YOUNG’S MODULUS PREDICTION OF LONG FIBER REINFORCED THERMOPLASTICS

F. Garesci\textsuperscript{a}, S. Fliegener\textsuperscript{b}

Corresponding author: F. Garesci: fgaresci@unime.it

\textit{a) DIECII – Department of Electronic Engineering, Industrial Chemistry and Engineering, con.da di Dio – University of Messina, Italy}

\textit{b) Fraunhofer Institute for Mechanics of Materials IWM, Freiburg, Germany}

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\textbf{Abstract}

The aim of this paper is to provide analytical models able to predict the elastic properties of long fiber reinforced thermoplastics (LFT) in dependence of microstructural parameters such as the fiber volume content ($v_f$), the fiber orientation distribution (FOD) and the fiber length distribution (FLD). The analytical predictions are compared to the experimental stiffness values from tensile tests on the composite material showing a good agreement. The FLD in terms of the probability density distribution as function of the fiber aspect ratio has been computed by an automated fiber separation and image analysis tool. On the other hand, the FOD in terms of the probability density distribution as function of fiber in-plane orientation was identified by using an image correlation procedure based on computer tomography scans of characteristic LFT specimens. Our analysis shows a good performance to predict the Young’s Modulus of LFT due to incorporation of the FOD as well as the FLD into the calculations.

\textbf{Introduction}

In the last years, the demand of high performing and economically producible materials for automotive applications is growing. LFT are a promising solution meeting the criteria cited
above, in particular they provide a good specific stiffness and strength if the part design is
adapted to the respective load cases. The efficient value of the material properties such as
elastic stiffness and strength varies in a broad range up to 250% [18], [19]. Those values
depend on the process-driven microstructural properties such as the fiber volume content (\(v_f\)),
the fiber orientation distribution (FOD) and the fiber length distribution (FLD) and they must
be considered in order to fully exploit the lightweight potential of LFT. In this work, the
elastic stiffness of LFT is derived from the characteristic values of the microstructure by
means of analytical models. There are several procedures in literature for predicting the
properties of composite materials in dependence of their microstructure. Tucker et al. [1]
provide a good overview on the micromechanical models used to calculate the stiffness of
short fiber composites. This traditional approach combines several methods such as the
Eshelby equivalent inclusion, the Mori-Tanaka models and the Halpin-Tsai equations [2] to
predict the elastic properties of short fiber reinforced plastics in dependence of both fiber
orientation and volume content. Other authors [3], [4], [5] use a unidirectional short fiber
composite and improve the micromechanical models by taking into account the spatial
distribution of the reinforced fibers. In this way, it is also possible to predict the local thermo-
mechanical properties excellently. Other studies focus on the closure approximations [6],[7],
[8] for evaluating the thermoelastic properties of short fiber composite to optimize the
injection moulding process. A recent study [9] is based on the elastic prediction of the correct
experimental fiber length distribution (FLD) and the correct fiber orientation distribution
(FOD). The fiber dimension could be a relevant aspect for understating the elastic properties
of composites. Simple models are based on the prediction of spherical particles [12] or short
fiber composite, but recently studies focused on not aligned long fiber composite [13] and
classic theory applied to new materials like carbon nanotubes [14] and nano composite [15]
are very promising in term of understating the elastic properties. Other authors focused their
methodology in the effect of the interfacial bonding conditions only to evaluate the effective properties of the composite [10], [11]; while others used finite element method in order to investigate how two of the most widespread models (i.e. Tandon-Weng and Halpin-Tsai) are suitable to predict either the stiffness of short fiber composite [21] and a composite by applying the orientation averaging scheme (several hundred multi-fiber with different orientation states [22]). In Ref. 12 the two previous approaches are modified in two relevant steps. Firstly, by incorporating an experimental FLD as well as a fiber orientation distribution (FOD) and secondly by evaluating the Mori-Tanaka model to obtain the stiffness matrix of the aligned fiber composite containing the previous FLD.

