# Constitutive Modeling of Fiber Composites with a Soft Hyperelastic Matrix

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## Abstract

This paper presents an experimental and numerical study of unidirectional carbon fiber composites with a silicone matrix, loaded transversally to the fibers. The experiments show nonlinear behavior with significant strain softening under cyclic loading. The numerical study uses a plane-strain finite element continuum model of the composite material in which the fiber distribution is based on experimental observations and cohesive elements allow debonding to take place at the fiber/matrix interfaces. It is found that accurate estimates of the initial tangent stiffness measured in the experiments can be obtained without allowing for debonding, but this feature has to be included to capture the non-linear and strain-softening behavior.

*Keywords:* 

Fiber composites, Hyperelastic matrix, Strain softening, Random microstructure

## 1. Introduction

New designs for all-composite lightweight deployable structures require the material to undergo severe stowage curvatures without damage and a variety of schemes for achieving this behavior have been proposed. Two of the most promising approaches make use of materials that either remain in the elastic

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range while undergoing such extreme deformations (Mejia-Ariza et al., 2006; Rehnmark et al., 2007; Datashvili et al., 2010) or have a weakly linked matrix that becomes much softer above its transition temperature (Campbell et al., 2005; Barrett et al., 2006). These composite materials are often described as <u>memory matrix composites</u>. In this paper we will focus on the first of these two approaches and specifically on composites of unidirectional carbon fibers embedded in a silicone matrix.

One of the problems of greatest practical importance is determining the stresses and strains that occur when thin sheets of these materials are folded. This problem was considered by Murphey et al. (2001) and then by Francis et al. (2007). For memory matrix composites they found that fiber microbuckling provides a key stress relief mechanism that allows the fibers to survive without damage when a sheet is folded to extreme curvatures.

The folding behavior of fiber composites with a soft hyperelastic matrix was recently studied by López Jiménez and Pellegrino (2011), who showed that during folding the matrix may be subject to very large strains, with maximum principal strains in excess of 200% near the fibers in sheets with a thickness on the order of 100  $\mu$ m and a fiber volume fraction of around 50%. Cyclic bending tests carried out by López Jiménez and Pellegrino (2011) also showed significant strain softening, meaning that the bending moment needed to impose a given curvature decreases during the first few cycles. The measured loss of stiffness for a given curvature was largest after the first cycle and gradually decreased in subsequent cycles. The details of the damage mechanisms were not investigated.

Instead of considering the folding behavior, as in previous studies, this paper focuses on the behavior under transverse tension, i.e. tension perpendicular to the fibers. This loading case is of particular interest because it does not produce fiber microbuckling, thus eliminating the effects of geometric nonlinearity, but instead emphasizes the effects of material nonlinearity including any effects related to material damage. It will be shown that experimental results obtained for this type of loading can be used to calibrate simulations in which a source of material damage, such as fiber debonding, is considered. Hence, both experimental and numerical studies of such tensile behavior under repeated loading will be presented in this paper.

The paper is laid out as follows. Section 2 presents a review of the relevant literature. Section 3 describes the construction of the test specimens and the test procedures, and presents the experimental results. Section 4 presents the finite element model that was set up to capture the behavior of the test specimens, with the specifics of the fiber arrangements used in the simulations presented in Section 5. The results of the numerical simulations are then presented and compared with experimental data in Section 6. Section 7 compares the behavior of fiber composites with a soft hyperelastic matrix with the behavior of composites with a standard epoxy matrix; it is shown that the incompressibility of the matrix is responsible for important differences in behavior. Finally, Section 8 summarizes the findings of this research and concludes the paper.

#### 2. Background

There is an extensive literature on the large strain behavior of fiber reinforced composites. Merodio and Ogden (2002, 2003) produced a model based on Spencer (1972), using two invariants to model the effect of the incompressible fibers on the strain energy of the material. Several biological materials, such as the cornea and blood vessels, fit the description of stiff fibers in a very soft matrix and models based on such an approach have been used by Holzapfel et al. (2000); Pandolfi and Manganiello (2006); Pandolfi and Holzapfel (2008). The second order homogenization theory developed by Ponte Castañeda (2002) has also been applied to fiber-reinforced hyperelastic materials. This approach is still under development, see for example Lopez-Pamies and Ponte Castañeda (2006), Agoras et al. (2009) and Lopez-Pamies and Idiart (2010); a comparison to numerical results has been presented in Moraleda et al. (2009b).

The models outlined above neglect the strain softening observed in materials composed of a soft matrix with hard inclusions, such as biological tissue, particle-reinforced rubbers, or solid rocket fuel. In rubbers strain softening is usually known as Mullins effect and in general the degree of strain softening increases with the concentration of reinforcement. Different mechanisms have been proposed to explain this effect, including debonding of particle and matrix, rupture of the filler, chain disentanglement or an increase of the rubber crystallization due to stress concentration (Diani et al., 2009).

Silicone rubber is more stable than other elastomers and shows very little to no strain softening when pure (Meunier et al., 2008), however strain softening is still observed in reinforced silicone rubber (Machado et al., 2010; López Jiménez and Pellegrino, 2011). López Jiménez and Pellegrino (2011) carried out bending tests on silicone specimens uniaxially reinforced with carbon fibers and found that an irreversible softening of the moment-curvature relationship takes place every time the curvature applied to the specimen is increased beyond the maximum value achieved in its prior history, see Figure 1. The process is also characterized by a marked hysteretic behavior under cyclic loading.



Figure 1: Moment-curvature relationship for a specimen with  $V_f = 55\%$ , showing strain softening under cycling loading, taken from López Jiménez and Pellegrino (2011).

