containing a fiber length distribution is given by:

$$C_{ijkl} = \frac{\int_0^\infty C^*_{ijkl}(l/d)\,p(l)\,dl}{\int_0^\infty p(l)dl} \tag{7}$$

where $C^*_{ijkl}(l/d)$ is the stiffness matrix of the UD fiber composite having the aspect ratio *l/d*. $p(l)$ is the probability density function that can be expressed for number or weight of fibers. In this paper $p(l)$ for weight of fiber lengths ($p(l) = w(l)$) is used in the calculation of the composite elastic properties. The Eshelby-Mori-Tanaka (EMT) method developed in [16-17] was applied to compute $C^*_{ijkl}(l/d)$, and after the calculation of the resulting C_{ijkl} using Eq. (7), the elastic stiffness of the actual composite having an as-formed fiber orientation state is computed using the orientation averaging method [9-10]:

$$\begin{aligned}\overline{C}_{ijkl} &= B_1 A_{ijkl} + B_2(A_{ij}\delta_{kl} + A_{kl}\delta_{ij}) + B_3(A_{ik}\delta_{jl} + A_{il}\delta_{jk} + A_{jl}\delta_{ik} + A_{jk}\delta_{il}) \\ &\quad + B_4\delta_{ij}\delta_{kl} + B_5(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})\end{aligned} \tag{8}$$

where the coefficients B_i (*i* = 1, 5) are related to the stiffness components of the UD transversely isotropic composite (Eq. (7)). These coefficients are given in [9-10].

Results and Discussion

Long glass fiber/polypropylene compounds were procured from Montsinger Technologies. Injection-molding was carried out for the center-gated disk and ISO-plaque geometries. The 13-mm pellets were injected using two different volume flow rates (16.4 cc/sec and 131.1 cc/sec) in order to study the effect of the injection speed on the composite microstructure. In this paper, a center-gated disk formed under fast fill conditions and an ISO-plaque injection-molded under slow fill conditions are analyzed. The weight fraction of glass fibers for both geometries is 40%. This corresponds to 19.2% fiber volume fraction. The center-gated disk is 3 mm thick and has a diameter of 177.8 mm. The ISO-plaque is also 3mm thick, and is 90 mm long and 80 mm wide. Three regions denoted as A, B, and C located at 6 mm, 34 mm, and 64 mm on the center-gated disk, and at 15 mm, 45 mm, and 75 mm on the ISO-plaque, respectively, were considered for fiber length and orientation measurements. A population of 2000 fibers was taken in each region for fiber length measurement [6]. Tensile specimens were cut from the injection-molded samples for mechanical testing in order to obtain the longitudinal and in-plane transverse moduli. In the ISO-plaque, tensile specimens were cut along the sample longitudinal and in-plane transverse directions while in the center-gated disk, the specimens were cut along two perpendicular radial directions.

Fiber Length Distribution Results

For the each type of specimens, similar FLD results have been obtained at the three above-mentioned regions. Therefore, the FLDs for the regions B are illustrated in this paper. Figures 1 and 2 show the FLDs in terms of number of fibers vs. fiber length and the probability density function for weight of fibers for the center-gated disk. These FLDs are the results corrected using a method explained in [6] to obtain an unbias length distribution from the raw data. This method considers the experimental sampling region specified by the diameter, *d*. On these figures are also presented the Weibull's fits. The FLD results for the ISO-plaque are illustrated in Figs 3 and 4. For both specimens, it is obvious that the Weibull's distributions for number of fibers can only represent the actual distributions in an average manner and cannot capture the peaks of these distributions. However, the Weibull's fits for the PDFs for weight of fibers capture the actual FLDs very well.

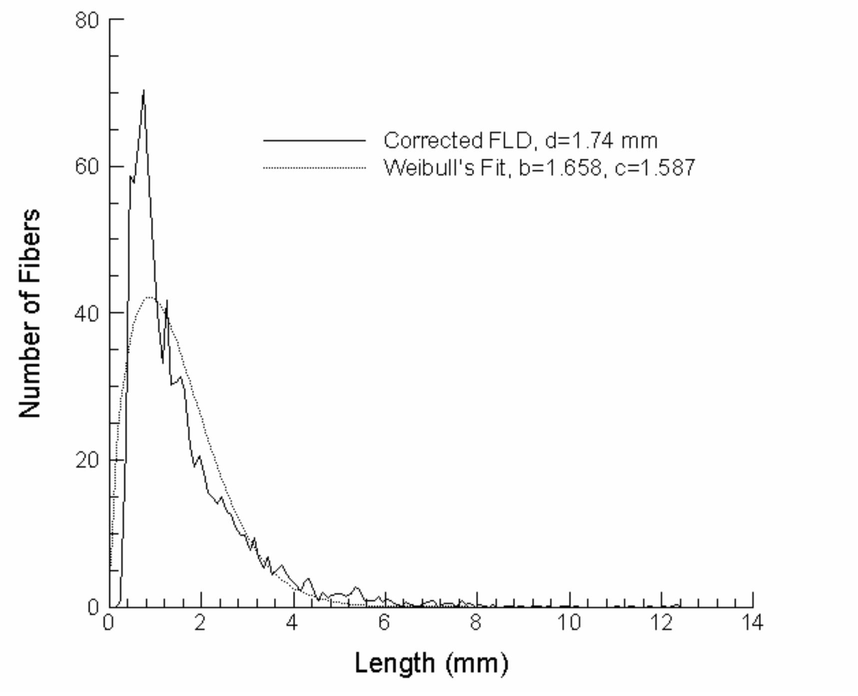

Figure 1: Corrected FLD in terms of number of fibers vs. fiber length for the center-gated disk.

