PROPERTY CALCULATION SYSTEM FOR INJECTION MOLDING AND COMPRESSION MOLDING OF FIBER-FILLED POLYMER COMPOSITES

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1 Abstract
This paper presents property calculation system for injection molding and compression molding simulations of short and long fiber-filled polymer composites. It also discusses its importance in molding induced residual stress calculation and warpage prediction, as well as in the subsequent stress analysis for molded plastic parts. The composite property calculation system consists of fiber orientation calculation, fiber breakage calculation, and micro-mechanics model. Major differences between short fiber and long fiber composite property calculations include the non-uniform fiber length distribution across injection molded parts and possible de-bonding between fiber and matrix. They are addressed by a recently implemented micro-mechanical model specific to long fiber composites, and use of calculated long fiber orientation and fiber length distributions. In this paper, case studies on fiber orientation distribution (FOD), fiber length distribution (FLD) and subsequent fiber-filled composite property distributions for short and long fiber-filled composites are given for injection molding and compression molding simulations.

2 Introduction
Discontinuous glass and carbon fibers are commonly added to injection molded plastic materials to reinforce the mechanical and thermal properties with small change of weight. The addition of fibers introduces anisotropy of the properties mainly due to the preferential orientation of fibers induced by the flow during the injection molding process. It is typical with composite materials that quantitative evaluation of the anisotropic thermo-mechanical properties is essential in the part designs and applications. Furthermore, these properties are non-uniformly distributed for injection molding or compression molding processes.

Computer simulation of injection molding of fiber-filled polymer composites has been available for industrial applications for quite some time. Recently a three-dimensional compression molding simulation has been developed [1] to meet the application demands, in particular, the need for simulating Direct Strand Deposition (DSD) compression molding for relatively large flat or moderately curved parts. Although compression molding has been mainly used for processing thermoset resins, it can also be employed to process thermoplastics materials, especially for long fiber-filled thermoplastics (LFT) because it produces much less breakage of long fibers than injection molding process.

The discontinuous fiber-filled composite property calculation needs correctly predicted as-formed microstructure due to the polymer flow and solidification in the molding process. Fiber orientation distribution (FOD) and fiber length distribution (FLD) are major factors for the composite property calculation. Another major aspect is the mechanics modeling and the fiber-matrix micro-structure. Perfectly bonded or de-bonded fiber-matrix in the micro-mechanics models have been used for these fiber-filled composite property calculations. The fiber-matrix interface in reality, for long fiber in particular, can involve quite a few factors that affect the adhesion between fiber and matrix in different situations. Therefore this paper presents fiber orientation models evolved from short fiber-filled orientation to long fiber-filled polymer composites, a fiber breakage model, and a de-bonding micro-mechanics model that addresses the fiber-matrix adhesion factor and is applied to the long fiber composite property calculation.
3 Fiber Orientation Models
A concise description of fiber orientation is the second-order orientation tensor [2], which is defined as

$$A = \langle pp \rangle$$  \hspace{1cm} (1)$$

where the unit vector $p$ directs along the fiber length, and the bracket “$\langle \rangle$” is the average over a volume domain. The trace of $A$, $\text{tr}A$, is unity due to the unit length of $p$ and the normalization condition.

The Folgar-Tucker model in the term of the orientation tensor $A$ is [3]:

$$\frac{DA}{Dt} = (W \cdot A - A \cdot W)$$
$$+ \xi [D \cdot A + A \cdot D - 2A : D]$$
$$+ 2C_i \dot{\gamma}(I - 3A)$$ \hspace{1cm} (2)$$

Here, $DA/Dt$ is the material derivative of $A$; $W$ and $D$ are the vorticity and the rate-of-deformation tensors, respectively; $\xi$ is the particle shape parameter and is very close to unity due to the large length-to-radius ratio of fibers; $C_i$ is the interaction coefficient, and a larger $C_i$ implies more fiber-fiber interactions; and $\dot{\gamma}$ is the scalar magnitude of $D$. $A$ is the fourth-order orientation tensor, and is approximated as a closure function of components of $A$.

