MULTISCALE MODELLING OF FIBRE REINFORCED COMPOSITES

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ABSTRACT

A multiscale modelling technique has been developed to predict the properties of a carbon fibre reinforced composite system. The full yield behaviour of an epoxy resin matrix consisting of amine-cured tetruglycidyl 4,4'-diaminodiphenylmethane has been predicted using a mean field modelling technique (Group Interaction Modelling). These curves are used in a 3D finite element model to obtain strain concentration factors of fibres adjacent to a fibre break in a unidirectional composite. A Monte Carlo simulation is performed to predict the tensile failure strain of a chain of single composite layers containing the yielding matrix.

1. INTRODUCTION

The use of multifunctional epoxy resins as the matrix in fibre reinforced composite materials is widespread. The cured resins display excellent strength and toughness, superior environmental resistance along with good thermal and electrical properties [1]. The resin chosen for this work is tetruglycidyl 4,4'-diaminodiphenylmethane (TGDDM) cured with the amine diaminodiphenylsulphone (DDS), both of which are tetrafunctional (see Figure 1). The curing mechanism is well characterised [2], but exact structural details of the 3D network are unknown. The use of predictive modelling to estimate engineering properties of composites is becoming more popular [3]. A multiscale approach to the modelling of composite systems is presented in this work. Firstly, the stress-strain curves to yield of a TGDDM/DDS matrix are predicted using Group Interaction Modelling (GIM). This technique has proven successful in predicting the properties of linear polymers [4] and is now beginning to be applied to crosslinking polymers too [5].

![TGDDM and DDS](image)

Figure 1: Chemical Structures of TGDDM and DDS.

Most of the statistical models for composite failure strength are based on the linkage of several single fibre elements of a given ineffective length and incorporated into a Monte-Carlo simulation. However, overlooking the nonlinear behaviour of the matrix in the analytical models [6,7] has led to unreliable results and limited the capability of the models. It is believed that interfacial shear yielding of a yielding matrix decreases the probability that a crack will propagate through the matrix as well as reducing the strain (or stress) concentration factor (SCF) in the adjacent fibres [8,9]. The SCF in adjacent fibres of a fibre break is the key parameter used in statistical models to predict the failure strength of a UD. Several models have been proposed for the prediction of the strength of UD composites, two of which are key. Firstly, Rosen’s model [6] which...
considers the equal load sharing rule between fibres and predicts a significantly higher failure strength than that of real composites. Secondly, Zweben’s model [10] which used the Hedgepeth and Van Dyke [11]-predicted SCF values and reported a lower strength than that of a real composite. It is suggested [12] these models are able to predict the upper and the lower bounds of the composite failure strength. Curtis [13] established an uncomplicated model which predicts the progressive growth of fibre breaks and the failure strain of UD composites under tension. However, this study did not take into account the effect of a yielding matrix in the model. This study attempts to establish a link between the GIM-predicted matrix yield properties and the ultimate failure strength of UD composite using the Curtis model [13].

2. GROUP INTERACTION MODELLING OF THE EPOXY RESIN MATRIX

Group Interaction Modelling is established as a tool for predicting polymer properties in the original text [4] and previously reported work [4,14]. In this paper, it is used to predict the properties of the epoxy resins matrix used in a fibre composite. GIM uses a hexagonal geometry so that a single polymer chain is surrounded by six neighbouring chains. The system is treated as a strong one-dimensional oscillator (the single chain) in a weak three-dimensional field (the neighbouring chains). The interactions between the chains are defined using a potential function that consists of several thermodynamic energy terms. The energy balance between attractive, cohesive energy ($E_{coh}$) and repulsive, thermal energy of skeletal mode vibrations ($H_T$) represents the equation of state for the system under investigation. This is shown in equation (1) where the total energy as defined by the model is

$$E_{total} = 0.89 E_{coh} - H_T = E_{coh} \left( \frac{V_0}{V} \right)^6 - 2 \left( \frac{V_0}{V} \right)^3$$

(1)

The polymer chain is defined as a series of repeating mer units, each of which is the smallest representative unit that still retains the chemical properties of the polymer. Each mer unit is further subdivided into functional groups each of which makes a well-defined contribution to the mer unit whole. The ensemble average thermodynamic properties of the functional groups are summed to give mer unit values which are used in the model. This method has the advantage of not requiring exact structural information such as atomic coordinates that are extremely difficult to obtain in branched polymers.