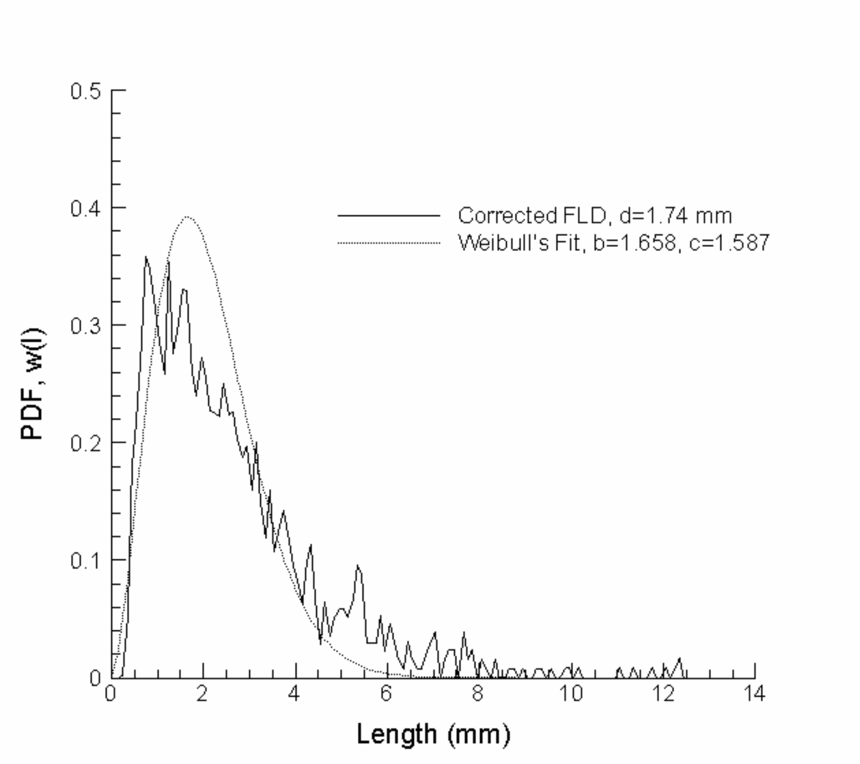

Figure 2: Corrected FLD in terms of PDF for weight of fibers vs. fiber length for the center-gated disk.

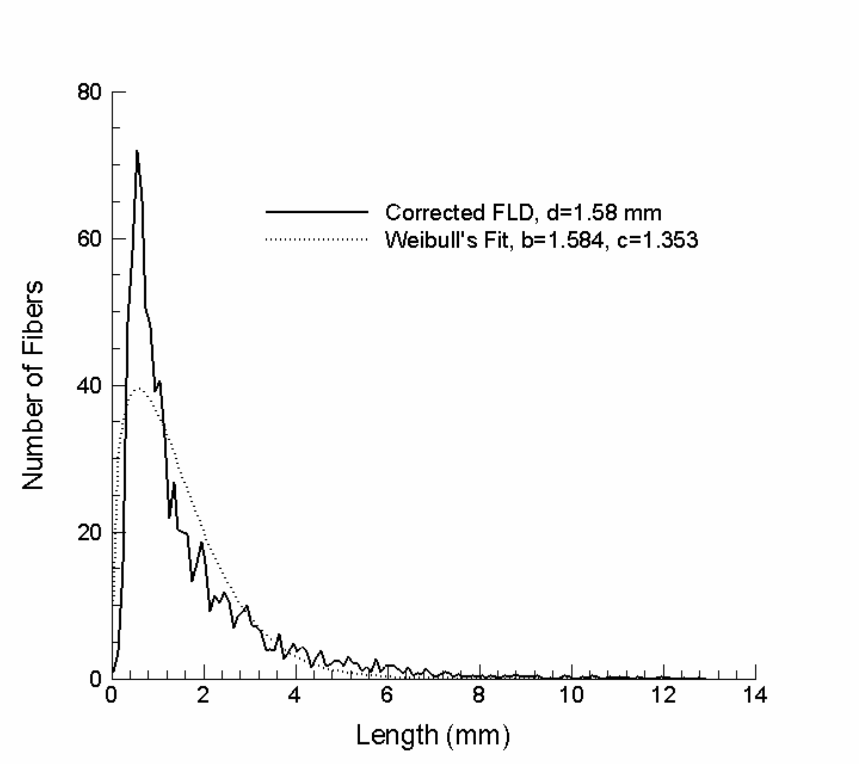

Figure 3: Corrected FLD in terms of number of fibers vs. fiber length for the ISO-plaque.

