K. Engineering Property Prediction Tools for Tailored Polymer Composite Structures

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Objective

- Enable the optimum design of lightweight automotive structural components using long-fiber-reinforced injection-molded thermoplastic composites (LFTs).

Approach

Phase I: Technical-feasibility assessment

- Perform a technical assessment of current process models and computational methods for short-fiber polymer injection-molded composites in order to determine their accuracy, limitations and potential applications to LFTs.

- Identify the research directions needed to develop process and constitutive models for LFTs.

- Understand and examine the LFT emerging microstructure, characterization techniques and material processing.

- Characterize LFT parts for both fiber orientation and attrition by applying x-ray tomography, automated methods utilizing advanced microscopy combined with polymer matrix etching, and ultrasonic inspection.

- Examine the molding behaviors of glass and carbon fibers.

Phase II: Research and development

- Develop an integrated approach that links process to structural modeling allowing the optimum design of LFT composite structures.

- Establish experimental methodologies for material characterization and testing to develop material property and performance databases and to verify modeling predictions.

Accomplishments

- The technical assessment of current process models has pointed out that the fiber-orientation equation and constitutive relation for short-fiber systems needs to be improved to account for the features of long-fiber systems which are characterized by limited fiber mobility, collective movement of fiber domains, fiber attrition, and bending of fibers.

- An approach to the fiber-suspension flow problem using the smoothed particle hydrodynamics method was formulated.

- Numerical assessment of current process modeling capability was performed for a center-gated disk and an end-gated plaque.

- The project kickoff meeting was organized at the USCAR headquarter on September 1st, 2005. A steering committee composed of Automotive Composite Consortium, American Plastics Council, and National Labs members was formed to follow this project.

- Identified several methods of non-destructively and destructively characterizing LFT composite fiber orientation, and have experimented and conducted proof-of-concept verification of these techniques.

- Identified a specialty material supplier and arranged procurement of materials for molded parts.

- Have identified and secured Tier 1 supplier commitment for molding LFT plaques and test articles for feasibility program. Have begun process of Tier 1 supplier commitment to the full project.

- Have secured commitment with LFT material supplier, both for the feasibility and full program.

- Completed material mold evaluation, identified US manufacturer, obtained mold quotation, and completed investigation of adaptation of PNNL Aarborg 350 injection-molding machine to mold LFT materials.

Future Direction

- The technical feasibility assessment phase will be completed by March 2006 that includes the completion of the assessment of process models and available computational tools to perform the work, the establishment of an experimental methodology, and research directions to develop process models for LFTs,

- Develop a material performance matrix to obtain material parameters for model input.

- Begin developing process and property prediction models for LFTs from April 2006.

- Further develop non-destructive techniques to determine fiber orientation, fiber length, and fiber attrition in molded parts.

- Procure materials and subsequently mold test samples with selected Tier 1 molder.

Introduction

Injection molding of fiber-filled thermoplastics is a multi-step process which consists of i) pre-mixing and pelletizing the fibers and thermoplastic resin, ii) putting the pellets in an injection molding machine that melts them and injects them under high pressure into a mold (*mold filling*) iii) solidifying the as-formed composite in the mold (*packing, holding and cooling*), and iv) ejecting the part. Mold filling is of greater importance in injection molding of fiber-reinforced thermoplastics since the fiber

orientations in the part are almost entirely determined by the flow patterns during filling. This phase is characterized by a *time-dependent non-isothermal and non-Newtonian flow* with free surface. As a result, it is very challenging to accurately predict the flow-induced fiber orientation. The difficulty is greater when the fibers are long (> 1 mm) and/or concentrated fiber volume fractions are considered, and the part to be molded presents three-dimensional features. Basically, the mold-filling problem is governed by three general equations from continuum mechanics which express

the balances of mass, momentum and energy of the fiber suspension. To complete the problem formulation, a constitutive relation is needed to describe the suspension behavior, and an equation to describe how the fibers can orient in response to the flow and fiber-fiber interaction must be added. These additional equations are essential but are the main sources of difficulty in process modeling. Therefore, assumptions have often been made to simplify the suspension rheology, flow behavior, coupled phenomena, fiber-fiber interactions, etc.

In the next section, the formulation of the mold filling problem is summarized. Standard assumptions will be mentioned. Also, the limitations of the current models will be clarified. After that, an approach to the mold-filling and fiber-orientation problems using the smoothed particle hydrodynamics method will be presented. The modeling investigation continues with the numerical assessment section in which the particularities of the microstructures of short- and long-fiber systems are discussed. Through the analyses of the end-gated plaque and center-gated disk problems, the limitations of the current process model are pointed out. Finally, the report is completed with the material selection, and non-destructive and destructive techniques evaluations for LFTs.