The reduced strain closure (RSC) model introduces a scalar factor $\kappa < 1$ to slow the fiber orientation kinetics, and the equation is given as [4,5]:

$$\frac{DA}{Dt} = (W \cdot A - A \cdot W)$$
$$+ \xi [D \cdot A + A \cdot D - 2A : D]$$
$$+ 2C_i \dot{\gamma}(I - 3A)$$
$$+ 2\kappa C_i \dot{\gamma}(I - 3A)$$ \hspace{1cm} (3)$$

Here, the fourth-order tensors $L$ and $M$ are functions of the eigenvalues $\lambda_i$ ($i = 1,2,3$) and the eigenvectors $e$, ($i = 1,2,3$) of the orientation tensor $A$, defined as $L = \sum_{i=1}^{3} \lambda_i e e e$, and $M = \sum_{i=1}^{3} \lambda_i e e e$. The scalar factor $\kappa$ is a phenomenological parameter, and its value is determined by fitting the fiber orientation or rheology prediction to experimental data. Setting $\kappa = 1$ reduces the RSC model of Eq. (3) to the Folgar-Tucker model of Eq. (2).

For a long fiber orientation, Phelps and Tucker [6] proposed an anisotropic rotary diffusion (ARD) term that describes the hydrodynamic fiber-fiber interaction in LFT systems, combined with the RSC procedure mentioned above to slow down the orientation kinetics. This combined ARD-RSC model is written as

$$\frac{DA}{Dt} = (W \cdot A - A \cdot W) +$$
$$\xi [D \cdot A + A \cdot D - 2A + (1 - \kappa)(L \cdot M : A) : D]$$
$$+ \gamma [C + (I - \kappa)M : C - 2\kappa(\text{tr}C)A]$$
$$- 5(C \cdot A + A \cdot C) + 10(A + (1 - \kappa)(L \cdot M : A) : C)$$ \hspace{1cm} (4)$$

Here, the rotary diffusion tensor $C$ is constructed from $A$ and $D$ as:

$$C = b_1 I + b_2 A + b_3 A^2 + b_4 \frac{D}{\dot{\gamma}} + b_5 \left(\frac{D}{\dot{\gamma}}\right)^2$$ \hspace{1cm} (5)$$

where $b_i$ ($i = 1, \ldots, 5$) are scalar constants and selected by matching experimental steady-state orientation and requiring stable orientation. Setting $b_1 = C_i$ and $b_i = 0$ ($i = 2, \ldots, 5$) reduces the ARD-RSC model of Eq. (4) to the RSC model of Eq. (3).

4 Fiber Breakage Model
Phelps and Tucker also developed a fiber length attrition model to predict fiber length distribution [6,7].

$$\frac{F_i}{F_{\text{crit}}} = \frac{8D \gamma \rho^2}{\pi^3 E_j d_f^4} (-D : A) > 1$$ \hspace{1cm} (6)$$

First, using a model by Dinh and Armstrong [8], an expression for the hydrodynamic force acting along the fiber axis is obtained. The condition for buckling that leads to fiber breakage compares this hydrodynamic force to the buckling force from the classical Euler buckling theory. The probability of a single fiber breakage can be expressed as:

$$P_i = C_b \dot{\gamma} \max \left\{0, \left[1 - \exp \left(1 - \frac{F_i}{F_{\text{crit}}} \right) \right] \right\}$$ \hspace{1cm} (7)$$

where $C_b$ is a scalar rate constant which can be used as a fitting parameter for scaling the shearing influence.

It can be seen that the buckling criterion dominates this probability form. If it is less than one, $P_i$ is zero meaning that there is no chance to break. If Equation (6) is true, the probability $P_i$ should increase monotonically as $F_i/F_{\text{crit}}$ and $\dot{\gamma}$ increase. The shear rate $\dot{\gamma}$ introduced in this scalar function is an additional force in the slow field for fiber breakage, and it is found as an important factor in matching the pattern of fiber length distribution (FLD) with reality.
The fibers, broken or not, have to follow a conservation law because they cannot disappear nor can they grow in a flow field. This conservation law can be expressed as:

\[
\frac{\partial N(l, t)}{\partial t} + \vec{v} \cdot \nabla N(l, t) = \nabla \cdot (g(l) \nabla N(l, t)) \tag{8}
\]

where \( L \) is the maximum initial fiber length, \( N(l, t) \) is the number of fibers with length \( l \) at time \( t \), \( g(l) \) is the scalar probability function of breaking a fiber of length \( l \), and \( R(l, l') \) is the probability function of a fiber of length \( l' \) breaking to form a fiber of length \( l \) (where \( l < l' \)) and another one of length \( l' - l \) in time \( t \). It can be expressed as a Gaussian breaking profile, such as:

\[
R(l, l') = G_{norm} \left( \frac{l'}{2}, S \right) \tag{9}
\]

where \( G_{norm}() \) is the Gaussian normal probability density function for the variable \( l \) with mean \( l'/2 \) and standard deviation \( S \). \( S \) is a dimensionless fitting parameter that can be used to control the shape of the Gaussian breaking profile. The range of this parameter is from 0 to 1, and a small value corresponds to the probability of breakage occurring exactly at the center of the length \( l' \), while a large value distributes the probable breakage points more evenly along the fiber length.

This fiber breakage model has three adjustable parameters: \( D_p \), \( C_b \), and \( S \). \( D_p \) can be considered as a parameter that determines the minimum length of fiber that cannot be broken, \( C_b \) is a breakage rate parameter, and \( S \) is a breakage probability profile shape parameter. It can be seen in the validation section that all of these have been used as fitting parameters.

5 Long fiber composite property models

Micro-mechanics and orientation averaging models are used in the thermo-mechanical property calculation for fiber-filled materials. The basic method adopted in micro-mechanical level modeling with an inclusion is the method proposed by Mori-Tanaka [9]. The part of this model is based on earlier work by Eshelby [10] on equivalent inclusion. Benveniste [11] later on proposed a complete tensor form of the Mori-Tanaka theory, and this tensor form is implemented as the first step of the fiber-filled composite property calculation in this paper.

The imperfect fiber-matrix interface model by Qu [12] is adopted for the expression of the stiffness tensor for a unidirectional composite that contains partially debonded fiber/matrix interfaces. The Eshelby-Mori-Tanaka formulation for the unidirectional composite is modified as follows:

\[
\begin{bmatrix} S_1 \end{bmatrix} = \begin{bmatrix} S_x \end{bmatrix} + \sum_{i} \left( v_i v_i T \right) \left[ I + v_i T + v_i LS \right]^{-1} \tag{10}
\]

in which \( S_x \) is the fourth-rank effective stiffness tensor of the two-phase composite, \( S_x \) and \( S_2 \) are the stiffness tensors of the matrix and inclusion, respectively, \( v_i \) and \( v_2 \) are volume fractions of the two phases. The Eshelby tensor \( E \), which depends on the aspect ratio of the inclusion, is used to derive the following tensor:

\[
T = \left[ I + E \right] \left[ \begin{bmatrix} S_x \end{bmatrix} - \begin{bmatrix} S_2 \end{bmatrix} \right]^{-1} \tag{11}
\]

A modified Eshelby tensor \( E' \) is given as

\[
E' = E^{\phi} \left( I - ELS \right) \left( I - E \right) \tag{12}
\]

in which the fourth-order tensor \( L \) is given by

\[
L = \phi' \left( P - Q \right) \tag{13}
\]

where

\[
P_{ijkl} = \frac{3}{16 \pi} \int_0^r \left[ \delta_{ij} \delta_{kl} \nabla \nabla \delta_{ij} + \delta_{ik} \delta_{jl} \nabla \nabla \delta_{ij} \right. \\
+ \delta_{jl} \delta_{ij} \nabla \nabla \delta_{ij} + \delta_{ik} \delta_{jl} \nabla \nabla \delta_{ij} \right] d \theta \sin \phi \phi d\theta d\phi \tag{14}
\]

\[
Q_{ijkl} = \frac{3}{4 \pi} \int_0^r \left[ \delta_{ij} \delta_{kl} \nabla \nabla \delta_{ij} - \delta_{ik} \delta_{jl} \nabla \nabla \delta_{ij} \right. \\
+ \delta_{jl} \delta_{ij} \nabla \nabla \delta_{ij} + \delta_{ik} \delta_{jl} \nabla \nabla \delta_{ij} \right] d \theta \sin \phi \phi d\theta d\phi \tag{15}
\]

where \( r \) is the radius of the fiber. \( \beta \) represents the compliance in the tangential direction of the interface, and can be expressed in terms of the matrix elastic modulus. \( \beta = 0 \) corresponds to a perfect fiber/matrix interface while an infinite value of \( \beta \) represents a completely debonded fiber-matrix interface.