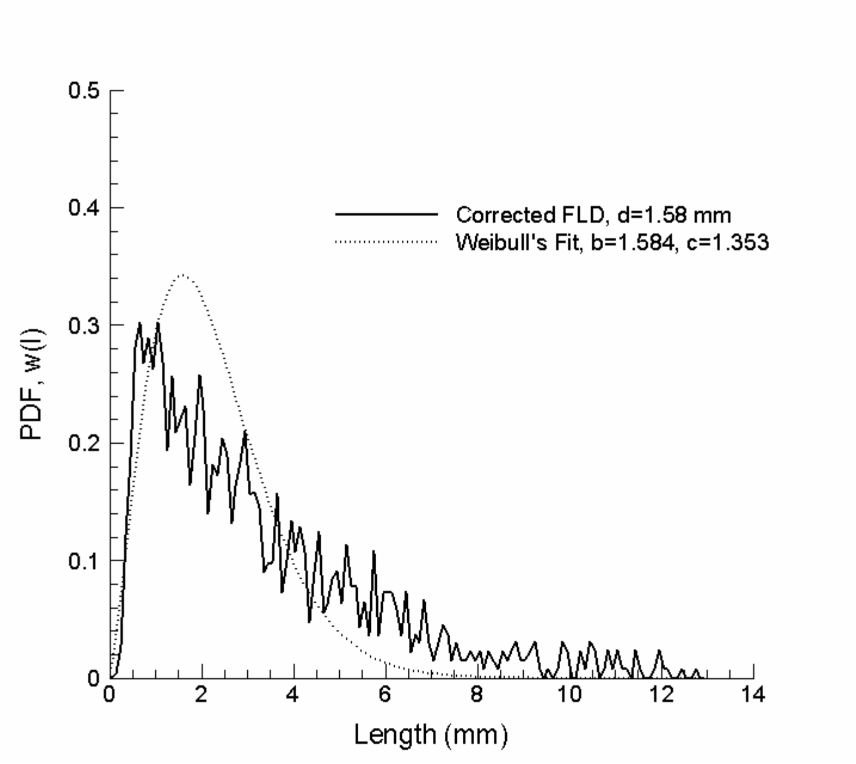

Figure 4: Corrected FLD in terms of PDF for weight of fibers vs. fiber length for the ISO-plaque.

Fiber Orientation Distribution Results

Figures 5 and 6 present the predicted and measured through-thickness values for the components of the second-order orientation tensor for the region B of the center-gated disk. The results for the ISO-plaque are given in Figs 7 and 8. The predictions were performed using the RSC model implemented in ORIENT, a finite difference code developed at the University of Illinois [18]. On these figures is presented the *z*-coordinate normalized by the specimen thickness *h*. The *SRF* and C_I parameters identified for both specimens are 30 and 0.03, respectively. The measurements were carried out using the method developed in [19]. The correlation of results shows that only the predictions for A_{11} are reasonably good while the A_{22} values are under-predicted, and this leads to the over-predictions of A_{33} since $A_{11} + A_{22} + A_{33} = 1$. A_{31} is poorly fit for both specimens.

Results for Elastic Properties

The elastic modulus of glass and polypropylene used in the computation are 73 GPa and 1.5 GPa, respectively. Poisson's ratios are 0.25 for glass fibers and 0.4 for polypropylene. The fiber length and orientation distributions given in Figs 2, 4-8 were used in the computation. The results for the center-gated disk and ISO-plaque are illustrated for the moduli E_{11} and E_{22} in Figs 9-12, respectively. As the Weibull's FLDs for weight capture the actual FLDs very well, the predictions using these Weibull's distributions are indistinguishable from those using the actual FLDs. Also, since the predictions for A_{11} are good, these result in good predictions for the modulus E_{11}.

However, the under-predictions for A_{22} lead to under-predicting E_{22}. Tables 1 and 2 provide the overall elastic properties predicted for the center-gated disk and ISO-plaque, respectively. The overall elastic properties of the specimen can be determined by considering the specimen as being formed by a stacking sequence of composite layers having the same thickness. Each layer is characterized by an orientation state (see Figs 5-8). The same tables also provide the results computed using the Weibull's FLDs and the experimental results. There are good correlations between the experimental results and the moduli computed based on the corrected experimental FLDs (Figs 2 and 4) and the experimental FODs. The use of the Weibull's FLDs and the experimental FODs also has provided good predictions. However, the use of the predicted FODs can only provide acceptable predictions for the modulus E_{11}. As previously mentioned, the less accurate prediction of A_{22} has not allowed to accurately predict the modulus E_{22}.