In this article, the elastic stiffness of a commercial long fiber reinforced thermoplastic has been investigated by using a hybrid procedure based on a theoretical approach describing the transversely isotropic equivalent composite. The model uses the experimental data of the fiber orientation and length distributions. To validate the procedure the predicted stiffness values are compared with experimental values from tensile tests. The scenario of the experimental framework presented in this manuscript is different from the works that studied short fiber composite (aspect ratio near the value of 20); in fact, in our experimental data the smallest value measured of aspect ratio is 27. In addition, if we considered the long fiber studies, most of them don’t use directly experimental FLD but the corrected FLD data or the Monte Carlo method. The advantage of our procedure is due to the direct use of the experimental data and thus to predict the stiffness properties of LFT rather quickly. The working scheme is given by three main steps:

1. **Experimental methods and materials:**
   
   1.1. Tensile tests to determine the elastic stiffness of the LFT material;
   
   1.2. Microstructure analysis by means of computer tomography to analyse the Fiber Orientation Distribution (FOD)
1.3. Automated fiber separation and analysis procedure to investigate the Fiber Length Distribution (FLD)

2. Analytical Procedure:
   
   2.1. Halpin-Tsai theory to evaluate the properties of the transversely isotropic composite;
   
   2.2. Fiber Orientation Distribution function to determine the tensor rank2 $a_{ij}$ and the tensor rank4 $a_{ijkl}$ after Advani, Tucker [16];

   2.3. Mean Tensor Averaging procedure to predict the Elastic Properties of LFT.

3. Fit functions:

   3.1. Curve Fitting Procedure for Fiber Orientation Distribution (FOD);

   3.2. Curve Fitting Procedure for Fiber Length Distribution (FLD).

1. EXPERIMENTAL METHODS AND MATERIALS

The LFT material was produced by a so-called direct LFT production route, where the continuous fiber roving are introduced directly into a double screw extruder and are broken into fiber fragments by the overlapping edges of the contra-rotating screws. The resulting material is characterized through a broad FLD reaching from a large amount of fiber fragments well below 1 mm until very long fibers up to 50 mm. The matrix consists of polypropylene resin (DOW® C711-70RNA) with some stabilizers and coupling agents and is merged with 30 mass-% glass fibers (TufRov® 4575) to form the LFT composite. To simulate different material states in terms of a locally varying microstructure, 3 mm thick plates with outer dimensions of 400 x 400 mm were produced by compression molding. The LFT-D strand as it comes out of the extruder was placed asymmetrically in the mold to separate the so-called press region near the strand inlay position from the flow-region, where a lateral flow is responsible for a higher degree of fiber orientation compared to the press-region.

1.1. Tensile tests to determine the elastic stiffness of the LFT material
The tensile stiffness of specimens with different orientation relative to the fiber mean orientation or flow direction, but similar fiber volume fraction and fiber length distribution was determined with a Hegewald&Peschke “Inspekt 100” testing machine. The specimen geometry was chosen according to DIN EN ISO 3167. The tests were performed at a strain rate of approximately 0.00022 [mm/s] to a maximum stress of 10 MPa in order avoid any damage. Three loading-unloading cycles per specimen were performed. The stiffness is taken as the mean value of the three loading cycles. At least three specimens per orientation were tested. The stiffness values for $E_{11}$ and $E_{22}$ are 7.83 and 3.25 GPa and 6.49 and 3.77 GPa for flow and press region LFT (30 mass-% / 13 volume-% fiber fraction) respectively.

1.2. Microstructure analysis by means of computer tomography

Microstructure analysis has been performed on specimens from different regions of a 3 mm thick LFT-plate. Because of the thin cross section, the fibers are aligned in planes parallel to the specimen surface and the out-of-plane component of the fiber orientation is negligibly small. The specimens for computer tomographic analysis have been prepared from similar regions as the specimens for mechanical testing were taken from. The tomography scans have been performed on a Phoenix “nanome|x 180NF” with a voxel size of 5 - 8 µm, so the complete plate thickness could be analyzed with one scan. The data was analyzed by the plug in “Directionality” for ImageJ [20], after converting the 3D voxel data to a stack of 2Dimages parallel to the surface. The 2D images parallel to the surface 2D orientation histograms have been extracted for each of the stack. The resulting orientation distribution for the whole stack has been generated by summing up the single histograms and re-normalizing.