Status of Current Process Models

Generally, it has been verified that the use of orientation tensors provides an efficient and accurate way to quantify the orientation state. Although injection molding of fiber-reinforced polymers, in general, involves flows of non-Newtonian suspending liquids, the assumption of a generalized Newtonian fluid is useful and allows qualitative predictions of fiber orientations in injection-molded parts. In addition, many of these parts possess thicknesses that are much smaller than other relevant dimensions and, consequently, the generalized Hele-Shaw model, which reduces the fully three-dimensional flow problem to a two-dimensional (2D) formulation for the pressure, is valid to model flows in thin cavities. However, the Hele-Shaw approximation has severe limitations to model flows in three-dimensional (3D) geometries since it does not provide accurate velocity fields in 3D features. Another important assumption consists of solving the flow equations as if the fibers were absent, then

using the obtained velocity results to solve the fiber-orientation problem. This uncoupled approach is often acceptable especially in the major (lubrication) part of the flow regions. Fiber-fiber interactions which are important in semi-concentrated and concentrated regimes have been modeled by adding a rotary diffusion term in the Jeffery equation. This term is the key element of the Folgar-Tucker model that is implemented in current commercial process modeling codes. It is noted that capabilities of 3D solutions for the governing equations and/or coupled flow-orientation also exist. 3D analyses using coupled or uncoupled approaches have improved the results for 3D feature problems.

Current process models for injection molding have been applied to short-fiber filled polymers. The fact that these models are able to provide good qualitative fiber orientations is thanks to the acceptable validity of the simplifying assumptions for short-fiber suspensions. However, to date, no models exist to describe the flow of a long-fiber suspension. Here, we arrive at a principal difficulty which is linked to the rheology of the suspension and its microstructure. Long fibers have limited mobility due to their length and multiple fiber-fiber contacts. Hence, they do not freely rotate and convect with the flow. Their behavior needs to be understood and accounted for in the establishment of the fiber-orientation equation. This equation should be able to describe the fiber-fiber interaction when the fibers are long and it should include the effect of fiber breakage during injection molding.

By examining the Folgar-Tucker equation, the limitation of this model can be understood. Advani and Tucker have expressed the Folgar-Tucker model in terms of fiber-orientation tensor components. According to this model, the fibers are convected with the fluid, and they re-orient in response to the deformation and rotation of the fluid according to:

$$\frac{Da_{ij}}{Dt} + \frac{1}{2}(\omega_{ik}a_{kj} - a_{ik}\omega_{kj}) =$$

$$\frac{1}{2}\kappa(\dot{\gamma}_{ik}a_{kj} + a_{ik}\dot{\gamma}_{kj} - 2\dot{\gamma}_{kl}a_{ijkl}) + 2C_1\dot{\gamma}(\delta_{ij} - 3a_{ij})$$

$$(1)$$

where a_{ij} and a_{ijkl} are, respectively, the second-and fourth-order orientation tensors, ω_{ij} is the vorticity tensor, $\dot{\gamma}_{ij}$ is the rate of the deformation tensor whose scalar magnitude is $\dot{\gamma}$. κ and C_I are material constants; κ depends on fiber aspect ratio, r_e, and C_I is called the interaction coefficient. If $C_I = 0$ and $\kappa = (r_e^2 - 1)/(r_e^2 + 1)$, Equation (1) is then Jeffrey's equation for the motion of a rigid, ellipsoidal shape fiber in a Newtonian solvent. This is strictly valid for dilute suspensions in which the fiber-fiber interaction is absent or negligible. Jeffrey's equation imposes no restriction on the fiber aspect ratio. The last term of Equation (1) has been added to model the randomizing effect of fiber-fiber interaction in semi-concentrated suspensions. Here, an important question is raised when applying Equation (1) to long-fiber suspensions, that is, long fibers do not freely rotate and convect with the suspending liquid; they remain organized in domains and have limited mobility. As a consequence, representing the fiber-fiber interaction through a rotary diffusion term dependent on a scalar coefficient (C_I) would be only a rough approximation. Furthermore, the assumption that the fibers are rigid cylinders of equal length and diameter is not valid for injection molding of long-fiber-filled polymers since fibers undergo bending and rupture which reduces the fiber length considerably. This is an important issue that has not been addressed until now.

A Smoothed Particle Hydrodynamics Approach to the Fiber Suspension Problem

An alternative approach to model multiphase free surface flows and heat transfer in domains with complex solid boundaries is to use a *Lagrangian particle-based method*. To simulate the mold-filling problem during injection molding, a smoothed particle hydrodynamics (SPH) approach, a meshless Lagrangian method, can be used. In this approach, the fluid is modeled by a discrete set of SPH particles, where each particle represents a finite volume of the fluid. One of the main advantages of SPH for modeling fiber-filled thermoplastics injection molding is that the SPH method does not require explicit interface tracking or contact angle models. In addition, the SPH simulations explicitly

conserve mass, momentum and energy. Also, the SPH method is manifestly Galilean-invariant because particle-particle interactions depend on the *relative* particle positions and velocity differences. In particle-based methods, the fluid-fluid interfaces move with the particles, hence there is no need to explicitly track the fluid-fluid interfaces. Processes such as fluid fragmentation and coalescence can be handled without difficulty. In addition, the particle nature of the SPH model allows fluid-solid interactions to be modeled through simple particle-particle interactions. The number of particles needed in SPH simulations is determined only by the required resolution and accuracy of the numerical solutions of the macroscopic governing equations such as the Navier-Stokes equation, continuity equation and energy conservation equation.