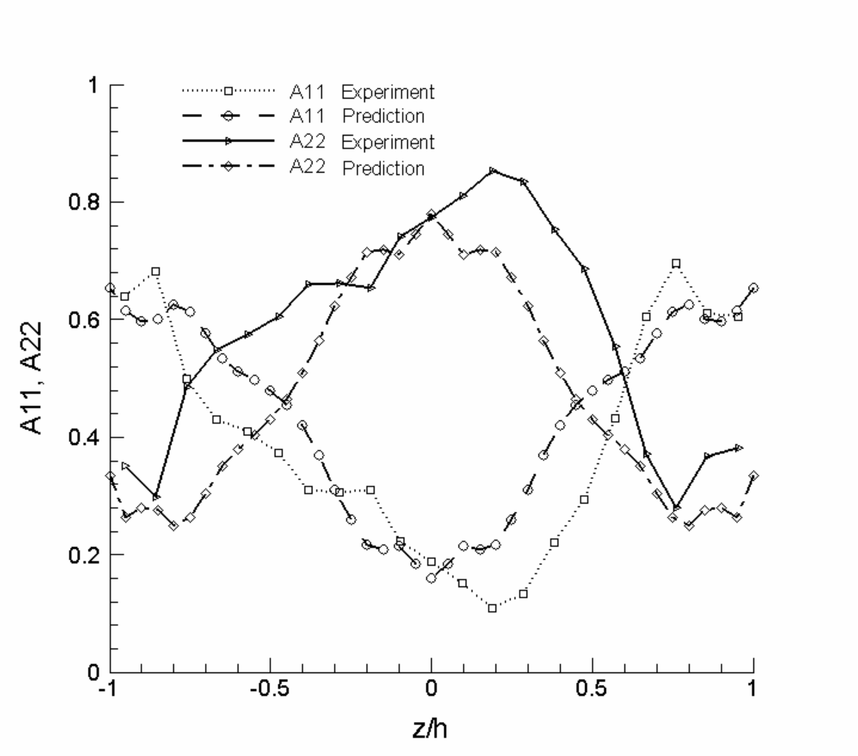

Figure 5: Predicted and measured orientation tensor components A_{11} and A_{22} for the center-gated disk.

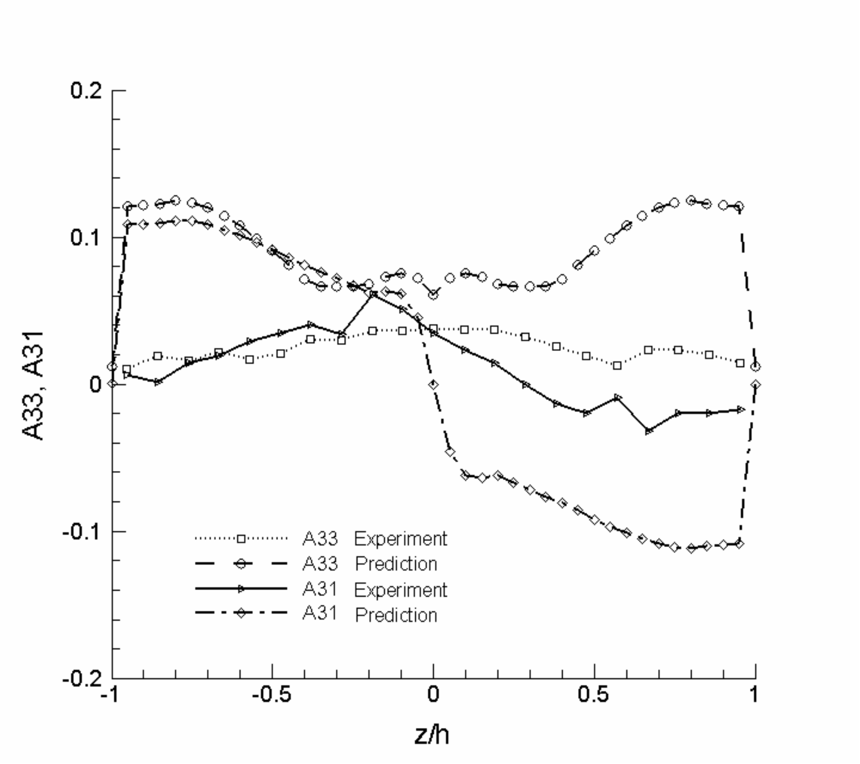

Figure 6: Predicted and measured orientation tensor components A_{33} and A_{31} for the center-gated disk.