In the present paper it is assumed that the main reason for strain softening

is debonding/dewetting between matrix and reinforcement. This approach was explored by Tong and Ravichandran (1994) and Ravichandran and Liu (1995), who provided expressions for the elastic properties of particle composites undergoing damage by dewetting, assuming a linear response of the material. Debonding was incorporated in the finite element analysis of elastomers reinforced with particles (Zhong and Knauss, 1997, 2000) and fibers (Moraleda et al., 2009) by introducing cohesive elements. The results were in good qualitative agreement with the behavior observed experimentally, but to the authors' knowledge no direct comparison of experimental results and predictions has been published.

#### 3. Experiments

This section details the specimen fabrication and characterization. In particular, the fiber distribution in three sets of specimens with different fiber volume fractions is characterized through micrographs and the results are used in Section 5 to set up finite element models. The section also presents a set of tension tests that show nonlinear behavior and link strain softening to fiber volume fraction.

## 3.1. Test Specimen Fabrication

The fibers used were HTS40-12K, produced by Toho Tenax (retrieved August 2010) and supplied by the Itochu Corporation as a uniaxial dry fabric with an areal weight of 40 g/m<sup>2</sup> for a single ply. This fabric had been made by spreading each tow to a width of approximately 20 mm, leaving no visible separation between the individual tows. The properties of the fibers are listed in Table 1.

The matrix used was CF19-2615, produced by NuSil Silicone Technology (March 2007). It is a two part, optically clear silicone chosen for its low viscosity, see Table 1, in order to facilitate flow between the fibers. The modulus and elongation properties provided by the manufacturer were verified experimentally through uniaxial tests (López Jiménez and Pellegrino, 2011). Failure typically occurs at elongations in the range 120-140% and for a Cauchy stress of

Fiber properties: HTS40		
Diameter	$7 \ \mu m$	
Tensile modulus	$240~\mathrm{GPa}$	
Density	$1.77~{\rm g/cm^3}$	
Matrix properties: CF19-2615		
Viscosity (part A)	1300 mPa s	
Viscosity (part B)	$800~\mathrm{mPa}$ s	
Density	$0.96~{\rm g/cm^3}$	
Manufacturer supplied tensile modulus	$0.8 \mathrm{MPa}$	
Manufacturer supplied elongation at failure	100%	

#### Table 1: Material properties

approximately 1.25 MPa. Failure criteria for silicone rubber under multi-axial loading conditions have yet to be developed. In particular there is no information about cavitation (Gent and Lindley, 1959), a failure mechanism consisting of the sudden appearance of internal flaws, which is often associated with the failure of rubbers. For this reason, the value of the stretch at failure cannot be readily applied to other loading conditions, although it will be used as an indication that the material may be close to failure.

In the fabrication process, the two parts of the silicone were mixed and placed under vacuum to extract any air bubbles. The mixture was then poured over the fibers and the material was cured for 30 minutes at 150°C. Internal vacuum and external pressure (up to 85 psi, 0.586 MPa) were applied during curing to increase the fiber volume fraction and to help consolidate the material. No significant porosity was observed in the specimens. More details on the fabrication process are provided in López Jiménez and Pellegrino (2011).

#### 3.2. Specimen Characterization

The actual distribution of the fibers in a thin sheet of composite material is different from the regular lattices that have been used as idealizations for finite element or analytical modeling (López Jiménez and Pellegrino, 2011). To set up a high fidelity model it is necessary to ensure that the fiber distribution captures the main attributes of the actual microstructure.

The first step in this process is obtaining micrographs of the cross section of a specimen. A standard micrograph would require a perfectly flat surface, which is traditionally achieved by embedding the specimen in epoxy, and then grinding and polishing it. However, this is not possible in the case of fiber composites with a soft matrix. Instead, the material was cut with a razor blade and then placed end-on under a Nikon Eclipse LV100 microscope set at an amplification of 50x and attached to a Nikon DS-Fi1 digital camera. The cut surface was not sufficiently flat to lie within the depth of field of the lens, hence several images of the cross section were taken at different focus distances and were then processed as an image stack using Adobe Photoshop CS4 (2008). A sharp image of the whole cross-section was thus obtained.

Next, a characterization of the fiber distribution in the cross-section was obtained. Several studies have proposed parameterizations based on the Voronoi tessellation of the fibers and Figure 2 shows such a tessellation obtained with the voronoi command in Matlab after providing the centroids of the fibers as an input. Different authors have used the distance from the fiber to the boundary of the cell (Davy and Guild, 1988) or the topological entropy of the polygonal network (Pyrz and Bochenek, 1998). Other researchers have suggested functions based on the position of the inclusions, such as the power spectral density of the indicator function (Povirk, 1995) or the radial distribution function (RDF), which measures the probability of, given the position of a fiber, finding another fiber in an annulus of radius r and r + dr centered on the first one (Rintoul and Torquato, 1997).

The function used in the present work was the second-order intensity function K(r) (Pyrz, 1994a,b), defined as the number of fibers expected to lie within a radial distance r from an arbitrary fiber, normalized by the overall fiber density. This function is proportional to the integral over the radius of the RDF and the two functions provide the same information. The algorithm used to



Figure 2: Micrograph showing Voronoi tessellation of fibers.

construct a finite element model based on the value of K(r) observed in the micrographs is detailed in Section 5.

The Voronoi tessellation can also be used to study in more detail the fiber volume fraction, which will be denoted as  $V_f$ . In particular, the difference between global and local volume fractions can be established by calculating the volume fraction of each of the Voronoi cells. Figure 3 shows its histogram, with a vertical line marking the average value (55%, for a three ply specimen), which agrees with the value obtained from weight measurements. This figure shows that the most common values of  $V_f$  are in the range 60% - 65%. The peak at very low volume fractions corresponds to fibers next to the surface of the material, whose Voronoi cells extend to infinity.