In SPH, the fluid is modeled by a discrete set of N SPH particles, where each particle represents a finite volume of the fluid. The size of the SPH particles is defined by the desired resolution of the simulation. The position of the ith SPH particle is denoted by the vector r_i, $i = 1,\ldots,N$, and each particle has a mass, m, and density, $\square_i$. The SPH theory is based on the idea that a continuous field $A(r)$ at the position r can be smoothed by the convolution integral:

$$A_s(\mathbf{r}) = \int A(\mathbf{r}')W(\mathbf{r} - \mathbf{r}', h)d\mathbf{r}' \qquad (2)$$

where $A_s(\mathbf{r})$ is the smoothed field, and the weighting function, W, with a range or support scale h satisfies the normalization condition:

$$\int W(\mathbf{r} - \mathbf{r}', h)d\mathbf{r}' = 1. \qquad (3)$$

The integration in Equation (2) is performed over the entire space of the field, and in the $h \rightarrow 0$ limit where W becomes a Dirac delta function, $A_s(\mathbf{r}) \rightarrow A(\mathbf{r})$.

Properties associated with any particle i are calculated by approximating the integral in (2) by the sum

$$A_i = \sum_j \Delta V_j A_j W(\mathbf{r}_i - \mathbf{r}_j, h)$$

$$= \sum_j m_j \frac{A_j}{\rho_j} W(\mathbf{r}_i - \mathbf{r}_j, h) \tag{4}$$

where ΔV_j is the volume occupied by particle j, ρ_j is the density at position $\mathbf{r}_j$, and the gradients of these properties are approximated by

$$\nabla A_i = \sum_j \Delta V_j A_j \nabla_i W(\mathbf{r}_i - \mathbf{r}_j, h)$$

$$= \sum_j m_j \frac{A_j}{\rho_j} \nabla_i W(\mathbf{r}_i - \mathbf{r}_j, h) . \tag{5}$$

The motion of each particle is governed by the momentum conservation described by the Navier—Stokes equation which can be written in terms of SPH approximation as

$$\frac{d\mathbf{v}_i}{dt} = -\sum_j m_j \left(\frac{\sigma_j}{\rho_j^2} + \frac{\sigma_i}{\rho_i^2} \right) \nabla_i W(\mathbf{r}_i - \mathbf{r}_j, h) + \mathbf{g} \tag{6}$$

where σ and τ are total and extra stress tensors given by:

$$\sigma = P\mathbf{I} + \tau . \tag{7}$$

Most of the existing models for fiber orientation in injecting molding operate under a simplifying assumption of fluid incompressibility. SPH allows the fluid flows ranging from essentially incompressible fluids (compressibility less than 0.01%) to highly compressible fluids to be modeled. The density ρi that can be determined from the continuity equation or directly from Equation (4), is given by:

$$\rho_i = \sum_j m_i W(\mathbf{r}_i - \mathbf{r}_j, h) \tag{8}$$

for $A_i = \rho i$ Mass m_i depends on the volume fraction of the suspending liquid that is related to the volume fraction of fibers. Given the density ρi, the pressure P in Equation (7) is obtained from the equation of state. To simulate the effect of surface tension and fluid-solid interactions, particle-particle interactions were added

into Equation (6) leading to the particle equation of motion:

$$\frac{d\mathbf{v}_i}{dt} = \mathbf{F}_i \tag{9}$$

Where $\mathbf{F}_i = -\sum_j m_j \left(\frac{\sigma_j}{\rho_j^2} + \frac{\sigma_i}{\rho_i^2} \right) \nabla_i W(\mathbf{r}_i - \mathbf{r}_j, h)$

$$+ \mathbf{g} + \frac{1}{m_i} \sum_j \mathbf{F}_{ij} \tag{10}$$

where $\sum_j \mathbf{F}_{ij}$ is the force acting on particle i due to the particle-particle interactions with the interaction forces given by

$$\mathbf{F}_{ij} = \begin{cases} s_{ij} \cos\left(\frac{1.5\pi}{3h} |\mathbf{r}_j - \mathbf{r}_i| \right) \frac{\mathbf{r}_j - \mathbf{r}_i}{|\mathbf{r}_j - \mathbf{r}_i|}, & |\mathbf{r}_j - \mathbf{r}_i| \le h \\ 0, & |\mathbf{r}_j - \mathbf{r}_i| > h, \end{cases} \tag{11}$$

where s_{ij} is the strength of the force acting between particles i and j. Since $\mathbf{F}_{ij} = -\mathbf{F}_{ji}$, the particle-particle interactions conserve momentum.

The fiber orientation at each point of the fluid domain represented by the particle i can be obtained by solving the Advani-Tucker Equation (1) that can be expressed in terms of the SPH discretization.

Current modeling tools for flow-induced fiber orientation relies on finite element (FE) analyses. After the prediction of fiber orientation by a FE processing code, the same mesh is imported into a structural FE code for a structural analysis. By doing so, some numerical problems could arise because the mesh that is good for flow modeling might not be appropriate for a structural analysis (e.g., damage analysis, etc.). The advantage of the SPH approach is that fiber-orientation prediction is not linked to a finite element mesh, but it is linked to the domain which corresponds to the part to be molded. Hence, once fiber orientation has been known at each point of the domain, the domain can be meshed as appropriately as necessary for a structural analysis that makes use of the orientation results.

