

Available online at www.sciencedirect.com



COMPOSITES Part A: applied science and manufacturing

Composites: Part A 38 (2007) 469-483

www.elsevier.com/locate/compositesa

Fiber-fiber interactions in carbon mat thermoplastics

A.C. Caba^{a,*,1}, A.C. Loos^b, R.C. Batra^c

^a Alliant Techsystems, Inc., Route 114, P.O. Box 1, Radford, VA 24143-0100, USA ^b Department of Mechanical Engineering, Michigan State University, East Lansing, MI 48824, USA ^c Department of Engineering Science and Mechanics, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061, USA

Received 6 October 2005; received in revised form 26 February 2006; accepted 27 February 2006

Abstract

Carbon mat thermoplastics (CMT) consisting of 12.7 mm long, chopped carbon fibers in a polypropylene matrix were manufactured using the wetlay technique at fiber volume fractions (FVF) from 10% to 25%, and tests simulating the compression molding process were conducted. The packing stress of the CMT followed a power law relation with FVF. A single fiber pull-out fixture was used to measure the frictional and hydrodynamic lubrication coefficients at fiber–fiber touch points, and results were fit with an existing relation for glass mat thermoplastics. In isothermal squeeze flow the load–displacement behavior for the 10% FVF CMT was similar in shape to that for a fluid with a yield stress. However, for FVFs of 15–25%, the load–displacement curves showed a load spike at the beginning of the flow, then followed the curve for a fluid with a yield stress. The spike was attributed to fiber breakage that increased with increasing FVF of the sample.

© 2006 Elsevier Ltd. All rights reserved.

Keywords: A. Carbon fibre; A. Polymer-matrix composites (PMCs); B. Mechanical properties; Fibre-fibre contacts

1. Introduction

Compression molded parts made with standard longfiber glass-mat thermoplastics (GMT) have begun to reach limitations in meeting part thickness, weight, and strength requirements. With current commercial materials, increased part strength requires thicker parts and hence extra weight. Thicker GMT parts begin to exceed allowable dimensions, forcing the use of heavier traditional materials such as steel. These restrictions also limit new automotive applications for GMT such as T-tops or floor pans, where space is at a premium. While GMT provides significant weight savings over metal stampings, further weight and space savings can be realized if a higher performing reinforcement such as carbon fiber is used. Long-fiber carbon-mat thermoplastic (CMT) offers increased performance over GMT in strength, stiffness, and weight. Research on GMT material has focused on how easily it can be processed into desired parts and the determination of its mechanical properties, but no research has explored long-fiber CMT material. Before using CMT in a production environment, it is necessary to characterize its flow properties.

Designs with long-fiber thermoplastics typically rely on a trial-and-error approach for designing the mold and charge placement. Simulation packages specifically designed for simulating the flow of long-fiber thermoplastic composites require measuring particular flow parameters of the material, and these flow parameters have been measured for GMT but not for CMT [1–6]. Accurate flow simulations will help to reduce the design cycle time and improve the quality of parts manufactured by eliminating weld lines through informed mold design and charge placement. Additionally, models of the flow behavior of GMT have been developed that include macro- and micromechanical descriptions of the process [5–12].

^{*} Corresponding author. Tel.: +1 540 552 7118.

E-mail addresses: Aaron.Caba@atk.com, aaroncaba@yahoo.com (A.C. Caba), aloos@egr.msu.edu (A.C. Loos), rbatra@vt.edu (R.C. Batra).

¹ Supported by the Center for High Performance Manufacturing at Virginia Polytechnic Institute and State University.

¹³⁵⁹⁻⁸³⁵X/\$ - see front matter © 2006 Elsevier Ltd. All rights reserved. doi:10.1016/j.compositesa.2006.02.024

A 300 mm wide lab-scale wetlay line at Virginia Tech was used to produce ready-to-mold mat from carbon and thermoplastic fibers. All of the material needed for this study was produced on this line, ensuring that direct control over processing variables was kept under the supervision of the researchers. A full description of the line is given by Lu [13] and Caba [14].

Much research effort has been focused on GMT materials, including optimization of their processing capabilities and the determination of their mechanical properties. Short carbon fiber ($\leq 2 \text{ mm}$ long) thermoplastics have also been studied, but no research has explored the long-fiber CMT material. The present work develops the knowledge base for CMT materials by performing the following tasks:

- Manufacture the CMT material using the wetlay process. The wetlay processing characteristics of the carbon and thermoplastic fibers were investigated, including varying lengths of carbon fibers and the resulting areal weight of the final mat.
- Characterize the flow properties of the CMT material. This included measuring the flow parameters of the material under typical compression molding conditions, developing a relation that adequately describes the experimental findings, and comparing the measured characteristics of the CMT with published values of GMT.

2. Theory

The CMT will be considered to be a concentrated suspension of uniform fibers in a viscous medium where the fibers in the suspension are cylindrical, slender $(l/d \gg 1)$, and straight. The fiber bed is assumed to be well dispersed and all fibers are taken to lie in a 2-D plane.

2.1. Fiber-fiber interaction forces

It has been shown that the major forces acting in a flow of fiber filled polymer material occur at the fiber-fiber touch points [15], therefore it is necessary to establish the distribution of contacts in the medium and the forces at these contact points. Toll [9,16] has exactly calculated the number of interactions between a test fiber and other fibers in a network. Consider the orientation and length distribution function $\Psi'(\vec{p}, l)$ where \vec{p} is a unit vector along a fiber and l is the fiber length. For a mat of mono-disperse fibers of length l and diameter d, the average number N of fibers intersecting a tube of radius d surrounding a fiber of interest is

$$N = 2n_{\rm f} l^2 df + \pi n_{\rm f} l d^2 g + \pi n_{\rm f} l d^2, \tag{1}$$

where n_f is the number of fiber center points per unit volume of the mat, and the orientation functions f and g are given by