Three different sets of specimens made from three plies were characterized and the same specimens, with  $V_f$  equal to 65%, 50%, and 22%, were studied by means of experiments and simulations. Note that because the total number of fibers in a unit width of each ply is the same in all cases, <u>specimens with</u> lower fiber volume fraction are thicker and hence have a lower homogenized <u>longitudinal modulus</u>. Figure 4 shows micrographs of the two extreme cases: note that the fiber distribution in the specimen with  $V_f = 22\%$  is very irregular and there is a high degree of fiber clustering.

#### 3.3. Experimental Results

100 mm long by 25 mm wide specimens made from either three or four plies were made and tested, see Figure 5, with an Instron 5569 materials testing



Figure 3: Histogram of volume fractions of Voronoi cells produced from micrographs. The vertical line marks the average value.



Figure 4: Specimens with three plies and: (a)  $V_f=65\%$  and 150  $\mu$ m average thickness, (b)  $V_f=22\%$  and 300  $\mu$ m average thickness.

machine with a 10 N load cell at a uniform diplacement rate of 0.1 mm per minute.



Figure 5: Experimental setup for tension test in direction transverse to the fibers.

The extension,  $\delta$ , of a gauge length defined by two retro-reflecting strips at a distance  $L \approx 25$  mm was measured using an Epsilon LEO1 laser extensioneter. The corresponding average engineering strain was computed from

$$\overline{\epsilon_y} = \frac{\delta}{L} \tag{1}$$

The average Piola-Kirchhoff stress  $\overline{\sigma_y}$  was obtained by dividing the load F by the initial cross-sectional area A of the specimen

$$\overline{\sigma_y} = \frac{F}{A} \tag{2}$$

Figures 6-7 show a series of stress vs. strain plots from these experiments.

Figure 6 shows the response of four nominally identical specimens, subjected to three cycles of increasing maximum strain followed by further extension until failure. The maximum strains applied in the three cycles corresponded to  $\overline{\epsilon_y} = 0.01, 0.02$ , and 0.03 over the whole specimen, as measured by the testing

machine. The corresponding strains measured with the laser extensioneter were similar.

The responses of the four specimens were similar during the first two cycles. Failure typically began to occur towards the end of the second cycle and hence the behavior of the specimens varied widely from the third cycle. The second specimen, Figure 6 b, was so damaged that the third cycle could not even be started. The other three specimens survived the three cycles, although with noticeable damage in the case of the fourth specimen. The first and fourth specimens failed after the first three cycles; the larger failure strain of the first specimen is explained by the position of the main crack producing failure, which was inside the laser gauge for the first specimen, and outside for the fourth one, leading to a higher measured strain. Tests carried out 24 hours after a previous test showed no signs of recovery.

The observed variability in the failure behavior shows an essential difference between composites with a silicone matrix vs. those with a standard epoxy matrix. In the latter case failure is sudden, whereas in the former case stable cracks form in the matrix and a load increase is needed for the cracks to grow. Hence composites with a silicone matrix often retain much of their stiffness even after visible cracks have formed; physically this behavior can be explained by a small misalignment of the fibers that results in a few fibers bridging across the crack. These fibers become aligned with the loading direction and, since the fibers are much stiffer than the matrix, even a small number of fibers is then able to sustain the same loads as the rest of the specimen.

In the case of specimens with a lower fiber volume fraction, the behavior is approximately linear for a much larger range of strains and the hysteresis is also much lower. These specimens reach a much higher strain at failure, see Figure 7, because the matrix is less constrained by the fibers. This results in lower stress concentrations, which are the main source of strain softening and nonlinearity.



Figure 6: Stress vs. strain under transverse loading for four different three-ply specimens with  $V_f = 65\%$ . The specimens were tested until failure occurred: (a) the first specimen failed after the three cycles, inside the measurement gauge, (b) the second specimen failed during the second cycle, (c) the third specimen failed after the three cycles and outside the measurement gauge and (d) the fourth specimen failed after the three cycles, with noticeable damage occurring during the third cycle.



Figure 7: Stress vs. strain under transverse loading for three three-ply specimens with  $V_f = 22\%$  subject to three load-unload cycles with a maximum strain  $\overline{\epsilon_y} = 0.1$ , followed by monotonic extension up to failure.

## 4. Finite Element Model

A finite element model of the test configuration was created using the package ABAQUS/Standard (2007). Plane strain conditions and non-linear geometry were assumed and both matrix and fibers were modeled as elastic continua, using plane strain elements. In order to reduce the computational effort, only a representative volume element (RVE) was modeled using periodic boundary conditions. The size of the RVE, the fiber arrangement and the mesh size were varied during the analysis, as discussed in detail in Section 5.

The RVE is defined with periodic boundary conditions such that the faces of a rectangular piece of composite material of size  $L_1 \times L_2$  deforms periodically in the x and y directions, where both x and y are perpendicular to the direction of the fibers and y is aligned with the direction of extension of the specimens tested in Section 3. The meshing of opposite faces of the RVE is identical, and hence the boundary conditions can be applied directly on the edge nodes using the EQUATION command in ABAQUS. The boundary conditions are expressed mathematically as:

$$u(x,0) = u(x,L_2)$$
 (3)

$$v(x_1,0) - v(x_1,L_2) = v(x_2,0) - v(x_2,L_2)$$
(4)

$$v(0,y) = v(L_1,y)$$
(5)

$$u(0, y_1) - u(L_1, y_1) = u(0, y_2) - u(L_1, y_2)$$
(6)

where u and v are the displacements in the x and y direction, respectively. The above boundary conditions imply that the dimensions of the RVE are allowed to change with respect to the undeformed configuration, but they have to remain uniform through the model. The loading of the element was imposed as a prescribed relative displacement between two opposite faces. No constraint was applied in the direction of the fibers.