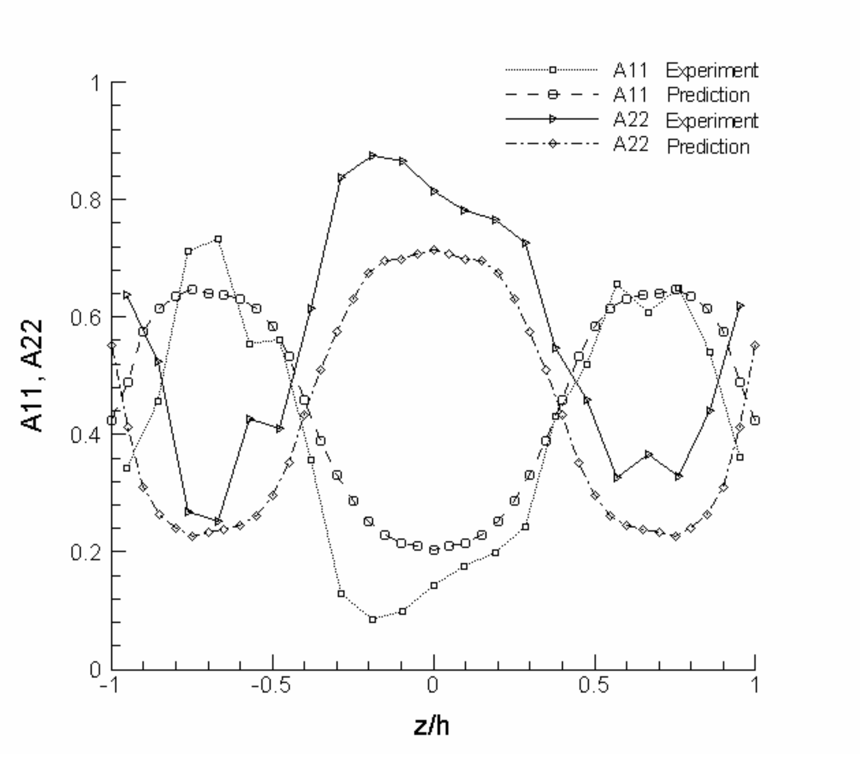

Figure 7: Predicted and measured orientation tensor components A_{11} and A_{22} for the ISO-plaque.

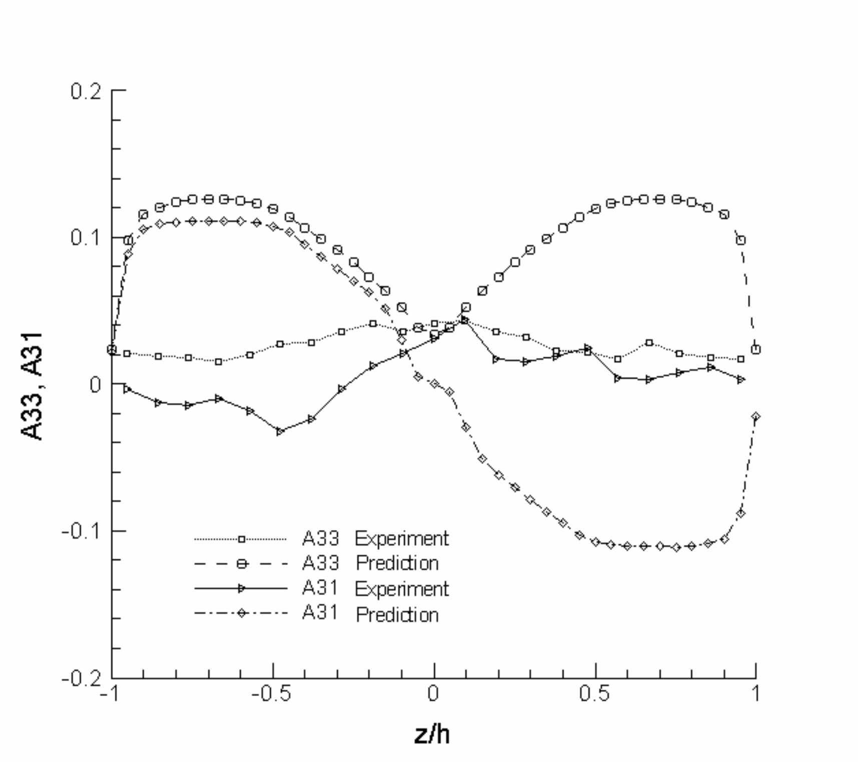

Figure 8: Predicted and measured orientation tensor components A_{33} *and* A_{31} *for the ISO-plaque.*