Flow region specimens (figure 1.a) were taken from a region of approx. 100 mm distance from the LFT strand inlay position, so a relatively high amount of fiber alignment results from the lateral flow field inside the mold. A low degree of fiber waviness can be observed in a characteristic CT scan of a flow region specimen. However, the scan shows only a section of
approx. 2.8 mm edge length due to resolution limits of the scanning device. An exemplary CT scan of a press region specimen (figure 1.b) exhibits remarkable deviation from the flow region structure which can be explained by a different flow field during specimen fabrication and thus a significantly lower degree of fiber alignment.

1.3. Automated fiber separation and analysis procedure

The FLD was determined with a characteristic LFT specimen of a size of approx 70x40x3 mm³. The specimen was first incinerated and then dispersed in a dilute solvent to separate the fibers. Finally, the fibers were analyzed by the image analysis software “FASEP®”. The procedure was carried out by the company “xyz high precision” [17].

2. ANALYTICAL PROCEDURE

2.1. Halpin-Tsai theory to evaluate the properties of the transversely isotropic composite

The Halpin Tsai equations are used to predict the mechanical properties of continuous long fiber composites [2]:

\[
\frac{P_c}{P_m} = \frac{1 + \zeta \eta V_f}{1 - \eta V_f} \quad (1)
\]

where \(P_c\) and \(P_m\) are the composite properties (\(E_{11}, E_{22}, G_{12}\)) and the matrix properties respectively, \(\zeta\) is a measure of reinforcement geometry and loading conditions and \(\eta\) is a fiber parameter computed as:

\[
\eta = \frac{P_f / P_m - 1}{P_f / P_m + \zeta} \quad (2)
\]

For continuous, cylindrical fiber reinforced composites the geometry factor is given as:

\[
\begin{align*}
\zeta_{E_{11}} &= 2 \cdot AR + 40V_f^{10} \\
\zeta_{E_{22}} &= 2 + 40V_f^{10} \\
\zeta_{G_{12}} &= 1 + 40V_f^{10}
\end{align*}
\]
If the fibers are aligned along 1-direction and the behaviours in the 2-direction and 3-direction are identical, the composite can be treated as transversely isotropic. Under this hypothesis the following identities hold: \( E_{22} = E_{33}, \nu_{12} = \nu_{13}, G_{12} = G_{13} \) and the following relationship is verified:

\[
G_{23} = \frac{E_{22}}{2(1 + \nu_{23})}
\] (4)

In this scenario, the independent constants for the transversely isotropic material are only five (i.e. \( E_{11}, E_{22}, G_{12}, \nu_{12}, \nu_{23} \)) and the Compliance Matrix \([S]\) can be expressed by:

\[
[S] = \begin{bmatrix}
\frac{1}{E_{11}} & \frac{\nu_{21}}{E_{22}} & -\frac{\nu_{31}}{E_{33}} & 0 & 0 & 0 \\
-\frac{\nu_{12}}{E_{11}} & \frac{1}{E_{22}} & -\frac{\nu_{32}}{E_{33}} & 0 & 0 & 0 \\
-\frac{\nu_{13}}{E_{11}} & -\frac{\nu_{23}}{E_{22}} & \frac{1}{E_{33}} & 0 & 0 & 0 \\
0 & 0 & 0 & \frac{1}{G_{23}} & 0 & 0 \\
0 & 0 & 0 & 0 & \frac{1}{G_{13}} & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{1}{G_{12}}
\end{bmatrix}
\] (5)

The Stiffness Matrix \([C]\) is the inverse of \([S]\) matrix.

\[
[C] = \begin{bmatrix}
C_{11} & C_{12} & C_{13} & 0 & 0 & 0 \\
C_{21} & C_{22} & C_{23} & 0 & 0 & 0 \\
C_{31} & C_{32} & C_{33} & 0 & 0 & 0 \\
0 & 0 & 0 & C_{44} & 0 & 0 \\
0 & 0 & 0 & 0 & C_{55} & 0 \\
0 & 0 & 0 & 0 & 0 & C_{66}
\end{bmatrix}
\] (6)

The stiffness matrix for the transversely isotropic material is completely defined if every parameter of matrix and fiber is known and all terms inside the matrix simply depend on the aspect ratio and the volume fraction values.