Table 1

Calculated	values	of <i>f</i>	and	g for	three	orientations	[9]
------------	--------	-------------	-----	-------	-------	--------------	-----

Orientation	f	g
Unidirectional	0	1
2-D random	$2/\pi$	$2/\pi$
3-D random	$\pi/4$	1/2

$$f = \oint \oint |\sin \widehat{\vec{p}\vec{q}}| \Psi'(\vec{p}) \Psi'(\vec{q}) \,\mathrm{d}\vec{p} \,\mathrm{d}\vec{q}, \tag{2}$$

$$g = \oint \oint |\cos \widehat{\vec{pq}}| \Psi'(\vec{p}) \Psi'(\vec{q}) \, \mathrm{d}\vec{p} \, \mathrm{d}\vec{q}, \tag{3}$$

where $\hat{p}\hat{q}$ is the angle between two fibers. Values of f and g for different fiber orientations are listed in Table 1. Assuming that the fibers are oriented in a plane and that $l/d \gg 1$, Eq. (2) reduces to

$$f = \oint \oint |\sin(\theta - \theta')| \Psi(\theta') \Psi(\theta) \, \mathrm{d}\theta' \, \mathrm{d}\theta, \tag{4}$$

where $\Psi(\theta)$ is the 2-D orientation distribution function, and Eq. (1) can be rewritten as

$$n^{(i)} = \frac{8}{\pi} \frac{l}{d} f \phi, \tag{5}$$

where $n^{(i)}$ is the number of contacts along a given fiber and ϕ is the fiber volume fraction (FVF). Assuming a well dispersed planar bed of fibers, Toll and Månson [17,10] derived equations for normal and tangential contact forces at fiber–fiber touch points. Fig. 1 shows forces acting at the point where two fibers touch. The average normal force $|\vec{c}^n|$ at the touch points is

$$\overline{|\vec{c}^n|} = \frac{32}{5\pi^2} E_{\rm f} d^2 f^3 \phi^3, \tag{6}$$

where $E_{\rm f}$ is the modulus of the fibers. Using Eq. (6) the tangential force at each contact point on the fiber can be expressed as

$$\vec{c}^{\rm f} = G(|\vec{c}^n|, |\vec{v}|)\vec{v},\tag{7}$$

where \vec{c}^{f} is the in-plane force vector, \vec{v} is the sliding velocity vector, and *G* is a friction function. Servais et al. [6,8] have shown that \vec{c}^{f} can be separated into a frictional component \vec{f}_{f} and a hydrodynamic lubrication component \vec{f}_{h} :

$$\vec{c}^{\rm f} = \vec{f}_{\rm f} + \vec{f}_{\rm h}.\tag{8}$$



Fig. 1. Forces at a fiber-fiber touch point.

Assuming that the shear viscosity, η , of the suspending fluid behaves according to the Carreau relation [18]

$$\eta(\dot{\gamma}) = \eta_0 [1 + (\lambda \dot{\gamma})^2]^{(n-1)/2}, \tag{9}$$

the force due to the shearing of the fluid layer was found to be

$$\vec{f}_{\rm h} = k_{\rm h} \eta_0 \left[1 + \left(\lambda \frac{|\vec{v}|}{\alpha} \right)^2 \right]^{(n-1)/2} \frac{\vec{v}}{\alpha},\tag{10}$$

where η_0 , λ , and *n* are the Carreau parameters of the fluid, α is the lubricating film thickness, \vec{v} is the relative sliding velocity at the touch point, and $k_{\rm h}$ is the hydrodynamic lubrication coefficient. The actual physics of this sliding contact is very complex, thus the coefficient cannot be interpreted in a physically meaningful way. Coulomb friction is assumed for the other component of the sliding force

$$\vec{f}_{\rm f} = k_{\rm f} |\vec{c}^n| \frac{\vec{v}}{|\vec{v}|},\tag{11}$$

where $k_{\rm f}$ is the Coulomb frictional coefficient. Static and kinetic friction coefficients are assumed to be equal.

In order to test the validity of Eqs. (10) and (11) for both dry and impregnated conditions, Servais et al. [6,8,19,20] devised a heated apparatus that allows one to vary the FVF and simultaneously measure the force required to pull either a single fiber or a fiber bundle through the mat. From these experiments they were able to measure both the frictional force and the hydrodynamic lubrication force at fiber–fiber contact points. For a fiber with length Lembedded in the molten material, the following relationship was obtained between the pulling force \vec{F} and the pulling velocity \vec{v}

$$\frac{\dot{F}}{L} = k_{\rm f} \left(\frac{256}{5\pi^3} \,\mathrm{d}E_{\rm f} f^4 \phi^4\right) \frac{\vec{v}}{|\vec{v}|} + k_{\rm h} \left(\frac{8}{\pi d} f \phi\right) \eta_0 \left[1 + \left(\lambda \frac{|\vec{v}|}{\alpha}\right)^2\right]^{(n-1)/2} \frac{\vec{v}}{\alpha}.$$
(12)

A similar relation was determined for fiber bundles. These relations were verified by oscillatory shear measurements of the molten material.