<table>
<thead>
<tr>
<th>Functional Group</th>
<th>N</th>
<th>$E_{coh}(0K)$ (J/mol)</th>
<th>$V_w$ (cm$^3$/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH$_2$</td>
<td>2</td>
<td>4500</td>
<td>10.25</td>
</tr>
<tr>
<td>Phenyl Ring</td>
<td>3</td>
<td>25000</td>
<td>43.3</td>
</tr>
<tr>
<td>N</td>
<td>2</td>
<td>9000</td>
<td>4</td>
</tr>
<tr>
<td>Epoxy Ring</td>
<td>4</td>
<td>15300</td>
<td>22</td>
</tr>
<tr>
<td>CH(OH)</td>
<td>2</td>
<td>20800</td>
<td>11.5</td>
</tr>
<tr>
<td>SO$_2$</td>
<td>2</td>
<td>45000</td>
<td>20.3</td>
</tr>
<tr>
<td>TGDDM Mer Unit</td>
<td>36</td>
<td>191700</td>
<td>232.9</td>
</tr>
<tr>
<td>DDS Mer Unit</td>
<td>12</td>
<td>113000</td>
<td>114.9</td>
</tr>
<tr>
<td>Cured TGDDM/DDS Mer Unit</td>
<td>18</td>
<td>152350</td>
<td>173.9</td>
</tr>
</tbody>
</table>

Table 1: GIM input parameters of constituent functional groups and mer units.
Each functional group contribution to the degrees of freedom, \( N \), the cohesive energy at absolute zero, \( E_{coh}(0K) \), the van der Waal’s volume, \( V_w \), is shown in Table 1 along with the uncured TGDDM and DDS mer unit totals. For TGDDM cured with DDS, the epoxy and amine are combined in a stoichiometric ratio of 1:1 as both components have 4 reactive sites. The GIM parameters for the cured TGDDM/DDS mer unit along with the length, \( L \), the molecular weight, \( M \), and the Debye temperature, \( \theta_1 \) are given in Table 1. Crosslinking is included by reducing \( N \) by 3 for each potential branching site on the mer unit; TGDDM and DDS have two each.

Two distinct transitions occur in the dynamic mechanical response of cured epoxy resins: the glass transition, which is already well-defined in Porter’s original text [4], and the low temperature beta transition which requires specific attention here. A characteristic beta temperature (\( T_\beta \)) and the cumulative loss tangent through the beta transition (\( \tan \Delta \beta \)) are applied to the loss tangent profile using a normal distribution. The beta transition is believed to be associated with an intramolecular crankshaft style motion of phenyl-phenyl segments within the polymer backbone [15]. The Arrhenius expression shown in equation (2) defines \( T_\beta \) in terms of the beta transition activation energy, \( \Delta H_\beta \), which is obtained using a simple quantum mechanics routine. Also, \( r \) is the strain rate, \( f \) is the characteristic vibrational frequency of the polymer chain (obtained from \( k\theta_1=hf \) where \( k \) and \( h \) are the Boltzmann and Planck constants respectively) and \( R \) is the gas constant. There is good agreement between the predicted and experimental \( T_g \) and \( T_\beta \), as shown in Table 2.

\[
T_\beta = \frac{-\Delta H_\beta}{R \ln \left( \frac{r}{2\pi f} \right)}
\]

(2)

An approximation of \( \tan \Delta \beta \) can be obtained using the ratio of elastic to plastic energy through the transition. In an individual ideal beta event, this can be expressed as the ratio of the energy required to perform the beta transition to the energy evolved as heat during it, as shown in equation (3). \( \Delta N \) and \( \Delta T \) represent the change in degrees of freedom and temperature through the transition respectively.