Finally, it is important to note that the major issue in SPH modeling is related to the high computational cost in comparison with the analyses using the grid-based numerical methods (e.g., finite element methods). To date, the automotive industry is not familiar with the SPH methods. On the other hand, finite element tools are available and will be more effectively explored to handle LFT analyses.

Numerical Assessment [1]

Conventional, short-fiber, thermoplastic composites (SFTs), processed by injection molding, have a layered structure. As shown in Figure 1, the fibers close to the cavity walls are aligned with their neighbors and oriented in the flow direction. This region of the part is called the shell layer. Fiber alignment in the shell is caused by shearing deformation during mold filling; the polymer has high velocity near the midplane of the part and does not slip on the cavity walls. Closer to the midplane of the part, the fiber orientation is nearly random in the flow/crossflow plane, and there may even be a dominant crossflow orientation. This layer is called the core. On a polished cross-section, the human eye will see a sharp transition between the core and the

Figure 1. Cross-section of injection-molded plaque, showing typical core/shell layered pattern of fiber orientation.

shell. This is apparent in Figure 2(a). However, quantitative measurements show that the transition is actually smooth, as in Figure 2(b).

A center-gated disk has the same layered structure as a plaque, but the shell layers are thinner, the core is thicker, and the core has a strong crossflow alignment. This is shown in Figure 2(b). The crossflow core in the disk is caused by fluid stretching in the tangential direction as it flows radially outward from the gate. This stretching is strongest at the midplane, where there is no gapwise shearing, whereas the gapwise shearing dominates the fiber orientation near the upper and lower surfaces of the part, as illustrated in Figure 3. At intermediate positions across the thickness, the orientation is a result of the competition between shearing and stretching.
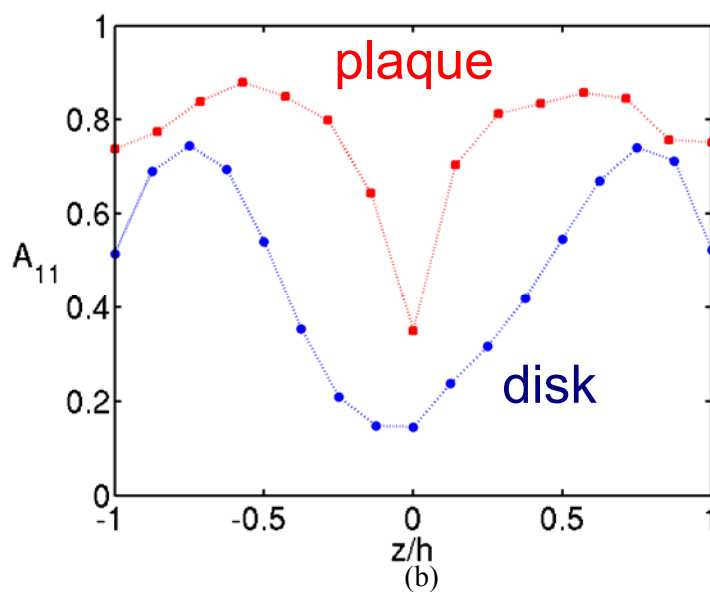

(a)   (b)
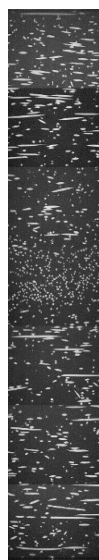
Figure 2. (a) Polished cross-section from an injection-molded SFT. Taken from a region like the white area in Fig. 2. (b) Measured flow-direction orientation in a plaque (upper curve) and center-gated disk (lower curve).
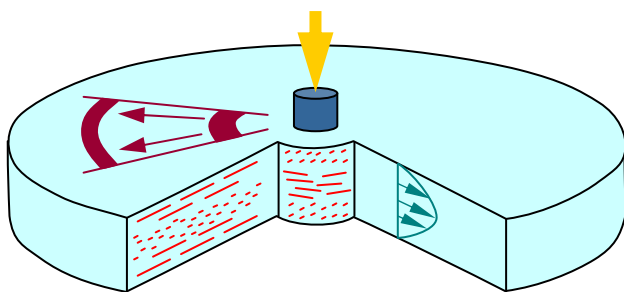
Figure 3. Sketch of fiber orientation structure in a center-gated disk.

Current fiber orientation models for injection-molded SFTs are accurate for parts with long flow lengths, but not as accurate when the flow length is short. Table 1 shows an example. For this table, plaques of two different sizes were molded from a 30% glass-reinforced PBT, General Electric Valox 420. The fiber orientation was measured in each type of plaque, and the elastic modulus was measured in the flow direction (E_{11}) and the crossflow direction (E_{22}). The fiber orientation was also predicted using the standard model. The same micromechanics model was then used to make two predictions for the elastic moduli, one using the measured fiber orientation and the other using the predicted fiber orientation.