2.2. Fiber axial force

During squeeze flow the resultant of the frictional forces on each individual fiber may generate in it an axial stress that exceeds its strength, causing it to break into shorter pieces. Two methods to estimate the maximum stress in a fiber in the flowing CMT are presented. A purely extensional flow field with no friction between the platens and the CMT material is shown in Fig. 2. The flow is assumed to be incompressible and axisymmetric about the z-axis and to satisfy

$$\frac{1}{r}\frac{\partial}{\partial r}(rv_r) + \frac{\partial v_z}{\partial z} = 0,$$
(13)



Fig. 2. Squeeze flow sketch.

where the velocity components v_r and v_z are assumed to have the following form:

$$v_r = f(r),\tag{14}$$

$$v_z = -\dot{\varepsilon}z. \tag{15}$$

Eq. (13) is integrated with respect to r and solved for f(r):

$$f(r) = \frac{ru}{2h} + \frac{C_1}{r},\tag{16}$$

where C_1 is a constant of integration. Since v_r must be finite at r = 0, $C_1 = 0$. Thus the velocity field between the plates is

$$v_r = \frac{r\dot{\varepsilon}}{2},\tag{17}$$

$$v_z = -z\dot{\varepsilon}.\tag{18}$$

A single fiber in a steady flow is shown in Fig. 3. The circles represent other fibers that are contacting the fiber under consideration. To an observer sitting on any fiber, the flow field will look the same, therefore we consider the fiber with its center located at r = 0. The following assumptions are made:

- The number of fiber touches $n^{(i)}$ is known (e.g. $n^{(i)} = 5$ in Fig. 3) and is large.
- The fiber touches are equally spaced along the fiber.
- The velocity of each contacting fiber is along the axis of the main fiber.
- The velocity at each contact point equals $v_r(r)$.
- The axial force at each contact point is based on the average contact force.

Since the fibers in the mat are oriented randomly, the actual contact problem is much more complicated than what is assumed here. These assumptions will allow us to



Fig. 3. Sketch of a single fiber in the flowing CMT.

determine a first order approximation of the forces on an individual fiber in the flowing CMT. The maximum force in each fiber will occur at the fiber's center. Summing one half of the forces on the fiber gives the maximum force F_{max}

$$F_{\max} = \frac{n^{(i)}}{2} |\vec{f}_{\rm f}| + \sum_{j=1}^{n^{(i)}/2} |\vec{f}_{\rm h}(r=ja)|, \qquad (19)$$

where $a = l/n^{(i)}$ is the average spacing between touches. The maximum axial stress in each fiber is given by

$$\sigma_{\rm f,max} = \frac{F_{\rm max}}{\pi d^2/4}.$$
(20)

For computational purposes, it is convenient to find an analytical expression for Eq. (19). Since $n^{(i)} \gg 1$,

$$F_{\max} \approx \frac{n^{(i)}}{2} |\vec{f}_{\rm f}| + \frac{n^{(i)}}{l} \int_{0}^{l/2} f_{\rm h}(r) \, \mathrm{d}r = \frac{n^{(i)}}{2} |\vec{f}_{\rm f}| + \frac{n^{(i)}}{l} \frac{2k_{\rm h}\eta_{0}\alpha}{\lambda^{2}\dot{\varepsilon}(1+n)} \left\{ \left[1 + \left(\frac{\lambda}{4\alpha}\dot{\varepsilon}l\right)^{2} \right]^{(1+n)/2} - 1 \right\}.$$
(21)

The above derivation assumes that the velocity of each contacting fiber is parallel to the stationary fiber's long axis. In order to relax this assumption, the average friction force at a distance χ from the center of the test fiber can be found by integrating over all possible fibers that contact at that point, as shown in Fig. 4. It is assumed that the contacting fiber has its center located at

$$\vec{p} = x\hat{\vec{x}} + y\hat{\vec{y}} \tag{22}$$

with respect to center of the test fiber, is translating as a rigid body with a velocity $\vec{v}(\vec{p})$, and it contacts the test fiber at a distance χ from the test fiber's center. Therefore, the average frictional force on the test fiber is

$$\overline{\vec{c}}^{f}(\chi) = \frac{1}{\pi R^{2}} \int_{\chi-R}^{\chi+R} \int_{-(R^{2} - (\chi-x)^{2})^{1/2}}^{(R^{2} - (\chi-x)^{2})^{1/2}} \vec{F}(\vec{p}) \,\mathrm{d}y \,\mathrm{d}x,$$
(23)

where R = l/2 and $\vec{F}(\vec{p})$ is the frictional force on the test fiber from a fiber centered at \vec{p} . The force is known as a function of velocity

$$\vec{F}(\vec{p}) = \vec{c}^{\rm f}(\vec{v}(\vec{p})),\tag{24}$$



Fig. 4. Circular region of integration to determine the average force at $x = \chi$.

where

$$\vec{v}(\vec{p}) = \frac{1}{2}\vec{p}\dot{\epsilon}.$$
(25)

From symmetry about the x-axis,

$$\overline{\vec{c}^{\mathrm{f}}}(\chi) = \frac{2}{\pi R^2} \int_{\chi-R}^{\chi+R} \int_0^{(R^2 - (\chi-x)^2)^{1/2}} \vec{F}(\vec{p}) \cdot \hat{\vec{x}} \,\mathrm{d}y \,\mathrm{d}x.$$
(26)

Substituting Eqs. (10), (11), (24), and (25) into Eq. (26) gives

$$\overline{c^{\mathrm{f}}}(\chi) = \frac{2}{\pi R^2} \int_{\chi-R}^{\chi+R} \int_0^{(R^2 - (\chi-d)^2)^{1/2}} \left\{ \frac{k_{\mathrm{f}} |\overline{c^n}| x}{(x^2 + y^2)^{1/2}} + k_{\mathrm{h}} \eta_0 \left[1 + (x^2 + y^2) \left(\frac{\lambda \dot{\varepsilon}}{2\alpha}\right)^2 \right]^{\frac{n-1}{2}} \frac{x \dot{\varepsilon}}{2\alpha} \right\} \mathrm{d}y \,\mathrm{d}x.$$
(27)

Eq. (27) can be numerically integrated to find the average force at $x = \chi$.