Two different sets of elements were used to model the fibers and the matrix. In the case of the fibers, which are modeled as linearly elastic materials, the elements used were CPE3, linear triangular elements for plane strain. The matrix was modeled as a hyperelastic purely incompressible material, which requires the use of elements with hybrid formulation. The elements chosen in this study were the linear quadrilateral hybrid elements CPE4H, with a mesh sufficiently fine to provide at least two elements between each fiber. Several other types of elements were investigated and were excluded for the following reasons. The linear hybrid triangular elements (CPE3H) resulted in lack of convergence and hence no analysis could be carried out to completion. The quadratic version, CPE6H, gave macroscopic results in agreement with the quadrilateral elements but resulted in high stress concentrations and a check-board stress pattern, see Figure 8. This was the case even for much finer meshes than those used for quadrilateral elements, and so the CPE6H elements were discarded.

The second order quadrilateral elements (CPE8H) gave no problems for a sufficiently fine mesh, but the linear version of these elements (CPE4H) pro-



Figure 8: Distribution of stress  $\sigma_x$  in a model with  $V_f = 50\%$  and CPE6H elements, showing check-board pattern.

duced better results for the same number of nodes. Generalized strain elements (CPEG3 and CPEG4H) were also tested, in order to allow deformation in the out-of-plane direction but the linear response was found to be practically the same as for plane strain elements, due to the high stiffness of the fibers.

The fibers were modeled as a linear elastic, orthotropic material. The transverse modulus was given a value of 20 GPa, typical of carbon fiber. It is roughly one order of magnitude lower than the axial modulus provided in Table 1. The value of the transverse modulus was varied by up to an order of magnitude to study the influence of this parameter, and was found to be negligible.

The matrix was modeled as a hyperelastic solid using a Gent (2005) potential modified to avoid singularities at large stretches. Although physically unrealistic, large stretches may occur in the course of an equilibrium iteration (López Jiménez and Pellegrino, 2011). This model was implemented in ABAQUS as a user-defined material, using the user subroutine UHYPER. The potential is defined in terms of the principal stretches  $\lambda_i$  and is given by:

$$W = -C_1 J_m \ln\left(1 - \frac{J_1}{J_m}\right)$$

$$+ C_2 \ln\left(\frac{J_2 + 3}{3}\right) \quad \text{if} \quad J_1 \le 0.9 J_m$$

$$(7)$$

$$W = -C_1 J_m \ln(0.1) + \frac{C_1 (J_1 - 0.9 J_m)}{0.1}$$

$$+ 0.5 \frac{J_1 (J_1 - 0.9 J_m)^2}{0.01 J_m}$$

$$+ C_2 \ln\left(\frac{J_2 + 3}{3}\right)$$
if  $J_1 > 0.9 J_m$ 
(8)

where

$$J_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3$$
 (9)

$$J_2 = \lambda_1^{-2} + \lambda_2^{-2} + \lambda_3^{-2} - 3 \tag{10}$$

with the parameters  $C_1 = 0.1015$  mJ,  $C_2 = 0.1479$  mJ and  $J_m = 13.7870$  obtained by fitting the model to uniaxial tests on pure silicone specimens. It has been shown by Ogden et al. (2004) that the parameters of the Gent potential can be predicted quite accurately from only unixaxial data, which is not the case with other hyperelastic potentials, such as the Ogden or Mooney-Rivlin potentials.

The model described so far includes no source of damage or dissipation. In order to capture the stress softening observed in the experiments, cohesive elements were introduced between the fibers and the surrounding matrix, as a way to model fiber debonding. The standard two-dimensional cohesive elements in ABAQUS/Standard, COH2D4, were used. The behavior of these elements is described in Figure 9. In tension, these elements keep their initial stiffness until a traction  $t_0$  is reached, corresponding to a separation  $\delta_0$ . The stiffness then decreases until it reaches a value of zero for a deformation  $\delta_f$ . The behavior of a damaged element can follow a straight line, as in the figure, or an exponential. In the case of unloading, the relationship follows a straight line passing through the origin. More details on the implementation of cohesive elements are provided in Section 6.2. Note that in compression the elements available in ABAQUS always retain the initial stiffness.



Figure 9: Typical traction-separation response in a cohesive element.

## 5. Geometry Details

This section details the geometry of the RVEs used in this study. It includes the reconstruction process that generates the fiber distribution and a sensitivity study of key geometric parameters such as the fiber density and the minimum distance between fibers.

## 5.1. Fiber Arrangement

The Random Sequential Adsortion algorithm by Rintoul and Torquato (1997) was used to generate the microstructure of the composite material. It is an iterative process that tries to minimize a potential E that quantifies the difference between a trial microstructure and an actual microstructure observed experimentally. E is usually defined using the parametrization function f(r) as  $E = \sum_{k} (f(r_k) - f_0(r_k))^2$ , where  $f_0(r)$  is the value of the function for the reference configuration, and the sum is made over the discretization intervals.

The algorithm works as follows. In every iteration a fiber is picked randomly and a random displacement is applied. The potential E' is calculated for this new configuration. The displacement is accepted according to the probability

$$P = \begin{cases} 1 & \text{if } \Delta E \le 0\\ e^{\frac{E-E'}{A}} & \text{if } \Delta E > 0 \end{cases}$$
(11)

where  $\Delta E = E' - E$  and A is a parameter that controls how fast the system should evolve (a value of 0.05 was used). This means that the algorithm would accept some of the displacements in which the energy increases slightly, but almost none implying a large increment. The iterations are repeated until the system converges to a stable value of E.

Two different potentials were used in this research, and the algorithm finally implemented is described in the next sections.