Table 1 shows the results. The parts are strongly anisotropic, being almost twice as stiff in the flow direction as in the crossflow direction. However, the part with the shorter 125-mm flow length has less anisotropy than the part with the longer 279-mm flow length. Using the measured orientation to predict the properties results in good agreement for both parts. The predictions based on the predicted orientation are quite good for the long part, but do

not capture the increased isotropy for the short flow length. The underlying issue is that the standard theory predicts a core that is too narrow and shell layers that are too thick, compared to experiments (see Figure 4). This deficiency in the standard model is currently being addressed, and there is an improved model developed, by Prof. Charles Tucker and his students at the University of Illinois. Continued work on fiber orientation models is required, even for the familiar SFT materials.

Material Selection and Injection Molding

Materials to be analyzed have been determined to be both glass- and carbon-filled polypropylene and Nylon 6 at a 30% fiber by weight. These materials will be procured by an industrial supplier; currently we are evaluating three material suppliers and will down-select to one supplier for full project participation. The samples will be long fiber between 6-15 □m in length.

Injection-molding-machine adaptation to minimize fiber attrition during long-fiber processing has been investigated during the course of this project. The injection-molding machine will be fixed with low-torque screws that will minimize fiber shearing during the heating and material transfer. Molds will be commercially-available designs produced from matched metal parts; we have identified a company in Greenville, MI that is experienced in producing multi-plate molds capable of producing a variety of sample configurations from a single mold base. In the experimental phase of this program, we will pursue the purchase and installation of one of these systems at PNNL.

Table 1. Comparison of measured and predicted elastic moduli for plaques molded from 30% glass-fiber-reinforced PBT. E11 is measured in the flow direction and E22 is measured in the crossflow direction.

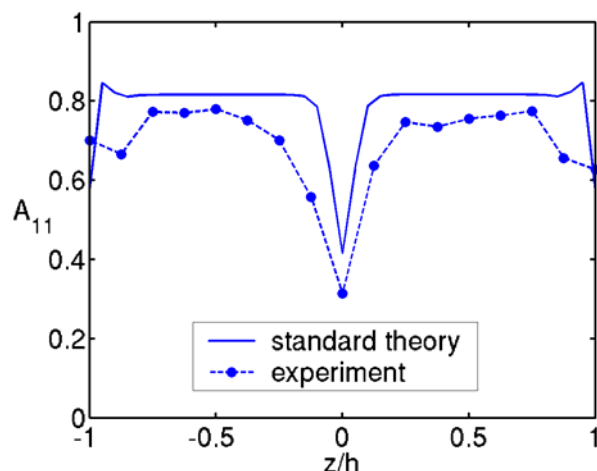| Plaque Dimensions (mm)	Property	Experimental (GPa)	Predicted using measured orientation (GPa)	Predicted using predicted orientation (GPa)
76x299x3	E_{11}	8.8	9.1	8.6
76x299x3	E_{22}	4.6	4.4	4.3
75x125x3	E_{11}	7.7	7.5	8.6
75x125x3	E_{22}	5.4	4.8	4.3

Figure 4. Fiber orientation in a 80x90x2mm plaque of 30% glass/PBT. The predictions using the standard theory give a much narrower core than the part actually has.

Non-Destructive and Destructive LFT Analysis

Short-fiber-reinforced thermoplastics are commonly utilized for applications in the automotive industry due to several factors including commonalities in process equipment with neat resin products, homogeneities within the fiber filler, consistent fiber lengths that are generally 1 mm or less, and a generally well-defined procedure for predicting fiber orientation within the molded part. Long-fiber-filled thermoplastic composites, on the other hand, pose significant challenges in both processing and fiber-orientation prediction. Specifically, factors that are inherent in long-fiber injection-molded parts include fiber breakage, large variations in viscosity due to decreased fiber mobility especially at the mold front, non-homogeneous yield stresses, increased fiber-wall interactions, and poor fiber dispersion that can be manifested in the presence of fiber bundles in the molding. Injection molding of simple geometries will be performed using long glass or carbon fibers. The injection-mold screw must be modified to provide the correct fiber shear to minimize fiber breakage and provide uniform distribution. However, the fiber aspect ratio of long-fibers results in an increased core thickness possibly reflecting the segregated state at which long fibers enter the mold cavity. Steric interactions that inhibit the motion and reorientation of longer fibers can result in the orientation of both the core and shell layers being lower than that of short-fiber composites; all of these factors require that careful experimental characterization and fiber orientation characterization be pursued in conjunction with any fiber orientation prediction modeling.

Nearly any measurement technique could be used provided that it effectively generates images of fibers on 2D sections through the material. Techniques could range from contact x-ray micro-radiography, scanning acoustic microscopy, to nuclear magnetic resonance. For studies of mesostructural artifacts, optical is preferred despite requiring a polished section of the sample.

If samples are to be prepared for 2D optical-reflection microscopy and analysis by image-processing techniques, great care must be taken with both sectioning and final polishing. Physical sectioning of long-fiber reinforcements can easily cause fiber pullout and the sectioning of aligned-fiber composites can leave chipped fibers and fragments at the surface. Clearly, fiber orientation characteristics that result from any 2D image analyzer depend upon the quality of the surface preparation.