2.3. Bulk flow forces

During squeeze flow, the through thickness stress in the material can be divided into the particle stress and the fluid stress. Ericsson et al. [15] have shown that the fluid stresses are much smaller than the particle stresses, therefore the average through thickness stress \overline{p} is the sum of the packing stress, the frictional stress, and the lubrication stress. Toll [16] derived an analytical expression for the packing stress σ_p exerted to squeeze the planar bed of fibers through the thickness

$$\sigma_{\rm p} = -\frac{16}{\pi^2} \phi^2 f \frac{\overline{|\vec{c}^n|}}{d^2}.$$
(28)

Substituting Eq. (6) into Eq. (28) yields an expression for $\sigma_{\rm p}$ as a function of ϕ

$$\tau_{\rm p} = -\frac{512}{5\pi^4} E_{\rm f} f^4 \phi^5.$$
⁽²⁹⁾

A comprehensive model for the squeezing flow of a concentrated long-fiber suspension is by Servais et al. [20]. The fibers are assumed to be slender ($d \ll l$), straight, and of uniform length l. Fiber bundles have an elliptical cross-section with minor axis of length a and major axis of length b. For dispersed fibers, both a and b reduce to r, the radius of the fiber. The fibers are assumed to lie in a plane and have an orientation function f given by Eq. (4). The suspending matrix is assumed to be shear thinning and described by the Carreau relation. The expression found for the average pressure \bar{p} over the squeezing area is

$$\overline{p} = \sigma_{\rm p} \left(1 + k_{\rm f} \frac{l}{3a} \right) + k_{\rm h} \frac{6}{\pi^2} \frac{l^2}{ab^2 \alpha} f \frac{\phi^2}{\phi_{\rm p}^2} \eta_0$$
$$\times \left[1 + \left(l_n \lambda \frac{l}{2\alpha} \dot{\epsilon} \right)^2 \right]^{(n-1)/2} \dot{\epsilon}, \qquad (30)$$

where ϕ_p is the volume fraction of fibers in a fiber bundle which is 0.8 for square packed bundles and 1.0 for dispersed fibers, and

$$l_n = \left(\frac{4}{3(n+3)}\right)^{1/(n-1)} \left(\frac{2n+1}{n}\right)^{n/(n-1)}.$$
(31)

3. Experimental work

The CMT material was manufactured using Zoltek (St. Louis, MO) PANEX33 Carbon fiber chopped to 12.7 mm long, and mixed with 15 denier Fibervisions Type 158 PP fiber, chopped to 5 mm, with no draw and no crimp. The CMT was manufactured at carbon FVF of 10%, 15%, 20%, and 25%, with areal weights of 85–350 g/m². Measurements were made of the through thickness packing stress, the frictional and hydrodynamic lubrication coefficients, and the bulk flow properties of the CMT.

3.1. Viscosity measurement

The complex viscosity, η^* , of the PP was measured using a cone and plate setup over frequencies between 0.5– 100 rad/s on a RMS-800/RDSII rheometer from Rheometrics, Inc. (Piscataway NJ), with a cone angle of 0.1 rad, diameter of 25 mm, and plate separation distance of 0.05 mm. The Cox-Merz rule [18] states that the shear viscosity, η , and complex viscosity, η^* , are identical when evaluated at the same values of $\dot{\gamma}$ and ω , respectively, i.e.:

$$\eta(\dot{\gamma}) = |\eta^*(\omega)|_{\omega = \dot{\gamma}}.$$
(32)

The Carreau equation (9) was fit to the measured data and the constants are listed in Table 2.

3.2. Packing stress measurement

As the CMT material is compressed in the thickness direction the fiber bed will elastically resist the compression. Finding a relation that relates this transverse normal stress to the FVF of the composite is important as an input to the micro-mechanical relations used later in this chapter.

The following assumptions are made when measuring the through thickness stress of the CMT:

(1) Fibers are oriented in a single plane. The wetlay process used to produce the CMT results in a thin web (≈0.5–4 mm thick) of material. Since the 12.7 mm fibers are much longer than the thickness of the material, this assumption is reasonable.

Table 2		
Carreau	parameters	of PP

Parameter	Value	Units
η_0	692.3	Pa s
n	0.7324	_
λο	0.4354	s^{-1}

- (2) The material is statistically homogeneous. In the wetlay process the chopped carbon fibers are introduced as bundles of fibers that are broken apart by the process down to single fibers. While some bundles are not completely broken apart, visual inspection of the mat indicates that the fibers are well dispersed.
- (3) There are a large number of fiber touches along each fiber. For a FVF of 10%, it was found that there are, on average, more than 100 fibers touching a single 12.7 mm fiber. This will tend to prevent any out-ofplane movement of the fibers.
- (4) The compression is quasi-static and allows the matrix to weep out while the fibers do not move in-plane.
- (5) Fibers do not slide relative to each other during compaction.

The packing stress, σ_p , was calculated assuming constant area and the measured load. Eq. (29) is modified when it is fit to experimental data to allow for non-ideal arrangement of fibers by changing the exponent on ϕ to an adjustable parameter, n'.

$$\sigma_{\rm p} = -\frac{512}{5\pi^4} E_{\rm f} f^4 \phi^{n'}.$$
(33)

Eq. (33) can be fit to experimental data by adjusting both f and n'.

Assuming that the squeezing speed is slow enough, the frictional force between fibers will keep them in place and allow the matrix to weep out. The in-plane dimensions of a sample are constant while the height changes.

A heated squeeze flow fixture, shown in Fig. 5, was designed and built. The platens are $152.4 \text{ mm} \times 152.4 \text{ mm} \times 38.1 \text{ mm}$ and each is heated using four 150 mm long by 12.7 mm diameter, 250 W, cartridge heaters. Zircar (Florida, NY) 25.4 mm thick insulation board, type RSLE-57, was mounted on the back and sides of each platen and the platens were then mounted in a ball-bearing die set to help ensure parallelism. Attached to the die set is an LVDT



Fig. 5. Heated squeeze flow fixture.

capable of measuring 0–10 mm displacement. The temperature of each platen is independently controlled using a digital PID temperature controller. The fixture was mounted in a 444 kN MTS servo-hydraulic load frame with hydraulic grips that limited the maximum loading to the 222 kN load range. A routine was written in the MTS TestStar language to control the motion of the actuator during the setup and testing and this routine was run on the computer that controlled the MTS frame. A separate computer running LabVIEW recorded load, displacement, and temperature data at a rate of 0.1–200 Hz. A LabVIEW Virtual Instrument was designed to interface with the fixture and the load frame to record the data.