#### 5.1.1. Initial Mesh

First, the fibers were randomly distributed with a hard-core process. This is a Poisson random process in which a limitation on the minimum distance between the centers of fibers is introduced. In cases with high fiber volume fraction not all the fibers can be located randomly as a jamming condition is reached. The jamming limit is given by Tanemura (1979) as 0.547 and this value is lowered if the imposed distance is higher than the fiber diameter. This is the case here as we need to make space for at least a small amount of matrix between neighboring fibers. In practice, jamming is observed when  $V_f$  approaches 50%.

If after 1000 attempts a fiber has not been accepted, the last attempted position is accepted even if the fiber overlaps with other fibers. In such cases the Random Sequential Adsortion algorithm is used, with the potential energy

$$E = \sum_{i} \sum_{j} \left( \left( \frac{100}{d_{ij}} + 10 \right) \delta_{ij} + 100 \delta_i \right)$$
(12)

where  $d_{ij}$  is the distance between fibers *i* and *j*,  $\delta_{ij}$  is equal to one if the distance between these fibers is less than the minimum imposed and zero otherwise, and  $\delta_i$  is one if fiber *i* is too close to the edges of the RVE and zero otherwise. The minimization of this potential until its value is zero will ensure that the geometric rules are satisfied.

This process only depends on the overall volume fraction assigned to the RVE and the geometric conditions imposed to avoid defective meshing, see Section 5.2.

## 5.1.2. Reconstruction of Actual Microstructure

Once an initial mesh has been obtained, the next step is to make it evolve to a configuration that is statistically equivalent to the actual microstructure observed in a micrograph. It is at this point that the second-order intensity function K(r) is used, in a potential of the form:

$$E = \sum_{k} (K(r_{k}) - K_{0}(r_{k}))^{2}$$

$$+ \sum_{i} \sum_{j} \left( \left( \frac{100}{d_{ij}} + 10 \right) \delta_{ij} + 100\delta_{i} \right)$$
(13)

Here, the large weights assigned to the penalty terms enforcing the minimum distance between the fibers ensure that fulfilment of the geometric constraints dominates over the term depending on K.

This process leads to two different types of RVE's: <u>purely random</u> i.e. obtained purely through the hard-core process, and <u>reconstructed</u> i.e. obtained from actual micrographs. Two examples are shown in Figure 10.

#### 5.2. Minimum Distance between Fibers

The minimum distance between the fibers is a very important parameter in the process described earlier. It affects the jamming limit at high fiber volume fractions and it also has a significant influence on the second-order intensity function of the initial configuration. Figure 11 shows the linear stiffness obtained from random models in which this distance varies from 0.1  $\mu$ m to 1  $\mu$ m (1.43% to 14.3% of the fiber diameter). Five different cases were considered for each distance and it was found that the minimum stiffness is approximately the same for all distances, while the maximum stiffness increases by nearly 30% as the distance decreases. Unless stated otherwise, a minimum separation of 0.25  $\mu$ m (i.e. 3.57% of the fiber diameter) was used for all simulations presented in this paper, in order to facilitate meshing. The same distance was also introduced as a limit on the distance to the edges of the unit cell. This condition does not have a physical interpretation: it has been included only to facilitate meshing.



Figure 10: Fiber distribution in 50  $\mu$ m ×50  $\mu$ m RVE with  $V_f = 50\%$ : (a) original random arrangement and (b) subsequent reconstruction of microstructure observed in micrographs.



Figure 11: Linear stiffness for different vales of minimum distance between the fibers. Purely random RVE's with  $L_1=L_2=50~\mu m,~V_f=50\%$ 

## 5.3. Mesh Density

The mesh density is another key parameter of the model. Due to the incompressible nature of the matrix, coarse meshes introduce very high stress concentrations and can also result in the presence of a checker-board pattern in the stress field.

Standard approaches to study mesh related convergence issues analyze the evolution of the stress at a given point with respect to the mesh size in terms of the maximum local stress divided by the homogenized stress, and to consider the evolution of an overall parameter, such as the homogenized stiffness. Results of these studies are presented in Figure 12, which shows that meshes with stress concentrations of up to two orders of magnitude only increased the overall stiffness by 10%. It was found that extreme stress concentrations could be avoided by including at least two elements between each pair of fibers and hence most of the results presented in this paper were obtained from models with 50.000 - 70.000 nodes, unless stated otherwise.



Figure 12: Effects of mesh density: (a) linear stiffness and (b) stress concentrations. The RVE is purely random and fixed, with  $V_f = 50\%$ 

# 5.4. Size of RVE

The RVE needs to include enough particles to capture the behavior of the material (Monetto and Drugan, 2004) and this effect was investigated by considering four different values of  $L_1 = L_2$ ; For each of these values of  $L_1 = L_2$  five different fiber arrangements were analyzed and the resulting linear stiffnesses are compared in Figure 13. According to the figure, there is a wider range of values for the smallest RVE, while for the other RVE's there is broadly similar behavior for all fiber arrangements.



Figure 13: Linear stiffness for different sizes of the RVE. Purely random distribution,  $V_f=50\%.$ 

The transverse stiffnesses in the x and y directions were also compared, to assess the orthotropy of the RVE's. Differences on the order of 0.2% were obtained, even for the case  $L_1 = L_2 = 25 \ \mu\text{m}$ . A value of  $L_1 = L_2 = 50 \ \mu\text{m}$ was chosen, except for the case  $V_f = 22\%$ , in which case  $L_1 = L_2 = 75 \ \mu\text{m}$  was chosen. The size and number of fibers of the different RVE's considered in the present study are summarized in Table 2.

It is important to mention that these conclusions do not extend automatically to the case of large strains. Müller (1987) showed that the use of a RVE may lead to incorrect solutions in non-linear elasticity.

$V_f$ (%)	Side length $(\mu m)$	Number of fibers
22	75	32
30	50	19
40	50	26
50	25	8
50	50	32
50	75	73
50	100	130
50	50	32
55	50	35
60	50	38
65	50	42

Table 2: Number of fibers included in each finite element model, as a function of fiber volumefraction and size of RVE.