Image reconstruction from projections using computed tomography (CT) produces a non-invasive measure of structure from external measurements. In x-ray CT, radiation is passed along straight lines through the object to a detector with the specimen located between the source and detector. With a stationary source and detector, the specimen is rotated and stopped at predetermined locations to acquire projections through the specimen. The projected signal is proportional to the amount of radiation reaching the detector. The logarithm of these measurements can be considered line integrals of the x-ray attenuation coefficients of the specimen; knowledge of all line integrals allows the reconstruction of the interior structure. X-ray tomographic reconstruction produces a two-dimensional map of x-ray attenuation coefficients of the irradiated cross-section of the specimen.

Transmission tomography such as x-ray CT employs scanners capable of discerning between linear attenuation coefficients values by as little as 0.1%. In practice, density measurement from x-ray tomographic data can be performed either by

calibrating the CT machines with objects of known density and obtaining a correlation equation that relates density with attenuation coefficients or by utilizing dual energy scanning to determine directly the density of the material. Typically, volumetric imaging of the sample is obtained by stacking a series of two-dimensional slices of CT image data. The quality and utility of the CT data ultimately depend on the machine resolution.

For many years, the standard method of acquiring a volumetric CT scan has been by scanning a sample one slice at a time. In this method, a linear detector array and an x-ray point source are mounted opposite each other. A fan-shaped x-ray beam traverses the sample and the attenuated fan is stored as a 1D linear image. By rotating the source/detector pair around the sample, a series of linear images are obtained which are subsequently used for the 2D slice reconstruction. A volumetric representation is obtained by advancing the table on which the sample rests after each rotation in order to acquire a stack of such slices. Recently, cone-beam CT has been developed to acquire a 3D data set with only one rotation of the sample. This provides for fast data acquisition and better x-ray utilization. In a cone-beam design, each projection of the object is, in essence, a radiograph. Attenuation measurements are simultaneously made for the entire object rather than for a single slice. The reconstruction algorithm is a generalization in three dimensions of the widely used convolution back-projection method.

Fiber Architecture of Long-Fiber-Reinforced Thermoplastics (LFTs)

The ability to characterize fiber orientation, fiber length and fiber curvature is critical for validation of predictive processing codes and for input to micro-mechanic-based life-prediction software. The ORNL team has been focusing on evaluating various destructive and non-destructive characterization methods. The objective of this study was to evaluate these methods for glass- as well as carbon-fiber-reinforced injection-molded parts. RTP Co. supplied tensile and flex bars molded from VLF 80107 CC natural polypropylene with 40% 11-mm-long glass fibers. Long-carbon-fiber-reinforced thermoplastics were not available on the market, therefore the evaluation was performed on specimens made from AMC 8590 sheet-molding compound by Quantum

Composites with 1-inch-long fibers. A flexible surface heater by Thermion was used for evaluation of x-ray techniques with Ni-coated carbon fibers.

X-ray Techniques

X-ray techniques can be used for obtaining two- and three-dimensional information about the fiber architecture of LFT. The utility of x-ray evaluation is dependent on differences in x-ray absorptance of materials used in the composite. Glass-reinforced thermoplastics can be used without modification, while carbon-fiber-reinforced thermoplastics must be modified to obtain sufficient contrast. Two techniques were considered for modification of carbon-fiber-reinforced thermoplastics. Dispersion of metallic particles in the matrix and coating of fibers was considered. Micrographs of particles dispersed in the polymer matrix, obtained from a previous project, were evaluated (Figure 5). It was concluded that this technique was not promising due to the necessity of uniformly dispersing sub-micron-sized particles in the matrix material. Large particles and non-uniformities in particle distribution would cause problems during image analysis. Ni-coated carbon fibers were deemed more suitable for x-ray analyses (Figure 6). It remains to be seen how the coating affects the manufacturing process, fiber length and fiber orientation in long-fiber thermoplastics.
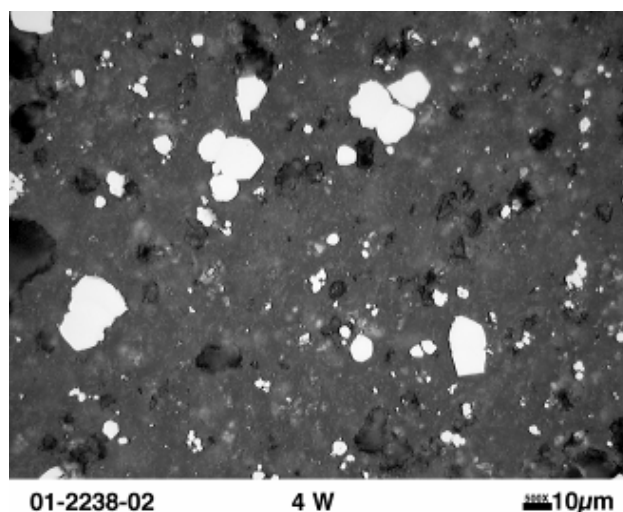

01-2238-02   4 W   10μm

Figure 5. Micrograph of 17% tungsten particles by volume in polymeric matrix (material supplied by Ecomass).