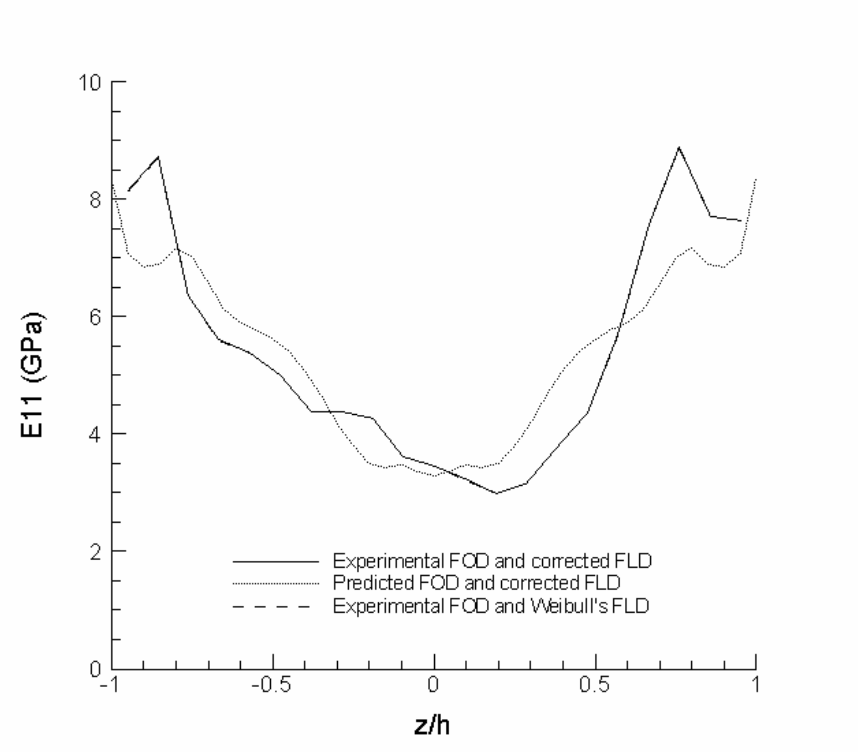

Figure 9: Predicted thru-thickness variation of the modulus E_{11} *for the center-gated disk.*

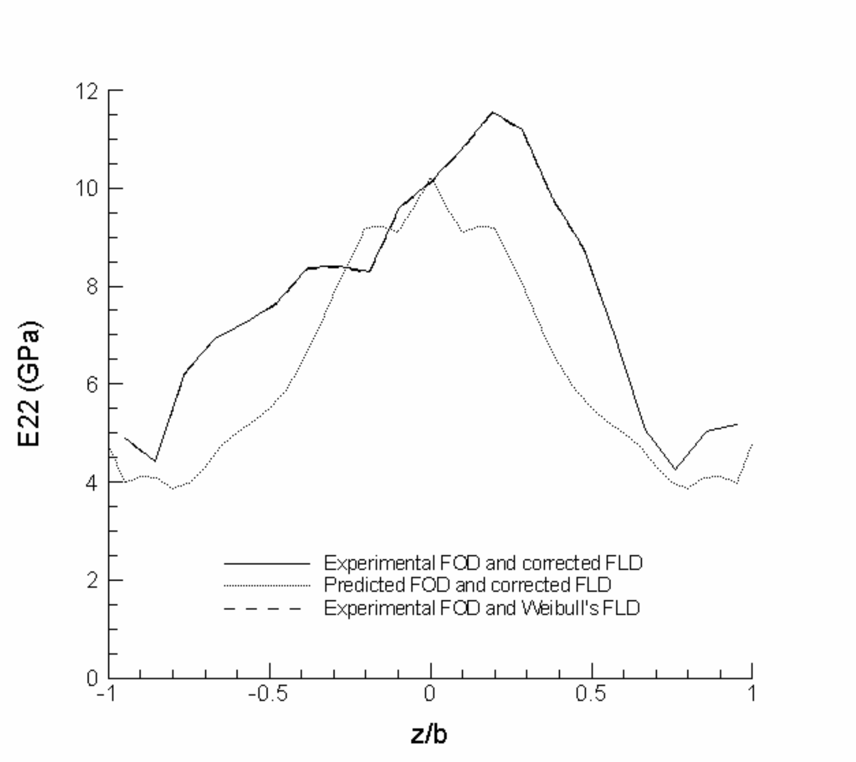

Figure 10: Predicted thru-thickness variation of the modulus E_{22} *for the center-gated disk.*

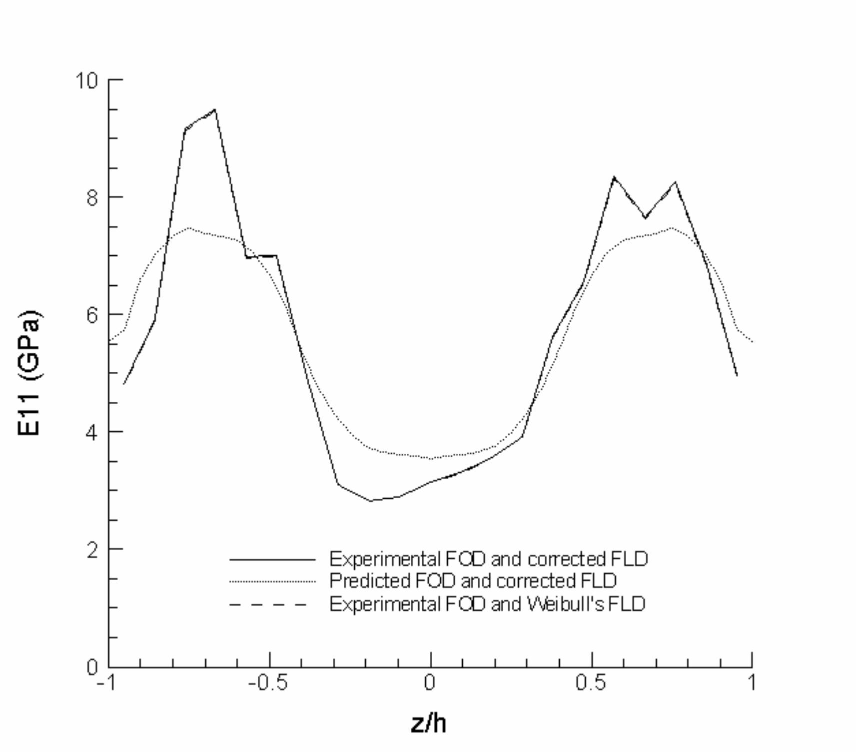

Figure 11: Predicted thru-thickness variation of the modulus E_{11} *for the ISO-plaque.*