## 6. Simulation Results

Two different sets of simulations were obtained. The objective of the first set was to study the initial response of the material, hence the bonding between fibers and matrix was assumed to be perfect and only the tangent homogenized modulus of the material was calculated. These simulations explored the dependence on the volume fraction and the difference between purely random vs. reconstructed RVE's. As it will be shown, this approach fails to capture the behavior of the material once the strain increases.

In the second set of simulations cohesive elements were included, to capture the softening due to debonding between the fibers and the matrix. In this set of simulations the different parameters controlling the behavior of the cohesive elements were explored and the results were compared with the large strain response of the composite. The effect of the cohesive elements on the microscopic stress and strain fields was also addressed.

## 6.1. Perfect Bonding

The linear responses of several RVE's were calculated for both purely random and reconstructed fiber distributions, see Figure 14. Since only three different volume fractions could be produced experimentally, due to the limitations in controlling the amount of silicone in the material, the microstructure regeneration process could be carried out only for three values of  $V_f$ . For each value of  $V_f$  five different configurations were generated and analyzed.

It can be seen in Figure 14 that the homogenized linear response is significantly different between the two sets of RVE's and, particularly for larger  $V_f$ , the reconstructed RVE's can be as much as 50% stiffer.



Figure 14: Linear stiffness obtained from simulations on purely random and reconstructed RVE's. Five different RVE's of each type were considered for each value of  $V_f$ .

The results of the simulations on reconstructed RVE's are compared to the experimental results in Figure 15. The experimental results show an elastic modulus that is up to 25% higher than the corresponding simulations; this difference can be attributed to the presence of slightly misaligned fibers, which carry part of the load and so increase the overall stiffness of the material. Experimental observations of the material after failure also indicated the presence

of misaligned fibers, see Section 3.3. Another possible source of disagreement is the fact that the matrix model was based on tests done on pure silicone specimens. The mechanical response of the silicone in the composite might be different, due to changes occurring in the fabrication process.



Figure 15: Comparison of experimental results with linear stiffness obtained from simulations on reconstructed RVE's.

The simulation results allow the study of the details of the stress and strain fields within the material. The strain remains basically zero in the fibers but reaches very high concentrations in the matrix, particularly between fibers that are very close. For the case  $V_f = 65\%$  there are several regions where the strain is 10 to 50 times higher than the homogenized applied strain,  $\overline{\epsilon_y}$ , see Figure 16.

The corresponding distribution of  $\sigma_y$  is shown in Figure 17. This plot shows two load transmission paths with high stress concentrations in matrix regions closely surrounded by fibers, due to incompressibility.

Another example, in Figure 18, shows the differences in the major principal strain between a reconstructed and a purely random RVE, both with  $V_f = 50\%$ . In the case of the reconstructed RVE the fibers tend to cluster more than in the random microstructure and this produces a higher strain concentration between the fibers, leading to a higher overall stiffness.



Figure 16: Major principal strain distribution due to  $\overline{\epsilon_y} = 0.01$ ; reconstructed RVE with  $V_f = 65\%$ : (a) complete 50 × 50  $\mu m^2$  RVE and (b) close up view.



Figure 17: Distribution of stress component  $\sigma_y$  due to  $\overline{\epsilon_y} = 0.01$ ; reconstructed RVE with  $V_f = 65\%$ .

The simulations presented in this section are unable to capture the softening behavior, see Figure 19. In order to capture this effect cohesive elements were introduced between the fibers and the matrix, as discussed in the next section.

### 6.2. Debonding Allowed

Cohesive elements were introduced to model the debonding between the fibers and the matrix. In order to reduce the influence of the cohesive elements before damage takes place, their initial stiffness was set to 100 GPa. This guarantees almost perfect initial bonding between matrix and fibers.

The failure behavior of the cohesive elements available in ABAQUS is controlled by three main parameters. The first parameter controls damage initiation, which in the present study was defined by a quadratic combination of the traction components. Hence, it was assumed that damage will occur when:

$$\left(\frac{t_n}{t_{n0}}\right)^2 + \left(\frac{t_s}{t_{s0}}\right)^2 = 1 \tag{14}$$

where  $t_n$  and  $t_s$  are the normal and shear components of the nominal traction



Figure 18: Major principal strain distribution due to  $\overline{\epsilon_y} = 0.01$ ; reconstructed RVE with  $V_f = 50\%$ : (a) reconstructed RVE and (b) purely random RVE.



Figure 19: Plot of average longitudinal stress vs. average longitudinal strain: experiment and simulation (reconstructed RVE,  $V_f = 65\%$ ) with perfect bonding.

stress vector and  $t_{n0}$ ,  $t_{s0}$  are normalizing factors. In the present study the same factor was used for all tractions, i.e.  $t_{n0} = t_{s0} = t_0$ , however different values were used in a set of preliminary analyses to determine the relative importance of each term. It was found that  $t_n$  tends to control damage initiation, as the models that considered only  $t_n$  showed practically no difference in damage initiation from the models that took both tractions into account. However, those parts of the simulations in which damage growth occurred were numerically more stable when both tractions were included, which suggested that  $t_s$  becomes more important once damage has been initiated.

The second parameter is the separation  $\delta_f$ , for which two main effects were found. For separations ranging from 1  $\mu$ m to values much larger than the fiber diameter, such as 100  $\mu$ m, the response was fairly independent of the actual value chosen for  $\delta_f$ , see Figure 20. The reason is that the cohesive elements limit the stress in the material and also allow the formation of small gaps between the fibers and the matrix, which relax the constraint on deformation due to matrix incompressibility. For values of  $\delta_f$  much smaller than the fiber radius, the formation of a gap between fiber and matrix is enough to produce a large decay in the stress, which in some cases resulted in complete debonding.