Two x-ray techniques were evaluated: 2D x-ray and 3D x-ray computed tomography. Information from 2D x-ray can provide useful information about fiber distribution spanning several fiber lengths (Figure 7). While it is impossible to resolve the individual fibers due to the number of fibers present in the thickness direction of a typical molded specimen, local fiber density, uniformity of fiber dispersion and general fiber-bundle orientation can be judged from the image. 2D x-ray imaging can be performed rapidly, and commercial equipment is readily available.

3D x-ray computed tomography was used to image individual fibers in LFT (Figure 8). Trade-offs must be made between image resolution, specimen size and time of imaging. Figure 8 shows images of 2x2 mm, 40% glass-reinforced sample with 4.5-micrometer resolution obtained in one hour with equipment available at ORNL. X-ray tomography machines are commercially available; however, the equipment is not as easily accessible as 2D x-ray machines.
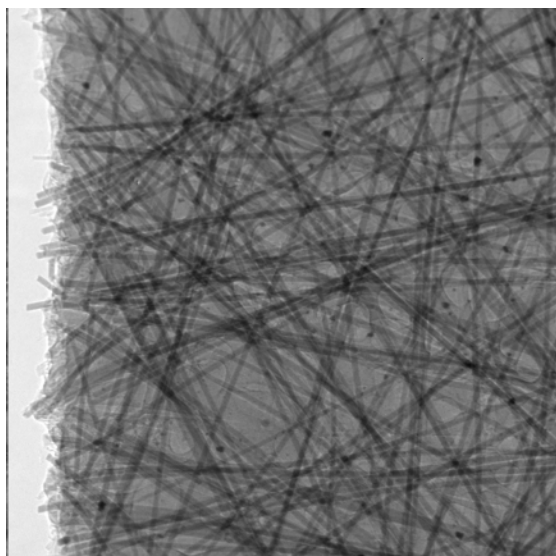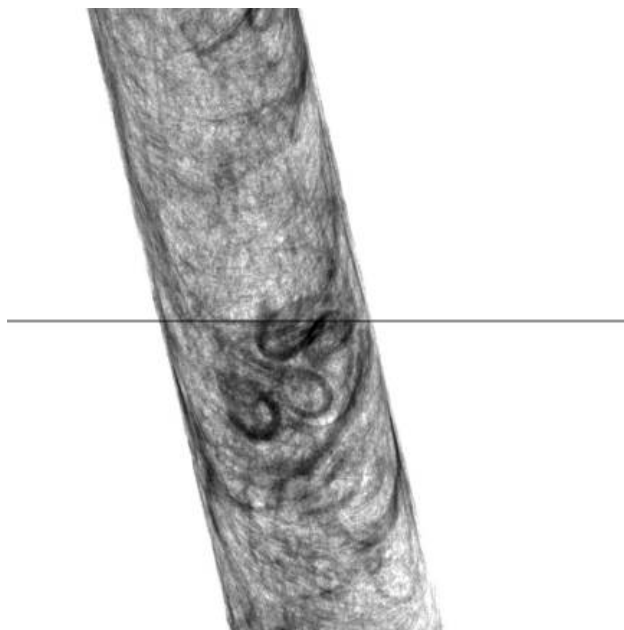

Figure 7. 2D x-ray of 12.5-mm-wide, 40% glass/polypropylene injection-molded tensile bar.
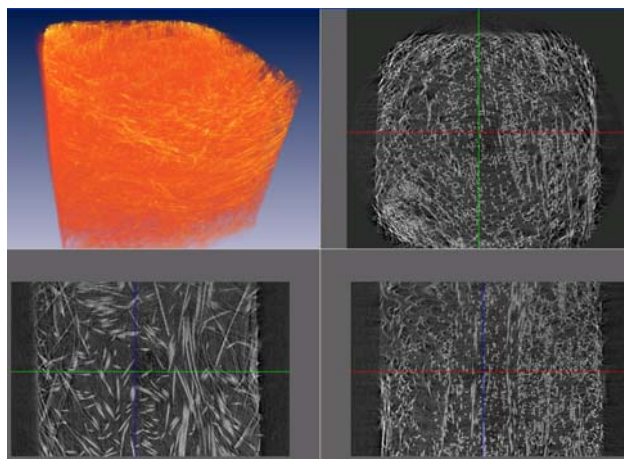
Figure 8. 3D x-ray of 2x2-mm sample of 40% glass/polypropylene - ISO view and three cross-sections.

Figure 6. X-ray of Thermion heater containing Ni-coated carbon fibers.

Obtaining the orientation tensor $\langle a_{ij} \rangle$ requires digital image processing on data obtained from 3D x-ray analysis. Software capable of this analysis in 3D was not available; however, a possible approach for determining $\langle a_{ij} \rangle$ was demonstrated in 2D by analyzing a cross-section. Steerable filters were applied to resolve fibers (Figure 9, Figure 10) before the distribution of fiber orientations was determined (Figure 11).

Figure 9. Threshold filter applied to cross-section obtained from 3D x-ray.

Figure 10. Edge filter applied to cross-section obtained from 3D x-ray.