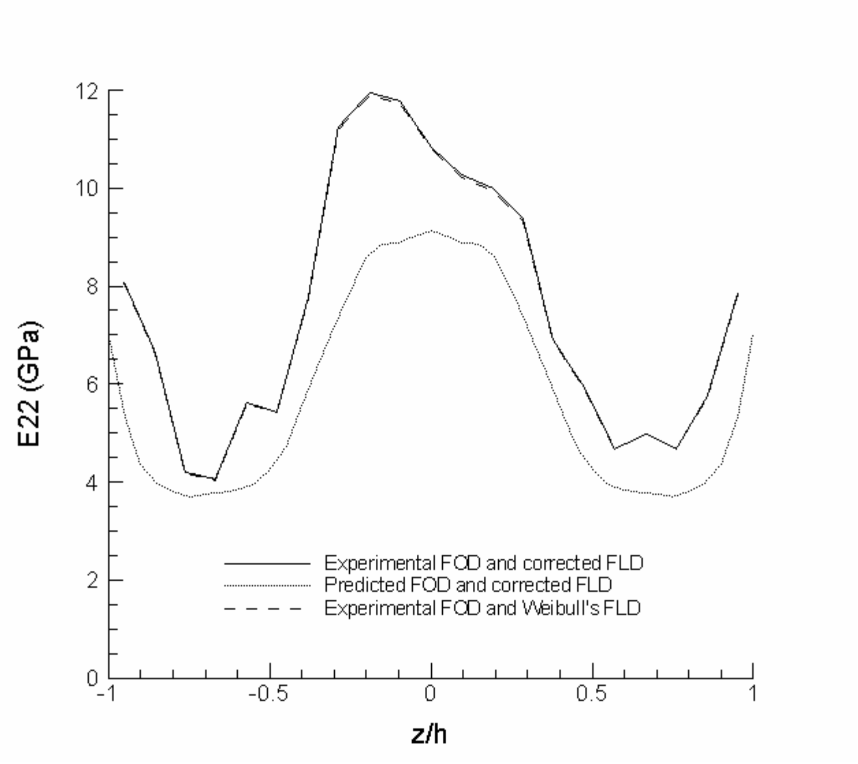

Figure 12: Predicted thru-thickness variation of the modulus E_{22} *for the ISO-plaque.*

Table I: Elastic properties for the glass/PP center-gated disk

Property (MPa)	Predictions (experimental FOD & FLD)	Predictions (Predicted FOD & experimental FLD)	Predictions (experimental FOD & Weibull's FLD)	Experiments
$\overline{E}_{11}$	5543	5730	5533	4737
$\overline{E}_{22}$	7703	6265	7686	6840
$\overline{E}_{33}$	3069	3127	3068	
$\overline{G}_{12}$	2123	2032	2120	
$\overline{G}_{13}$	951.4	1266	951.1	
$\overline{G}_{23}$	1014	1188	1014	

Table 2: Elastic properties for the glass/PP ISO-plaque

Property (MPa)	Predictions (experimental FOD & FLD)	Predictions (Predicted FOD & experimental FLD)	Predictions (experimental FOD & Weibull's FLD)	Experiments
$\overline{E}_{11}$	5786	5973	5766	5357
$\overline{E}_{22}$	7568	5874	7537	8012
$\overline{E}_{33}$	3085	3158	3082	
$\overline{G}_{12}$	2086	1998	2079	
$\overline{G}_{13}$	957	1321	957	
$\overline{G}_{23}$	1019	1202	1019	

Summary and Next Steps

This paper has developed a rigorous methodology to determine the elastic properties of long-fiber injection-molded thermoplastics. This methodology combines process modeling and micromechanical modeling based on the Eshelby-Mori-Tanaka model with experimental characterization of fiber length and orientation distributions. It has been shown that it is important to obtain an accurate fiber orientation distribution and a realistic fiber length distribution to accurately predict the composite properties. The current fiber orientation model cannot predict the fiber orientation in LFT samples to a level of accuracy needed for accurate property prediction. Therefore, research is ongoing to develop a fiber orientation model for LFTs. The developed methodology has been validated via experimental verification of the longitudinal and transverse moduli determined for long glass fiber injection-molded polypropylene specimens. The use of the corrected FLD for weight of fiber lengths and the experimental FOD has allowed reasonable predictions of the elastic properties for the studied glass/PP specimens. The Weibull's FLD can well capture the actual FLD for weight of fibers, thus the use of the Weibull's distribution for weight has led to practically the same results as those obtained using the actual FLD.

The next steps in our work will focus on (i) completing the development of a fiber orientation model for LFTs, (ii) linking process modeling to structural modeling for the analyses of LFT structures, and (iii) modeling the nonlinear behaviors due to damage and plastic deformation of LFTs.