Figure 20: Stress vs. strain under transverse loading for different values of  $\delta_0$ ,  $t_0 = 0.2$  MPa, reconstructed RVE,  $V_f = 65\%$ .

The model shows a better agreement with the experimental results for low values of  $\delta_f$  and the dependence of the solution on  $t_0$  for this case is shown in Figure 21. As expected, an increase in  $t_0$  delays the start of non-linear behavior and increases the total stress. The simulation with  $t_0 = 0.2$  MPa shows a decrease in stress for strains higher than 0.015, which indicates that the initiation of failure in the composite is not an instantaneous process, as already seen in Section 3.3.

The third parameter is the type of damage evolution, which in ABAQUS can be linear or exponential with respect to the separation. The results shown previously were all produced from models with linear damage growth, because using an exponential did not lead to significant changes.

Figure 22 shows a comparison between experiments and simulations, all for  $V_f = 65\%$  and for reconstructed RVE's. The numerical results provide a very good approximation of the test results. The response predicted by the simulations is not completely smooth due to the relatively small number of fibers included in the RVE: when a single fiber debonds, the force carried by



Figure 21: Stress vs. strain under transverse loading for different values of  $t_0$ ,  $\delta_0 = 0.1 \ \mu\text{m}$ , reconstructed RVE,  $V_f = 65\%$ .

the material drops, and then rises again as more strain is applied. This effect could be reduced by using larger RVE's.



Figure 22: Stress vs. strain under transverse loading for experiments and simulations using different reconstructed RVE's,  $V_f = 65\%$ .

If a loading-unloading cycle is applied, the model shows permanent damage due to the degradation of the cohesive elements, see Figure 23. There are several differences from the behavior observed in the experiments, mainly due to the fact that as long as no damage is being generated the cohesive elements behave linearly and therefore neither hysteresis nor permanent deformation are predicted by the simulation. Also, the unloading-reloading path is basically a straight line until new damage occurs. Despite these limitations the model is still able to provide a good prediction of the experimental behavior.



Figure 23: Stress vs. strain under cyclic transverse loading for experiment and simulation using reconstructed RVE,  $V_f = 65\%$ .

The simulations can be used again to look at the microscopic stress and strain fields. Figures 24-25 show the maximum principal strains and the stress components in the loading direction when cohesive elements are introduced in the simulations originally presented in Figures 16-17, for the case of perfect bonding. The same scale has been kept for ease of comparison: the strains have approximately the same values, although the distribution is different, and the stresses are much lower, as expected, due to the inclusion of the cohesive elements.



Figure 24: Distribution of maximum principal strain due to  $\overline{\epsilon_y} = 0.01$ . Reconstructed RVE,  $V_f = 65\%$ , cohesive elements introduced between fibers and matrix.



Figure 25: Distribution of stress  $\sigma_y$  due to  $\overline{\epsilon_y} = 0.01$ . Reconstructed RVE,  $V_f = 65\%$ , cohesive elements introduced between fibers and matrix. The scale is the same as in Figure 17.

The stresses are not only lower, but their distribution is significantly different from the case of perfect bonding. If the scale is changed according to the new stress levels, Figure 26, it can be seen that the highest stress no longer occurs between the fibers that are closest, because the cohesive elements degrade more in regions where the fibers are closer than where they are more spread out.



Figure 26: Distribution of stress  $\sigma_y$  due to  $\overline{\epsilon_y} = 0.01$ . Reconstructed RVE,  $V_f = 65\%$ , cohesive elements introduced between fibers and matrix.

As it was noted in Section 3.1, the values of the principal strain observed in the simulations are well beyond the failure point observed in uniaxial testing. Although these results cannot be used to assess quantitatively the integrity of the material, they suggest that debonding is sufficient to capture the observed softening.

# 7. Comparison with Analytical Bounds

Several approximations for the transverse stiffness of a fiber reinforced material have been proposed (Jones, 1999). The rule of mixtures gives a transverse stiffness of

$$E_2 = \frac{E_{2f}E'_m}{(1 - V_f)E_{2f} + V_fE'_m}$$
(15)

where  $E_{2f}$  is the transverse stiffness of the fibers and  $E'_m$  is equal to

$$E'_m = \frac{E_m}{1 - \nu_m} \tag{16}$$

where  $E_m$  and  $\nu_m$  are the properties of the matrix, which is assumed to be isotropic. This expression provides a rough lower bound.

A semi-empirical approximation can be obtained through the Halpin-Tsai equations (Halpin and Kardos, 1976), which give a transverse stiffness of

$$E_2 = E_m \frac{1 + \eta \xi V_f}{1 - \eta V_f} \tag{17}$$

where the parameter  $\eta$  is defined as

$$\eta = \frac{E_{2f} - E_m}{E_{2f} + \xi E_m} \tag{18}$$

and  $\xi$  is a parameter to be fitted experimentally, and whose value usually ranges from 1 to 2.

It is interesting to study the validity of these approximations in the case of composites with a hyperelastic matrix. Figure 27 shows a comparison between the numerical results from Section 6, the rule of mixtures, and the Halpin-Tsai prediction. The value used for the matrix stiffness was the initial tangent modulus of the silicone under uniaxial tension, obtained by differentiation of Equation 8 that gives  $E_m = 6C_1 + 2C_2 = 0.9048$  MPa, and the Poisson's ratio was that of incompressible materials,  $\nu_m = 0.5$ . The results were produced with purely random RVE's, i.e. no information from the micrographs was used. The simulations were repeated using the same fiber arrangement and finite element mesh, but giving the matrix the properties of a typical epoxy, with  $E_m = 4.5$  GPa and  $\nu_m = 0.2$ . In both cases the response was normalized by the elastic modulus of the matrix.