2.2 Fiber Orientation Distribution function to determine the Advani-Tucker tensors

In the earliest work, to study the elastic properties of a composite taking into account the fiber orientation, each fiber was modelled as a rigid cylinder with fixed length and diameter [16]. If the fiber concentration is spatially uniform, a spherical coordinate system \(\Psi(\theta, \varphi)\) that
represents the probability of the fiber orientation inside the unit sphere can be introduced. 

$\Psi$ is normalized and symmetric function. In addition, a vector $\mathbf{p}$ describing the position of the fiber in the spherical coordinate system can be defined as follows:

$$
\int \int \int_{\phi=0, \theta=0}^{2\pi, \pi} \sin \theta d\theta d\phi.
$$

The tensor Rank 2 and the tensor Rank 4 are:

$$
a_{ij} = \int p_i p_j \Psi(p) dp
$$

$$
a_{ijkl} = \int p_i p_j p_k p_l \Psi(p) dp
$$

being $i,j,k,l=1,2,3$. For a bi-dimensional material, where all fibers are located in the 1-2 plane (as it is the case for the investigated LFT), the orientation state is planar and the distribution function $\Psi_{\phi}(\phi)$ depends only on $\phi$ and the tensor Rank 2 and the tensor Rank 4 are:

$$
a_{ij} = \int_0^{2\pi} p_i p_j \Psi_{\phi}(p) dp
$$

$$
a_{ijkl} = \int_0^{2\pi} p_i p_j p_k p_l \Psi_{\phi}(p) dp
$$

Starting from the experimental data, it is not possible to describe and absolutely quantify the orientation of each fiber inside the composite, in this sense a probability distribution function to describe the spatial or plan probability distribution of the fibers is used.

2.3 Mean Tensor averaging procedure to predict the Elastic Properties of LFT

Basically, the properties of the composite are related to the probability distribution function $\Psi(p)$ and to the properties of the transversely isotropic tensor $T(p)$ by:

$$
\langle T \rangle = \int T(p) \Psi(p) dp
$$

Where $\langle T \rangle$ denotes the orientation average property. The tensor $T(p_{i,j,k,l})$ is given by:
where $\delta_{ij}$ denotes the Kronecker delta function and the scalar $B_i$ parameters are depending on

the stiffness matrix elements (eq.6) as shown:

\[
B_1 = C_1 + C_{22} - 2C_{12} - 4C_{16} \\
B_2 = C_{12} - C_{23} \\
B_3 = C_{16} + \frac{1}{2}(C_{23} - C_{22}) \\
B_4 = C_{23} \\
B_5 = \frac{1}{2}(C_{23} - C_{22})
\]

By applying the equations 12 and 13, it is possible to evaluate the elastic properties of the LFT composite weighted by the orientation distribution function. It is also important to remark that the fibers have a uniform length and constant aspect ratio (fiber length/fiber diameter) because the volume fraction $v_f$ is constant.

**Fit functions:**

### 3.1 Curve Fitting Procedure for FOD (Fiber Orientation Distribution)

From microstructure analysis (CT-scans) we obtain the Fiber Orientation Distribution (FOD) in terms of probability density distributions as function of the in-plane fiber orientation for Flow and Press Regions (figure 2). As described in section 2.2, the FOD function $\Psi_\theta$ is to be symmetric and normalized. Despite different functions can be applied, not all of them satisfy the two criteria. For example, the Gaussian Function (blue curve in figure 2) is normalized and symmetric but doesn’t reproduce the experimental data; on the other hand the Gauss function (Red curve) is a symmetric function suitable to fit the experimental data but it is not a normalized function. With this in mind, we selected the trigonometric function $y = \alpha + \beta \cos(x + \gamma)$ (orange curve in figure 2). The function is symmetric and normalized and it reproduces with a good agreement the experimental data, in fact it doesn’t
underestimate the presence of fibers far from the 1-direction. The shape of the trigonometric function well replicates the shape of the experimental data and shows the lowest value for the quadratic error function (figure 2). The $a_{ij}$ tensors are finally obtained by applying eq.9 and after choosing the trigonometric functions to model the Flow and the Press region. Their values are:

\[
\begin{bmatrix}
0.718 & -0.066 & 0 \\
-0.066 & 0.282 & 0 \\
0 & 0 & 0
\end{bmatrix}
\text{ and }
\begin{bmatrix}
0.594 & -0.005 & 0 \\
-0.005 & 0.406 & 0 \\
0 & 0 & 0
\end{bmatrix}
\]

respectively. The FOD function was used in the computation. Therefore, we evaluate the Young’s Modulus value of the composite starting from the properties of transversely isotropic material and by using the tensor procedure and the recovered FOD function. We focused on the flow region; this region is the part of the specimen where the highest degree of fiber alignment can be observed. Based on the information from section 1.3, the Halpin-Tsai theory which was developed for ideal straight fibers can most likely be applied for flow region LFT while it must be rated as critical to model press region specimens.

3.2 Curve Fitting Procedure for FLD (Fibers Length Distribution)

From specimen incineration and image analysis we can determine the Fiber Length Distribution (FLD) in terms of probability density distribution versus aspect ratio. The experimental fiber distribution as function of the aspect ratio is characterized by two main regions, see purple line in figure 3, in the first region achieved for values smaller than 500 the probability decreases strongly from 15.77% to 0.93%, in the second region scattered data are observed. For this reason, for this experimental data the distribution functions (logarithmic) used in [11] are not suitable. Our investigations indicate as more appropriate curves the ones shown in figure 3: (i) the exponential function (the red curve: $y = e^{ax+bx}$) and (ii) the hyperbolic function (the orange curve $y = a/x$). The experimental range of the aspect ratio is between 27 and 3340. We can observe that: both red and orange curves in figure 3 are in good

10
agreement to the experimental data for low values of aspect ratio (smaller than 200); the percentage amount of the fibers with aspect ratio less than 200 is 37.24% and 50.18% respectively. In particular, the 32.8% of the fibers have an aspect ratio values smaller than 120 and the 15.77% of the fibers have aspect ratio equal to 27. For higher values of aspect ratio scattered data are observed. In this condition, two main mistakes could be occur, (i) to underestimate the $E_{11}$ value if we focus on the lower aspect ratio or (ii) to exclude a high amount of fibers if we focus on the higher aspect ratio values. Furthermore the orange curve follows well the experimental data for high values of aspect ratio. All curves are fitted with data inside the experimental range of AR. Outside this range, it doesn’t make sense to recover the FLD, because values lower than 27 correspond to fiber fragments with low load-carrying ability and values higher than 2500 are less relevant for our investigation as the elastic stiffness has almost reached its saturation level for AR above 1000.

**Results obtained by using the recovered functions for FOD and FLD and by using the Mean Tensor Averaging Procedure**

The last part of this work is related to the prediction of the properties of LFT through the combination of the Halpin Tsai theory, the orientation distribution function, the mean tensor averaging procedure and the length distribution function.

- The **first** step is to evaluate the elastic properties of the transversely isotropic material from the Halpin-Tsai theory. As known, for a fixed fiber volume fraction, the elastic stiffness of the transversely isotropic composite in the 1 direction ($E_{11}$) is expressed as a function of the aspect ratio and its value increases highly for low values of aspect ratio and up aspect ratio equal to 200 reaches a constant value near. This behaviour is independent on the value of fiber volume fraction. To achieve the curves we used for fibers and matrix the data shown in the appendix (table a) under application of equations from eq.1 to eq.4.
The second step is to apply the mean tensor averaging procedure to take into account the orientation distribution function. The Elastic Modulus of the composite is evaluated by using the eq.11 and the FOD identified from experimental data. As shown in figure 4 for volume fraction values between 13% and 18%, \( E_{11} \) depends on the aspect ratio up to 200, for larger values the modulus does not change appreciably. In the figure 4, the purple points are the constant experimental value (see section 1.1). From this point of view, it seems that the composite with similar mechanical properties has a volume fraction between 17% and 18%, which is well above the nominal value of 13% of the investigated material. However, to have a full overview about the elastic modulus we have to take into account the amount of each fiber inside the composite that means that we have to weigh the Elastic modulus values on the FLD.