As the CMT material is compressed in the thickness direction the fiber bed will elastically resist the compression. Finding a relation between this stress and the FVF of the composite is important as an input to the micromechanical relations used later in this work.

Through thickness stress experiments were conducted on C/PP CMT at slow squeezing speeds on 10–25% FVF CMT consisting of 12.7 mm long PANEX35 fibers in PP. Sheets 101.6 mm square were cut from the roll of material and stacked with the machine direction aligned to form each sample with the number of sheets being varied so that all samples had approximately the same mass of fibers. The five samples that were tested are listed in Table 3.

Before the start of testing, the platens of the squeeze flow fixture were treated with the release agent Frecote 700-NC to prevent samples from sticking to the platens. The platens were pre-heated and allowed to come to a uniform temperature of 200 °C, then the empty fixture was closed and held at 1 kN load so the zero reference for the platen separation could be set. The fixture was opened and a sample was inserted, then the MTS program closed the platens to a separation of 10 mm and the samples were held for at least 9 min to allow them to be heated to the temperature of the platens. This was sufficient time for the center of the stack to heat to within 0.1 °C of the set temperature.

Each test began with the load cell on its 44 kN range. The data acquisition was started at a rate of 0.25 Hz and the platens were driven together at a constant speed of 0.005 mm/s. The program that controlled the load frame continued until the load reached 40 kN where the motion of the actuator was stopped to allow the load cell range to be manually changed from the 44 kN range to the

Table 3				
Low speed	through	thickness	stress	samples

FVF	Total mass (g)	Constituent mass (g)		
		Fiber	Matrix	
0.100	50.32	9.19	41.13	
0.100	52.48	9.58	42.90	
0.150	35.50	9.30	26.20	
0.200	28.48	9.53	18.95	
0.250	22.38	8.98	13.40	

222 kN range. Then the test proceeded until a limiting load of 220 kN was reached. After the maximum load was reached the overall load was reduced to 4 kN and the platens were held a fixed distance apart while the fixture cooled. When the temperature was below 130 °C the fixture was opened and readied for the next test.

3.3. Fiber pull-out

3.3.1. Equipment

In order to measure $k_{\rm f}$, $k_{\rm h}$, and α , a fiber pull-out fixture was designed and built. Fig. 6 shows a sketch of the fixture that allows a single fiber to be pulled through the suspension. The fixture was mounted in an oven heated by a 200 W IR heater. Interior dimensions of the oven were approximately 76 mm on a side. The oven was mounted in an electro-mechanical load frame as shown in Fig. 7. A close-up of the fixture is depicted in Fig. 8 that shows how the thermocouple was mounted and the aluminum



Fig. 6. Sketch of the fiber pull-out fixture.



Fig. 7. Pull-out fixture mounted in the load frame.



Fig. 8. Pull-out fixture closeup.

plates and wire clips that were fashioned to constrain the heated sample within the fixture. Temperature was controlled by switching the heater power on and off. A maximum duty cycle of 70% on, 30% off over 5 s was used for the IR heater to prevent it from overheating and burning out. Temperature control to within ± 0.5 °C was achieved using this setup. Since the heating was by IR and the furnace box was not air tight, the air temperature lagged behind the fixture temperature by a significant margin. Therefore the controlling thermocouple is attached directly to the metal fixture. Heating the fixture to 180 °C took about 10 min.

3.3.2. Sample preparation

A 3.1 mm thick panel was manufactured from the 10% FVF material. Pieces $10 \text{ mm} \times 19 \text{ mm}$ were cut from the consolidated panel to fit in the pull-out fixture. In order to maintain consistency in testing, CMT pieces were placed in the fixture so the side of the panel that was on the bottom of the mold sandwiched the traction fiber. All testing had the traction fiber pulled parallel to the transverse direction of the panel. Carbon fibers were extracted from a 48 K tow of unsized Zoltek PANEX33.

3.4. Squeeze flow

High speed tests used 10–25% FVF CMT with 12.7 mm long PANEX35 fibers in PP. The samples were 50.4 mm in diameter, and the number of sheets was chosen so that at full consolidation the sample height equaled 4 mm. The sheets were once again stacked with the machine direction aligned. Each sample was tested at closing speeds of 0.1, 0.2, 0.5, 1.0, 2.0, 5.0, or 8.5 mm/s, for a total of seven samples per FVF. Fig. 9 shows a sample before it is tested.



Fig. 9. Disks of CMT stacked for the squeeze flow experiment.

The platens were coated with Frecote 700-NC before each batch of seven samples. Before testing each individual sample, the platens were also sprayed with Miller Stephenson MS-122 Teflon release agent in an attempt to reduce sliding friction between the platens and the sample. The platens were pre-heated to 180 °C and the zero separation reference was found as above. The sample was inserted into the fixture, then the fixture was closed so the platens were 10 mm apart and held there for 9 min to allow the samples to come to thermal equilibrium.

Depending on the loads expected during the test, the load cell range was set to either 44, 88, or 222 kN before each test. After the hold time was complete the data acquisition system was started and the MTS control program was run to drive the platens together at a constant speed of 0.1-8.5 mm/s and each sample was reduced to approximately 25% of its original thickness. Once the maximum load was reached, the load was reduced to 4 kN and the platens were held a fixed distance apart until the fixture cooled to 130 °C when the fixture was opened and prepared for the next test.