Figure 27: Comparisons of transverse stiffness of two types of fiber-reinforced composites with (a) silicone matrix and (b) epoxy matrix. The simulations assumed random RVE's. The crosses show the values of the five different configurations for each value of  $V_f$ .

The results show two main differences. The first difference relates to the magnitude of the increase in normalized stiffness and its dependence on the volume fraction. The normalized stiffness of the composite with the silicone matrix increases rapidly with  $V_f$  and reaches a value of over 20. This behavior is also different from the Halpin-Tsai predictions; attempts to increase the value of  $\xi$  were still not able to reproduce the dependence on volume fraction observed in the simulations. The second difference is a much greater spread in stiffness values obtained from the simulations in the case of composites with silicone matrix than in the case of composites with epoxy matrix. For example, in the case of  $V_f = 60\%$  the extreme values differ by up to  $\pm 20\%$  from the mean value.

There are two possible explanations for this difference of behavior: the extreme disparity of stiffness between fibers and silicone matrix, and the incompressible nature of silicone. In order to study both options, two new sets of simulations were produced using the same RVE's but different matrix properties, see Figure 28. The first set of simulations modeled the matrix as a linear material with  $\nu_m = 0.2$  and modulus equal to the tangent modulus of the silicone,  $E_m = 0.9048$  MPa. In this case the material behavior could be fitted with the Halpin-Tsai equation using  $\xi = 1.5$ , which lies in the usual range of this parameter. In the second case the matrix retained the elastic modulus of epoxy,  $E_m = 4.5$  GPa, but incompressibility was also included. In this case the stiffness gain was much higher than in the case of compressible epoxy, and the Halpin-Tsai equation with  $\xi = 2$  gave a poor prediction. By using a value of  $\xi = 25$  the prediction was much improved and the dependence on  $V_f$  was captured. However, this value of  $\xi$  is well outside the usual range.



Figure 28: Comparison of transverse stiffness from simulations and analytical predictions. Matrix properties: (a)  $E_m = 0.9048$  MPa,  $\nu_m = 0.35$  and (b)  $E_m = 4.5$  GPa,  $\nu_m = 0.5$ . The crosses show the value of each of the five different configurations considered for each  $V_f$ .

Therefore, it can be concluded that incompressibility alone is enough to produce some deviation from the behavior of standard, epoxy based composites but the effect is much more significant when combined with a matrix that is several orders of magnitude softer than the fibers. In particular, a large spread in stiffness values will be seen only when both effects are present.

Finally, the stiffness predictions from the simulations were also compared to the Hashin-Rosen bounds (Hashin and Rosen, 1964), which include the effects of the bulk modulus of the matrix. The bounds were calculated using the linear stiffness of the silicone and different values for its Poisson's ratio ( $\nu_m =$ 0.4, 0.49, 0.499 and 0.4999). The results have been plotted in Figure 29, which shows that the upper bounds are highly dependent on the value of  $\nu_m$ , although they tend to converge for values very close to the incompressibility limit; the lower bound, on the other hand, show no dependency. The upper bounds for  $\nu_m > 0.49$  bound all numerical results, see Figure 29, however the disparity between higher and lower bounds is too high for these estimates to be useful in design applications.



Figure 29: Comparison of transverse stiffness from simulations and Hashin-Rosen analytical bounds, for  $E_m = 0.9048$  MPa and different values of the Poisson's ratio.

## 8. Conclusions

A composite material consisting of unidirectional carbon fibers in a silicone matrix was fabricated and tested under tension in a direction transverse to the fibers. Its behavior was found to be nonlinear and a marked strain softening was observed. Failure occurred due to tearing of the matrix, a process which was stabilized by slight misalignments of the fibers.

A 2D finite element model was created in order to study the micro-mechanics of this material, using a representative volume element with periodic boundary conditions. Two different fiber arrangements were considered: a random hard-core process and a reconstruction method using the second-order intensity function obtained from actual micrographs of the material.

In a first set of simulations perfect bonding between the fibers and the matrix was assumed and the modulus of the matrix was taken to be constant. This approach produced a linear response, and the analyses in which the fiber distribution had been based on direct measurements from micrographs showed a higher stiffness for low fiber volume fractions, due to fiber clustering. No significant difference in initial stiffness could be observed for fiber volume fractions of 55% or higher. These numerical predictions provided an adequate match to the measured initial stiffness but did not (attempt to) capture strain softening, which resulted in microscopic strains up to 50 times higher than the macroscopic average strain being obtained.

In a second set of simulations debonding between the fibers and the matrix was allowed to occur by introducing cohesive elements in the finite element model. This modification allowed the simulations to capture strain softening and thus replicate the observed nonlinear behavior as well as the damage under cyclic loading. Although these simulations were still not able to fully capture the observed hysteresis and permanent deformation, since the unloading-reloading behavior of the cohesive elements is elastic (a more refined model of debonding and crack propagation could be adopted for more realistic predictions), this approach was able to reduce the microscopic strains to values well below the failure stretch of the silicone matrix observed under uniaxial conditions.

It is concluded that the transverse loading test provides a very useful measure of the damage taking place in a uniaxial fiber composite with a soft matrix. By comparing experimental results and simulations that assume perfect bonding, parametric studies of different types of silicones, fiber sizing, etc. could be carried out in the future, to find the best combination of materials and processing techniques.

An additional result of this study is the fact that uniaxial fiber composites with a soft matrix show a much higher transverse stiffness than composites with traditional epoxy, once the results are normalized by the elastic modulus of the matrix. Numerical simulations have shown that the main reason is the incompressibility of the matrix; the high difference in stiffness between fibers and matrix is less important. Hence it is noted that traditional analytical tools like the Halpin-Tsai equations should not be used with this new type of composites.

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