\[
\tan \Delta \beta = \frac{R \Delta N T_\beta}{R N \Delta T}
\]

(3)

The prediction of a series of thermomechanical properties using GIM begins with the prediction of volume and bulk modulus as a function of temperature. First, the two terms in the equation of state in equation (1) are predicted from the fundamental input parameters. The heat capacity is calculated using the Tarasov extension to the Debye model which is then integrated over temperature to give the thermal energy, \( H_T \). The cohesive energy as a function of temperature (\( E_{coh} \)) is predicted using a normal distribution to account for the change through the glass transition. The volume can be calculated either by solving the Lennard-Jones form of the potential function directly in equation (1) or via the thermal expansion coefficient. As the latter is important later in predicting stress-strain curves, this route will be shown. The volumetric thermal expansion coefficient, \( \alpha_v \), is calculated using the heat capacity and cohesive energy as shown in equation (4).

\[
\alpha_v = \frac{1.38 C_v}{R E_{coh}}
\]

(4)
Integrating $\alpha_v$ over temperature gives the volume as a function of temperature as shown in Figure 2 for TGDDM/DDS. The predicted and experimental expansion coefficient and density [5] are in good agreement as shown in Table 2.

Table 2: Comparison between predicted and experimental properties for TGDDM/DDS. All properties are measured at room temperature at a strain rate of 1 Hz with the exception of E and $\sigma_y$ which are at 0.00167 Hz.

<table>
<thead>
<tr>
<th>Property</th>
<th>GIM Predicted</th>
<th>Experimental</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_g$ ($^\circ$C)</td>
<td>281</td>
<td>270 (ref [5])</td>
</tr>
<tr>
<td>$T_\beta$ ($^\circ$C)</td>
<td>-38</td>
<td>-45 (ref [5])</td>
</tr>
<tr>
<td>$\rho$ (g/cm$^3$)</td>
<td>1.30</td>
<td>1.29 (ref [5])</td>
</tr>
<tr>
<td>$\alpha_L$ (x10$^{-6}$/K)</td>
<td>50</td>
<td>$\sim$50 (ref [16])</td>
</tr>
<tr>
<td>$E$ (GPa)</td>
<td>5.17</td>
<td>5.04 (ref [34])</td>
</tr>
<tr>
<td>$\sigma_y$ (MPa)</td>
<td>201</td>
<td>200 (ref [34])</td>
</tr>
</tbody>
</table>

The elastic bulk modulus, $B_e$, can be expressed in terms of the energy density in the system, as shown in equation (5). The total bulk modulus, $B$, including the plastic loss associated with the beta and glass transitions, is plotted as a function of temperature for TGDDM/DDS in Figure 2.

$$B_e = 18 \frac{E_{total}}{V}$$  \hspace{1cm} (5)

The predicted elastic modulus and Poisson’s ratio, $\nu$, for TGDDM/DDS are shown in Figure 3, where a typical modulus vs. temperature profile for an epoxy resin is
observed. The room temperature value of $E$ compares well to experiment as shown in Table 2. The predicted modulus is shown in tension while the experimental value is in compression, but this isn’t a problem at the relatively low strain values used in this work [18].

Figure 3: GIM predicted Tensile Modulus ($E$ in MPa, solid) and Poisson’s ratio ($\nu$, dotted) for TGDDM/DDS.

For the purposes of the model, the elastic and plastic contributions to the strain are defined separately. The elastic strain, $\varepsilon_e$, is calculated using equation (7) where the integral of the linear thermal expansion coefficient ($\alpha_t \approx \alpha_v/3$) over temperature gives the dimension change required. The plastic loss associated with the beta and glass transition events are included via the loss tangent as shown in equation (7) which gives the total strain, $\varepsilon$:

$$\varepsilon_e = \int_0^T \alpha_t \, dT \quad \& \quad \varepsilon = \varepsilon_e \left(1 + \int_0^T \tan \delta \, dT \right)$$