4. Results

4.1. Packing stress

For the low speed tests the experimentally measured transverse normal stress versus FVF is shown in Fig. 10. The curves for the 15%, 20% and 25% FVF material are all coincident with the curve appearing bi-linear in the log-log plot with a knee at $\phi = 0.2094$. Eq. (33) was used to fit both sections of the curve with the left portion of the curve was fit with f = 0.3460 and n' = 4.759 while the right portion was fit with f = 0.1891 and n' = 3.248. The f value of 0.3460 is not very close to the analytical value of $2/\pi = 0.6366$ for randomly oriented fibers, but the exponent, n', is close to the theoretical value of 5 suggesting that there is some preferential alignment of the fibers in the CMT. The presence of the knee in the data is qualitatively predicted by Toll [16]. For brittle fibers such as carbon, he



Fig. 10. Through thickness stress versus FVF for the low speed tests.

suggests that above a certain FVF a loss of stiffness is expected as fibers begin to fracture. Since some fiber segments will be carrying a high load while others will have a low load, there should be a transition from purely elastic to inelastic behavior.

The results for the 10% FVF material differed dramatically from those for the other three FVFs. The recorded load signal from the two 10% FVF samples was erratic and jumped around significantly while the load for the other three FVFs steadily increased, as shown in Fig. 10. Also, the calculated stresses were about 7 times larger than those for the other samples. Fitting Eq. (33) to the data yields f = 0.5692 and n' = 5.168. This value of f is much closer to the theoretical value for randomly oriented fibers. It is possible that there was a manufacturing difference between the 10% FVF material and the other materials. The higher FVF material may have more fiber alignment or bundles of fibers that were not dispersed as well as those in the 10% FVF material.

4.2. Fiber pull-out

Testing was completed for PP CMT at 10.2–15% FVF. A typical force versus time curve is shown in Fig. 11. The time up to 1000 s includes the heat up time (about 600 s) and the initial settling time, from 600–1000 s where the fiber was pulled at 0.0167–0.0333 mm/s until the force leveled out. Any slack in the fiber was removed and the traction fiber was pulled straight during this time. Typically, 10 mm of fiber was pulled through the fixture to ensure steady state. Once the measured force leveled out, the pulling speed was steeped between 0.000167 and 1.67 mm/s. Additionally, part way through the test, at about 1225 s, the direction of the cross-head was reversed to completely unload the traction fiber. From this, any offset in the load signal could be measured and subsequently removed during data analysis.



Fig. 11. Typical force versus time curve for a single fiber pull-out experiment for 10.2% C/PP CMT.

As the pulling speed was stepped up (1300-2195 s), the measured force was seen to increase. At each speed below 0.5 mm/s, the measured force remained constant over time whereas at speeds of 0.5 mm/s and above, the measured force showed a spike at the beginning of each new speed, then the force decayed to a varying value. This can clearly be seen in the data collected at both the 0.5 mm/s and 1.0 mm/s speeds. All of the traction fiber had been pulled through the fixture at 2195 s.

The average force for each speed was calculated and normalized to force per unit length, and results from runs at 10.2–15% FVF are shown in Fig. 12. Eq. (12) was fit to the data using constants in Table 4. A least squares method was then used to find the parameters $k_{\rm f}$, $k_{\rm h}$, and α . Only data at speeds less than 1.0 mm/s were used in the data fit because the average force could not be calculated with certainty for the higher speeds.

Results for the 15% FVF material were harder to obtain. Instead of the traction fiber pulling smoothly through the CMT, the fiber tended to break upon loading. A total of six tests were attempted with the 15% FVF CMT material. Only one of the experiments exhibited a region where a steady force was measured, and that only lasted for about 70 s before the fiber broke. All other tests had the fiber break and no steady force regions were observed.



Fig. 12. Force versus pulling speed for C/PP CMT.

Table 4	
Values used in Eq.	(12)

Parameter	Value	Units
d	7.2	μm
E_{f}	228	GPa
f	0.5692	_
α	2.797	μm
$k_{\rm f}$	0.02451	_
$k_{ m h}$	0.001262	mm^2



Fig. 13. Force decay results for single fiber pull-out, fit with Eq. (12).

Results from the force-decay experiment are shown in Fig. 13. Eq. (12) was fit to the data with v = 0, and $k_f = 0.02380$ was found using a least squares method, which is close to the value obtained from the constant speed experiments.

5. Discussion

5.1. Squeeze flow

Fig. 14 shows a typical sample after squeezing. The originally circular disk is deformed into an elliptical disk. The black concentric circles in the middle of the sample were drawn on the top disk before testing. If there was no friction between the sample and platens, the circles would have deformed into ellipses along with the rest of the sample. Since the circles are still the same size and shape as they were before testing, a no-slip condition between the platen and the sample prevails. The tests were run at constant



Fig. 14. Squeezed sample.



Fig. 15. Load versus platen separation for the 2 mm thick, 10% FVF samples; the left figure shows entire range and the right figure shows a zoomed in region.



Fig. 16. Load versus platen separation for the 4 mm thick, 10% FVF samples.

velocity and it was found that the load frame could maintain a constant closing speed until the squeezing force was too large.

Figs. 15 and 16 depict the force versus displacement curves for the 10% FVF C/PP material. The shape of the

curves is similar to that of the low FVF GMT and SMC [1–3,21]. The force is very low as the platens approach h_0 , then near h_0 the force rises quickly while the material is consolidated. As squeezing continues the force shows a plateau, then increases sharply near the end of the test.

The 15% FVF material behaves similar to the 10% FVF material. Results from the 2 mm and the 4 mm thick samples are shown in Fig. 17.

The 20% and the 26.2% FVF materials show a different behavior (Figs. 18 and 19, respectively) than the lower FVF material. As the platens approach h_0 , the load rises quickly to a local maximum, then drops as the squeezing continues. As the test progresses the load begins to climb again. As far as we could discern, this behavior has not been reported in the literature for any other fiber filled polymer, though a similar curve was obtained for squeeze flow of mudcakes by Sherwood et al. [22]. The cause of this initial load spike was determined to be breaking of the fibers. It has been known anecdotally that the maximum fiber length in a GMT part is limited both by the distance the material must flow during molding and by its FVF. The GMT used in other studies [1,5,8,11] is about 8–13% FVF while the



Fig. 17. Load versus platen separation for the 15% FVF samples.