An ash method was evaluated for fiber length determination. The matrix was burned off from a section of a specimen in a furnace at 600°C. A small amount of fibers was separated from the middle of the section so that the sample would not contain any fibers that were cut before the burn-off. This was accomplished by placing a drop of epoxy adhesive and washing free fibers away after the
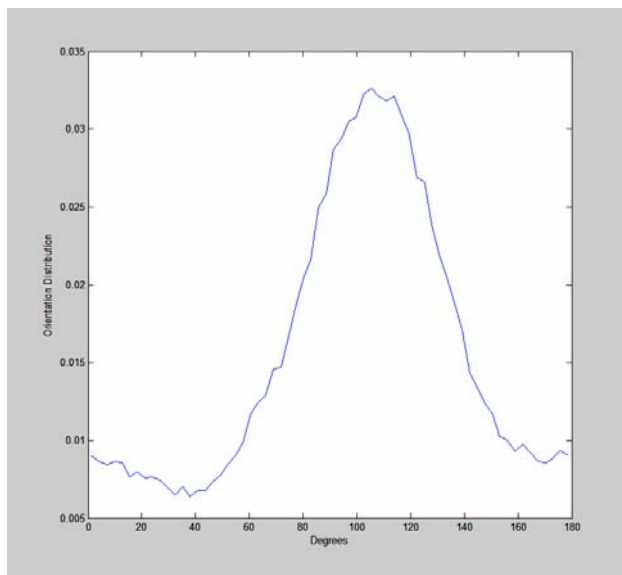

Figure 11. Orientation of fiber cross-sections in a slice from 3D x-ray data.

adhesive set. It is challenging not to break any fibers as the fibers are tightly wrapped and intermingled. The adhesive drop was then burned off and the fibers spread on a Petri dish. Again, separating fibers and fiber bundles is challenging without breaking the fibers. Fibers were then imaged via a scanner. The need for a microscope with an x-y stage and multiple image assembly was eliminated by the use of a scanner. Fiber lengths were then evaluated using ImageJ freeware (Figure 12).
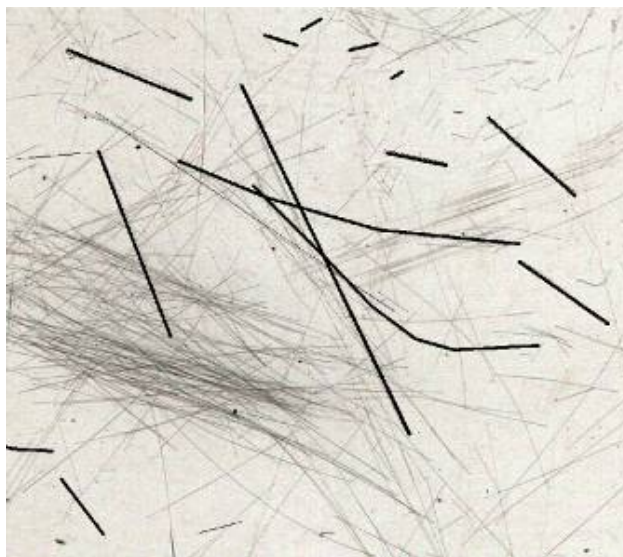

Figure 12. Fibers and their approximation for fiber-length determination.

MRI, c-scan and other acoustic methods were also evaluated; however, initial results did not indicate that these methods would be useful for determination of fiber architecture in LFT.

Conclusions

This report covers five months of work in FY 2005 (the project started in June 2005). Our efforts were focused on evaluating existing process models, computational methods, emerging microstructure, and fiber-orientation characterization tools to determine their accuracy and potential applications to LFTs.

When SFTs are compared to LFTs, many factors suggest that orientation models which work well for SFTs may not work so well for LFTs. Because the fibers in LFTs are nearly an order of magnitude longer than in SFTs, it is expected that fiber-fiber interactions are important over longer length scales. LFTs can have a patchy, domain-like orientational structure, where fibers are aligned similarly to their neighbors. This does not occur in SFTs. Previous experiments on LFTs show a variation in fiber volume fraction across the thickness of molded parts, with higher fiber concentrations near the midplane of the part. Finally, the average fiber length can change significantly during the processing of an LFT, and can vary from location to location within a molded part. None of these factors are accounted for in current models of fiber orientation in SFTs, but all of these factors will affect the mechanical properties of molded parts. Thus, there is a significant need to develop more advanced processing-microstructure models for LFT materials, in order to provide a solid foundation for predictive engineering.

Experimental characterization of the composite microstructure (i.e., fiber-orientation and aspect-ratio distributions, fiber organization structure) is essential since all the modeling work relies on it. Currently, there are promising techniques based on computed tomography that can be used effectively to characterize LFT microstructures.

Reference

[1] Tucker III, CL. "Material on Injection Molding Flow Modeling and Fiber Orientation Modeling." Internal Report submitted to PNNL.

Presentations/Publications/Patents

Nguyen, BN, Holbery, JD, Johnson, KI, Smith, MT. "Long Fiber Injection Molded Composites: From Process Modeling to Property Prediction" Proceedings of the 5th Annual Automotive Composites Conference and Exhibition, Troy, MI, September 12-14, 2005.