Based on the orthotropic assumption, a nine-parameter averaging approximation model was proposed by Jin and Lin [13] as follows:

\[
\begin{align*}
\langle S \rangle_{ijkl} &= B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) \\
&+ B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right) + B_x \left( a_{ijkl} \right)
\end{align*} \tag{16}
\]
This orientation averaging model is used if all nine parameters can be determined with two theoretical micro-mechanics models. Otherwise, Advani-Tucker’s five parameter orientation averaging model [2] is used.

Due to the fiber breakage which occurs during the molding process, the fiber length distribution is not uniform. The length averaging scheme can be described with the probability density function \( p(l) \), that is, the stiffness of the composite is

\[
S = \frac{\int_0 S^*(l)p(l)dl}{\int_0 p(l)dl}
\]

(17)

However, the stiffness tensor \( S^* \) needs to be calculated for each length segment \( l_i \), which requires a lot of computation. To simplify the computation, the average fiber length by weight, \( l_{w} \), is used in Equation (10) and (11) to calculate the stiffness. The averaging scheme mentioned in the previous section is then employed to calculate the composite stiffness averaged over the fiber orientation distribution.

The thermal expansion coefficient tensor is calculated with the Rosen-Hashin model [14] which was derived based on a connection to the effective elastic properties shown by Levin [15], and extended to general anisotropic phases. Later on, it was proven by Benveniste-Dvorak [16] that the Rosen-Hashin model has an important consistency property with the Mori-Tanaka principle in the context of thermo-mechanical problems. Benveniste-Dvorak tensor equivalent form of Rosen-Hashin model is implemented for the long fiber composite thermal expansion coefficient calculation, it is written as:

\[
\alpha_{xy} = \alpha_1 + (\alpha_1 - \alpha_2)(C_2 - C_1)^{-1}(C_1 - C_x)
\]

(18)

where \( \alpha_1 \) and \( \alpha_2 \) are the matrix and inclusion thermal expansion coefficient tensors, \( C_1 \) and \( C_2 \) are the compliance tensors of the matrix and the inclusion, respectively, and \( C_x \) the effective compliance tensor which can be calculated from the effective stiffness tensor \( S_e \) from equation (10).

6 Examples

6.1 Injection Molding

The first example is an injection molding simulation of an end-gated ISO plaque with 50% long glass fibers (LGF) in PA 66 (DuPont Zytel 75LG50HSL BK031). This was one of the validation cases documented in U.S. Department of Energy Energy Efficient & Renewable Energy website [17]. The dimension of the ISO plaque is 12 in. x 12 in. x 0.11 in with thinning-down thickness of the fan gate, and three fiber orientation measuring locations marked in green. Fiber lengths at inlet (light blue) and at center (coincided location with the center green bar) are measured as shown in Fig. 1.

Because the tensile specimens based on modified ASTM638 Type I standards were cut along the flow and cross flow directions at the center location from this ISO plaque, the stress-strain curves measured from these tensile specimens are the responses that can be seen as averaged responses across the tensile bar cross-section. A single point comparison with a predicted value would not be enough. In this regard, the tensile specimens are meshed and built into the plaque mesh as shown in Fig. 2.
Process conditions used for the analyses are:

- Fill time: 1.03 sec.
- Mold surface temperature: 85°C
- Melt temperature: 295°C
- Cooling time: 20 sec.

The fiber orientation distributions (FOD) for the three green locations shown in Fig. 1 were first compared to make sure the fiber orientation predictions are in good agreement with experimental measurements. The three locations’ cross-thickness profile comparisons in Figs. 3, 4 and 5 show that the fiber orientation results from mid-plane mesh are very consistent with those predicted with dual domain mesh, and they both are in good agreement with the experiments.