Fig. 18. Load versus platen separation for the 20% FVF samples.



Fig. 19. Load versus platen separation for the 26.2% FVF samples.

CMT shows the load spike beginning at 15–20% FVF. It is possible that glass fibers do not exhibit this trait, or researchers have not tested GMTs with high enough FVF.

Two squeeze-flow samples at each FVF had the matrix burned off after the squeezing test. Dissection and visual inspection of the resulting carbon fiber mats found an increasing amount of very short (<1-4 mm) fibers as the FVF of the sample increased. Photographs of the dissected specimens are shown in Figs. 20-23. For the 10% FVF sample (Fig. 20), the mat had very few visibly broken fibers and the mat had structural integrity from the many layers of long overlapping fibers. The 15% FVF (Fig. 21) sample showed some very short (<1 mm long) fibers that are visible in Fig. 21 against the white background. When the FVF was increased to 20% the dissection resulted in clumps of fibers about 2-5 mm across, and even more 'dust' from the very short, broken fibers. In contrast to the 10% FVF sample, the 26.2% FVF sample shown in Fig. 23 was very fragile, and broke apart much more easily. During dissection of this mat many small clumps of short fibers about 1-3 mm long were apparent, along with much more very fine carbon 'dust' than in the other material. In all samples there was a range of fiber lengths from 'dust' to 12.7 mm indicating that some fiber breakage occurred in all samples and that this fiber breakage is not uniform within a sample. Additional work needs to be done to determine the fiber length distribution after squeeze flow.

Fig. 24 compares the measured average squeezing stress \overline{p} with that calculated from Eq. (30). While other researchers have shown that fiber orientation can be affected by the flow [23–25], the orientation function f is assumed to be constant in this case due to the relatively short (5l) flow lengths involved. For the 10% FVF material shown in Fig. 24(a), both the theoretical prediction and the data show increasing trends with the same slope as the squeezing strain rate increases, but the theory over-predicts the measured force by a factor of about 8-10. As the FVF is increased from 10% to 26.2%, the predicted stress increases by almost two orders of magnitude and the character of the stress versus strain rate curve changes from linear to nonlinear. As the FVF increases the measured stress does not increase as fast as the predicted stress, and for the 26.2% FVF material the predicted and the experimental values do not agree at all.



Fig. 20. FVF sample (10%) after squeezing and burn-off (scale division = 1 mm).



Fig. 21. FVF sample (15%) after squeezing and burn-off (scale division = 1 mm).

The maximum tensile strength of the fiber is 3.8 GPa [26,27]. If the flow causes stresses greater than this in the fiber, it will break until the length is reduced below a critical length. For each FVF, the critical fiber length l_{max} was calculated by setting $\sigma_{\text{f,max}} = 3.8$ GPa in Eq. (20).

The calculated maximum critical fiber lengths at each FVF of the CMT are listed in Table 5. Since the fiber in this test began as 12.7 mm long, the calculated values of $l_{\rm max}$ indicate that fiber breakage would not occur in any of the material. Since significant breakage was observed, there must be other forces acting on the fibers that this formulation does not consider.

It is known that the length of the reinforcing fiber affects both the tensile strength and the impact strength [20], thus, it is important to keep the fibers as long as possible in parts molded with both GMT and CMT. Further studies should be conducted to fully quantify the forces acting on fibers in the CMT so that the average effective fiber length, or fiber length distribution, can be calculated for a given FVF. Designers could then pick appropriate fiber sizings and matrix materials that will reduce the frictional effects in order to obtain the longest fiber length during manufacturing.

5.2. Fiber pull-out

Results from the 10.2-12.1% FVF C/PP CMT material were fit with Eq. (12) very well, as shown in Fig. 12 for pulling speeds of 0.000167-0.5 mm/s. At higher speeds the



Fig. 22. FVF sample (20%) after squeezing and burn-off (scale division = 1 mm).



Fig. 23. FVF sample (25%) after squeezing and burn-off (scale division = 1 mm).

experimental data was difficult to interpret (see Fig. 11). When the speed was stepped up, the measured force showed a peak then dropped-off to an unsteady value. Since the fiber bed and traction fiber should have been at equilibrium due to the slower speed testing, it is possible that there is rearrangement of the fiber bed when the speed



Fig. 24. Z squeezing stress versus squeezing strain rate for the high speed testing. The theoretical stress curves were calculated using Eq. (30) with l = 12.7 mm and f = 0.3460.

Table 5Calculated maximum critical fiber lengths for different FVF

ϕ	l _{max} (mm)
0.100	172
0.150	124
0.200	85
0.262	48

is >0.5 mm/s. As the fiber bed changes configuration, the load would change, making the test invalid.

The 15% FVF samples were more difficult to work with than the lower FVF samples as the traction fiber tended to break rather than smoothly slide through the CMT. This is due to the higher FVF that leads to both a larger number of fibers touching the traction fiber, and a higher normal force at each fiber–fiber touch point. Eq. (12) predicts a five fold increase in the static friction force when the FVF is increased from 10% to 15%. Even with these difficulties, the 15% FVF data appears to be reasonable and brackets the predictions of Eq. (12). The extra scatter in the 15% FVF data seems reasonable since the remaining fiber length must be estimated.

6. Conclusions

A single fiber pull-out fixture was successfully designed, built, and used to measure the fiber–fiber frictional coefficients in CMT. These coefficients were used in relations to estimate the bulk flow of the CMT.

Squeeze flow experiments were completed on 10–26.2% FVF C/PP CMT material. The low FVF material showed flow behavior similar to that reported for GMT. As the FVF was increased, the squeezing force in the experiments showed a pronounced localized peak as the flow began. It is hypothesized that the flow stresses exceeded the tensile strength of the fibers and dissection of the samples showed increasing fiber breakage as the FVF was increased. The axial stress in the fibers was calculated and compared to the tensile strength of the fibers, and it was found that

the proposed relation based on the Servais et al. equation does not predict fiber breakage. Further study of the forces acting on carbon fibers during consolidation and flow is needed to fully understand the mechanics that govern the fiber fracture in the CMT.