The third step is to use the recovered fit function to achieve the experimental distribution lengths data, for weighting the \( E_{11} \) obtained from the previous Tensor procedure. A recovered fit function for each \( E_{11} \) values is found. As an example, figure 5 shows for \( v_f=13\% \), the \( E_{11} \) value for the following cases: (i) a perfect fiber aligned (in 1-direction) composite (thick blue line) computed by using Halpin-Tsai equations; (ii) the Tensor procedure explained in the second step (dashed blue line) computed by using the tensor procedure; and (iii) the recovered distribution function of the previous function (continuous blue line).

Applying the procedure we obtain the \( E_{11} \) value for each volume fraction value as indicated in table b. By using this procedure it is possible to evaluate the \( E_{11} \) value for a fixed volume fraction with the experimental distribution length function. For \( v_f=13.29\% \) the \( E_{11}=8.70 \) GPa.

Conclusion

A hybrid methodology to predict the Young’s modulus of a Long Fiber Thermoplastic composite was developed in detail in this paper. Experimental data for fiber length
distribution and for orientation distribution was used to obtain the fitting functions and to 
combine the fitting functions with the Halpin Tsai equations. Our results indicate that our 
approach can be used with success to predict the stiffness value for a fixed volume fraction, 
and to estimate the volume fraction using a fixed stiffness value. We conclude that: a) if we 
don’t take into account the experimental length distribution we overrate the volume fraction; 
b) to improve the good agreement with the experimental results, we need more accurate 
experimental data for FLD; c) if our threshold is the experimental value of stiffness (7.83 GPa) 
the obtained volume fraction assumes a lower value than the expected one (11.5%).

To reach the point b), we need to take into account a larger amount of fibers, in our case it is 
around 50% between the AR range (27<AR<1000); for higher AR values, experimental 
measurements show scattered data so we need more experimental investigations for 
improving this point. This could also explain the point c): the detected volume fraction value 
is lower than the experimental one because if we used only the 50% of the total amount of the 
real fiber present in the specimen we underestimate the presence of the fiber inside the 
composite. We focus on predicting the young’s modulus of LFT by using, as much as 
possible, the experimental data in terms of DOF and FLD. If we compared the value obtained 
with the experimental one, we can say that the proposed methodology can be considered 
useful for the purposes of the prediction of Young’s modulus.

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Figures and Tables

<table>
<thead>
<tr>
<th>E_f</th>
<th>72.00 [GPa]</th>
<th>Fiber Young's Modulus</th>
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<tbody>
<tr>
<td>ν_f</td>
<td>0.22</td>
<td>Fiber Poisson's ratio</td>
</tr>
<tr>
<td>d_f</td>
<td>0.017</td>
<td>Fiber diameter</td>
</tr>
<tr>
<td>E_m</td>
<td>1.40       [GPa]</td>
<td>Matrix Young's Modulus</td>
</tr>
<tr>
<td>ν_m</td>
<td>0.35</td>
<td>Matrix Poisson's ratio</td>
</tr>
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</table>

Table a – Fiber (f) and matrix (m) properties

<table>
<thead>
<tr>
<th>Volume Fraction</th>
<th>E_{11} [GPa]</th>
</tr>
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<tbody>
<tr>
<td>12%</td>
<td>8.06</td>
</tr>
<tr>
<td>13%</td>
<td>8.54</td>
</tr>
<tr>
<td>14%</td>
<td>9.05</td>
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<tr>
<td>15%</td>
<td>9.56</td>
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<td>10.05</td>
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<tr>
<td>17%</td>
<td>10.58</td>
</tr>
<tr>
<td>18%</td>
<td>11.09</td>
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</table>

Table b - Analytically predicted E_{11} values for flow region LFT at different volume fraction values
Figure 1: experimental Ct scan for Flow (a) and Press (b) region

Figure 2 – Flow region: Comparison and error variations (in the insertion) between experimental data and the FOD fitting functions and error variations
Figure 3 – Comparison between experimental data (dark purple) and several fit curves.

Figure 4 - $E_{11}$ as function of the aspect ratio computed by taking into account the tensor procedure for DOF at different volume fraction and experimental values (purple points)
Figure 5 – $E_{11}$ as function of the aspect ratio achieved for a transversely isotropic material (thick blue line), by using tensor procedure (dashed line) and by using the recovered fit function (continuous line) for a fixed volume fraction value (13%).