Acknowledgement

This work has been funded by the U.S. Department of Energy's Office of FreedomCAR and Vehicle Technologies. The support by Dr. Joseph Carpenter Jr., Technology Area Development Manager, is gratefully acknowledged.

References

1. Truckenmüller, F., H.-G. Fritz. "Injection Molding of Long Fiber-Reinforced Thermoplastics: A Comparison of Extruded and Pultruded Materials with Direct Addition of Roving Strands." Polymer Engineering and Science, (1991) 31, pp. 1316-1329.
2. Silverman, E.M. "Effect of Glass Fiber Length on the Creep and Impact Resistance of Reinforced Thermoplastics." Polymer Composites, (1987) 8, pp. 8-15.
3. Henning, F., H. Ernst, R. Brussel, O. Geiger, W. Krause. "LFTs for Automotive Applications." Reinforced Plastics, (2005) 49, pp.24-33.
4. Nguyen, B.N., J.D. Holbery, M.T. Smith, V. Kunc, R.E. Norris, J. H. Phelps, C.L. Tucker III. "Assessment of Current Process Modeling Approaches to Determine Their Limitations, Applicability and Developments Needed for Long-Fiber Thermoplastic Injection-Molded Composites." (2006) Internal Report, PNNL-16236.
5. Nguyen, B.N., V. Kunc, B.J. Frame, J.H. Phelps, C.L. Tucker III, S.K. Bapanapalli, J.D. Holbery, M.T. Smith. "Effects of Fiber Length and Orientation Distributions on the Elastic Properties of Long-Fiber Injection-Molded Thermoplastics: Part I – Property Prediction." (2007) submitted to Journal of Composite Materials.
6. Kunc, V., B.J. Frame, B.N. Nguyen, C.L. Tucker III, G. Velez-Garcia. "Fiber Length Distribution Measurement for Long Glass and Carbon Fiber Reinforced Injection Molded Thermoplastics." (2007), Proceedings of the 2007 SPE Automotive Composites Conference & Expositions (ACCE).
7. Folgar, F., C.L. Tucker III. "Orientation Behavior of Fibers in Concentrated Suspensions." Journal of Reinforced Plastic Composites, (1984) 3, pp. 98-119.
8. Phelps, J.H., C.L. Tucker III. "Assessing Fiber Orientation Prediction Capability for Long-Fiber Thermoplastic Composites." Technical Report (2007), University of Illinois at Urbana-Champaign, Urbana, IL 61801.
9. Advani, S., C.L. Tucker III. "The Use of Tensors to Describe and Predict Fiber Orientation in Short-Fiber Composites." Journal of Rheology, (1987) 31, pp. 751-784.
10. Camacho, C., C.L. Tucker III, S. Yalvac, R.L. McGee (1990). "Stiffness and Thermal Expansion Predictions for Hybrid Short-Fiber Composites." Polymer Composites, (1990) 11, pp. 229-239.
11. Eshelby, J.D. "The Determination of the Elastic Field of An Ellipsoidal Inclusion and Related Problems." Proceedings of the Royal Society London, (1957) A 241, pp. 376-396.
12. Mori, T., K. Tanaka. "Average Stress in Matrix and Average Elastic Energy of Materials with Misfitting Inclusions." Acta Metallurgica, (1973) 21, pp. 571-574.
13. Benveniste, Y. "A New Approach to The Application of Mori-Tanaka's Theory in Composite Materials." Mechanics of Materials, (1987) 6, pp.147-157.
14. Tucker III, C.L., E. Liang. "Stiffness Predictions for Unidirectional Short-Fiber Composites: Review and Evaluation." Composites Science and Technology, (1999) 59, pp. 655-671.
15. Hine, P.J., H.R. Lusti, A.A. Gusev. "Numerical Simulation of the Effects of Volume Fraction, Aspect Ratio and Fiber Length Distribution on the Elastic and Thermoelastic Properties of Short Fiber Composites." Composites Science and Technology, (2002) 62, pp.1445-1453.

16. Taya, M., T. Mura. “On Stiffness and Strength of An Aligned Short-Fiber Reinforced Composite Containing Fiber-End Cracks under Uniaxial Applied Stress.” Journal of Applied Mechanics, (1981) 48, pp. 361-367.

17. Chen, C.-H., C.-H. Cheng. “Effective Elastic Moduli of Misoriented Short-Fiber Composites.” International Journal of Solids and Structures, (1996) 33, pp. 2519-2539.

18. Bay, R.S., C.L. Tucker III. “Fiber Orientation in Simple Injection Moldings. Part I: Theory and Numerical Methods.” Polymer Composites, (1992) 13, pp. 317-331.

19. Hine, P.J., N. Davidson, R.A. Duckett, A.R. Clarke and I.M. Ward. “Hydrostatically Extruded Glass-Fiber-Reinforced Polyoxymethylene. I: The Development of Fiber and Matrix Orientation.” Polymer Composites, (1996) 17, pp. 720-729.