The fiber length distribution (FLD) predictions are the next to check. The fiber length predictions are also very dependent on the inlet conditions. Fig. 6 shows the fiber length inlet condition used for the calculation based on provided fiber length measurement at the inlet. Fig. 7 shows the experimental and calculated fiber length probability distributions at the center of the plaque. Please note that the fiber length for the first data point in Fig. 6 is non-zero, while that in Fig. 7 is zero. Therefore, the probability of first data point in Fig. 6 is non-zero, while that in Fig. 7 is zero.
As already prepared in the mesh models, the tensile specimens have been finely meshed and built into the plaque mesh in the same dimensions and locations as they were cut along the flow and cross flow directions from the ISO plaque. The idea is to give a proper comparison for the mechanical properties between experiments and predictions. Fig. 8 shows the debonding factor influence on the principal moduli. A few values have been tried before settling down to 0.5 which gives a reasonable match in the mechanical property predictions, as shown in the result pictures. From Fig. 9, it can be seen that tensile specimens can be taken out at exact locations with exact dimensions, with the predicted modulus distributions on them, as the experimental specimens for comparison purpose. Further examination with per frame range plot is shown in Fig. 10. We can see a clear range and distribution over each of the tensile specimen in its intended tensile modulus plot. These plots indicate that the tensile specimens in examination are actually averaged over a range, so the comparison cannot be done with a single value at a particular location in the ISO plaque. Rather, it must be done with the same way as the specimens were done in a tensile test machine.

Another point must be made that the principal directions vary in space even inside the specimens. Even though the specimens were cut along the flow and cross flow directions, they do not necessarily give the same slope between stress and strain as in the first principal modulus value. Therefore, only using the first principal modulus values to do the average for comparison is still just an approximation.

The response of the stress vs strain in a structural analysis of the tensile specimens with predicted set of stiffness tensor distribution over the tensile specimen is the right way to do the comparison.

Fig. 8. Predicted tensile moduli in the center of ISO plaque with different debonding factors.

Fig. 9. Predicted tensile moduli in the first (top) and the second (bottom) principal direction, with tensile specimens cut out from the plaque at the same locations and with the same dimensions as in experiments.

Fig. 11 shows the stress-strain curves obtained by Oak Ridge National Laboratory (ORNL) during the CRADA project on this sample. The initial slopes taken from these curves were averaged to be 16,100 MPa and 11,400 MPa, respectively, even though the overall stress-strain curves represent a clear nonlinear behavior. It can be readily seen that these experimental values fall into the predicted tensile specimen ranges in Fig. 11.
long glass fiber). This case is from Premix Inc. The initial fiber length is 12mm. The 3D mesh model of the final plate (300x300x2 mm) is shown in Fig. 12. The initial charge (300x75x12.7 mm) is placed along the left side with the intention to create a close to unidirectional fiber orientation by the compression molding process. The tensile specimens (200x25 mm) are cut from the plate in both X and Y directions, and they are highlighted in Fig. 12 as red and brown color, respectively. The fibers in the initial charge are assumed to have 2D random orientation at the start of molding. The calculated fiber orientation result is shown in Fig. 13. It shows that the fiber orientation is predominantly along the X direction because the resin mainly flows in X direction. The predicted distributions of tensile moduli are shown in Fig. 14. The tensile specimens taken out to be further scaled according to the range per frame are shown in Fig. 15.

Fig. 10. Predicted tensile modulus distributions inside tensile specimens for E1 (top) and E2 (bottom) with range scaled per frame.

Fig. 11. ORNL provided stress-strain curves for PA66 50% LGF tensile specimens [17].

6.2 Compression Molding
The second example is a pure compression molding of a long fiber filled thermoset material (with 34%
Fig. 13. Fiber orientation prediction (which shows alignment to the X direction in later stage of molding).

Fig. 14. Predicted tensile modulus distributions in the first principal direction (top) and in the second principal direction (bottom). The tensile bars of testing specimens are taken out for illustration purpose.

Fig. 15. Tensile modulus distribution in the tensile testing specimen in X direction (top) and in Y direction (bottom).

The experimental values of tensile modulus are 16 GPa for the X direction, and 12.4 GPa for the Y direction. On the other hand, the calculated average values for the tensile modulus are 16 GPa and 11 GPa in X and Y direction, respectively. The experimental and simulation values compare reasonably well.

7 Concluding Remarks
Following on from the long fiber orientation and long fiber breakage model implementations in the past, long fiber polymer composite property calculation models have been implemented for long fiber-filled injection moldings in this paper. Major differences between short fiber and long fiber composite property enhancements include the non-uniform fiber length distribution across injection molded parts and possible de-bonding between fiber and matrix. The recent implementation of micro-mechanics model by Qu provides a debonding coefficient to fit measurement results [12]. For the two study cases shown in this report, a debonding coefficient of 0.5 was used.

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9 References