The flow characteristics of the CMT were investigated. Two fixtures were designed and manufactured in the course of this study. The first fixture allowed us to measure the force required to pull a single carbon fiber through the molten CMT. This test measured the frictional coefficients at the fiber-fiber touch points. The force versus speed curves of the CMT were fitted with a model originally developed for GMT, and the fit was found to be very good. The second fixture constructed for this portion of the study was a heated parallel plate plastometer that was used to measure the through thickness packing stresses of the fiber bed and the bulk flow properties of the CMT. A method of estimating the axial stress in the fiber during flow of the CMT was developed. It was found that the axial stress in the fiber depends on the FVF of the CMT, and if the FVF is high enough, the axial stress in the fiber can exceed its tensile strength.

References

- Kotsikos G, Bland JH, Gibson AG, Chandler HW. Squeeze flow testing of glass mat thermoplastic material. Composites Part A 1996;27A(12):1195–200.
- [2] Kotsikos G, Gibson AG. Investigation of the squeeze flow behavior of sheet moulding compounds (SMC). Composites Part A 1998:29A:1569–77.
- [3] Kotsikos G, Bland JH, Gibson AG. Rheological characterization of commercial glass mat thermoplastics (GMT) by squeeze flow testing. Polym Compos 1999;20(1):114–23.
- [4] Bush SF, Torres FG, Methven JM. Rheological characterization of discrete long glass fiber (LGF) reinforced thermoplastics. Composites Part A 2000;32:1421–31.
- [5] Dweib MA, Ó Brádaigh CM. Extensional and shearing flow of a glass-mat-reinforced thermoplastics (GMT) materials as a non-Newtonian viscous fluid. Compos Sci Technol 1999;59:1399–410.
- [6] Servais C, Luciani A, Månson J-AE. Fiber-fiber interaction in concentrated suspensions: dispersed fiber bundles. J Rheol 1999;43(4):1005–18.
- [7] Dweib MA, Ó Brádaigh CM. Anisotropic modeling of isothermal squeezing flow of glass-mat reinforced thermoplastics (GMT). Polym Compos 1998;19(5):588–99.
- [8] Servais C, Månson J-AE. Fiber-fiber interaction in concentrated suspensions: disperse fibers. J Rheol 1999;43(4):991–1004.

- [9] Toll S. Note: on the tube model for fiber suspensions. J Rheol 1993; 37(1):123–5.
- [10] Toll S, Månson J-AE. Dynamics of a planar concentrated fiber suspension with non-hydrodynamic interaction. J Rheol 1994;38(4): 985–97.
- [11] Törnqvist R, Sunderland P, Månson J-AE. Non-isothermal process rheology of thermoplastic composites for compression flow moulding. Composites Part A 2000;31:917–27.
- [12] Törnqvist R, Servias C, Ebinger J-C, Månson J-AE. The role of fiber bundle interaction forces in non-isothermal compression molding of thermoplastic composites. Polym Compos 2001;22(1):1–10.
- [13] Lu Y. Mechanical properties of random discontinuous fiber composites manufactured from the wetlay process. Master's thesis, Virginia Polytechnic Institute and State University. Available from: http://scholar.lib.vt.edu/theses/available/etd-08132002-163349/, August 2002.
- [14] Caba AC. Characterization of carbon mat thermoplastic composites: flow and mechanical properties. PhD thesis, Virginia Polytechnic Institute and State University, September 2005.
- [15] Ericsson KA, Toll S, Månson J-AE. Sliding plate rheometry of planar oriented concentrated fiber suspension. Rheol Acta 1997;36:397–405.
- [16] Toll S. Packing mechanics of fiber reinforcements. Polym Eng Sci 1998;38(8):1337–50.
- [17] Toll S, Månson J-AE. Elastic compression of a fibre network. J Appl Mech 1995;62:223–6.
- [18] Baird DG, Collias DI. Polymer processing. New York: John Wiley & Sons Inc; 1998.
- [19] Servais C, Månson J-AE. The relationship between steady-state and oscillatory shear viscosity in planar randomly oriented concentrated fiber suspensions. J Rheol 1999;43(4):1019–31.
- [20] Servais C, Luciani A, Månson J-AE. Squeeze flow of concentrated long fiber suspensions: experiments and model. J Non-Newton Fluid Mech 2002;104(2–3):165–84.
- [21] Adams MJ, Edmondson B, Gaughey DG, Yahya R. An experimental and theoretical study of the squeeze-film deformation and flow of elastoplastic fluids. J Non-Newton Fluid Mech 1994;51:61–78.
- [22] Sherwood JD, Meeten GH, Farrow CA, Alderman NJ. Squeeze-film rheometry of non-uniform mudcakes. J Non-Newton Fluid Mech 1991;39(3):311–34.
- [23] Advani SG, Tucker III CL. A numerical simulation of short fiber orientation in compression molding. Polym Compos 1990;11(3): 164–73.
- [24] Jackson WC, Advani SG, Tucker CL. Predicting the orientation of short fibers in this compression moldings. J Compos Mater 1986;20:539–57.
- [25] Advani SG, Tucker III CL. The use of tensors to describe and predict fiber orientation in short fiber composites. J Rheol 1987;31(8):751–84.
- [26] PANEX 33 chopped carbon fibers. Zoltek product data sheet PX33CF. Available from: http://www.zoltek.com/panex_products/ pan33_chopped.pdf, September 1999.
- [27] PANEX 35 chopped carbon fibers. Zoltek product data sheet PX35CA. Available from: http://www.zoltek.com/panex_products/ PX35CA0250-15.pdf, June 2002.