Figure 4: Comparison between the experimental (solid, ref [19]) and GIM predicted (dotted) Compressive Stress-Strain curve for TGDDM/DDS.
Now the elastic modulus and strain are known, the prediction of stress is given in equation (8), with a correction for tension to compression equal to twice Poisson’s ratio to correct for biaxial expansion in the two axes normal to the compression. Predicted and experimental compressive stress-strain curves for TGDDM/DDS are shown in Figure 4. The agreement between the two curves is excellent pre-yield and predicts the yield stress and strain very well (see Table 2). There is a period of post-yield strain softening and hardening in the experimental curve that is not replicated in the predicted curve. This effect is not currently included in the model and doesn’t affect the composite modelling in later sections.

\[ \sigma_c = \frac{E \epsilon}{2\nu} \]  

(8)

3. FINITE ELEMENT ANALYSIS

A 3D fibre composite was modelled by Ansys 10.1 containing six hexagonally packed carbon fibres enclosed by the TGDDM/DDS resin matrix. The fibre volume fraction of 50% was fixed by adjusting the distance between the fibres. Perfect interfacial bonding was achieved between the fibres and the matrix by ensuring coincidence of nodes along the fibre/matrix interfaces in the model. GIM-predicted stress-strain curves of the resin matrix were introduced into the model. The properties of the transversely orthotropic HTA5131 carbon fibres were taken from reference [20] and shown in Table 3.

Table 3: Mechanical properties of the carbon fibre employed in the FE model [20].

<table>
<thead>
<tr>
<th>Diameter (µm)</th>
<th>7.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>Young’s modulus (GPa)</td>
<td></td>
</tr>
<tr>
<td>( E_z )</td>
<td>235</td>
</tr>
<tr>
<td>( E_x = E_y )</td>
<td>13.8</td>
</tr>
<tr>
<td>Poisson’s ratio</td>
<td></td>
</tr>
<tr>
<td>( \nu_{yx} )</td>
<td>0.35</td>
</tr>
<tr>
<td>( \nu_{zy} = \nu_{zx} )</td>
<td>0.2</td>
</tr>
<tr>
<td>Shear modulus (GPa)</td>
<td></td>
</tr>
<tr>
<td>( G_{xy} )</td>
<td>5.11</td>
</tr>
<tr>
<td>( G_{yz} = G_{xz} )</td>
<td>18</td>
</tr>
</tbody>
</table>

The 8-noded solid brick structural element, SOLID45, was employed to mesh both the fibres and the matrix. In a static FE model, a fibre-break can be introduced into the model prior to the loading, as it is equivalent to the fracture of the fibre during loading. The top layer of the elements is removed from the fibre-end as shown in Figure 5. An axial displacement of 1% was applied on the front face of the model in z-direction while the back face was constrained with \( z=0 \). The model was constrained on its other faces to ensure stability. Moreover, the matrix nodes at the rim of the fibre-break were constrained to prevent rotation during deformation.

Figure 5: The meshed FE model.
A strain analysis was carried out because of the known failure response of these composites. Figure 6 shows the development of the axial tensile strain along the centre of the broken fibre. It can be seen that the yielding of the matrix has resulted in an exponential increase in the broken fibre axial tensile strain following the fibre fracture. Because of the elasto-plastic behaviour of the matrix, the applied strain cannot be fully recovered even at the far-field level, so it falls below the applied strain. The ineffective length of the fibre is considered as twice the length of the fibre fragment over which 90% of strain recovery occurred. The ineffective length of 0.161 mm is within the range of values reported in the literature [21].

![Figure 6: The axial tensile strain profile away from a fibre-break at the centre of the broken fibre within a hexagonal array at an applied strain of 1%.

The introduction of a break leads to a considerable increase in axial strain around the break. Figure 7 shows the regions of intensified strain in the surrounding fibres in the plane of fracture. The SCF of the adjacent fibre is considered as the ratio of the averaged axial strain at its cross section to the applied strain. The strain concentration factors for the nearest neighbouring and the next-nearest neighbouring fibres in the FE model containing the TGDDM/DDS resin matrix were found to be 1.11 and 1.009, respectively. However, the latter is almost one and can therefore be disregarded.

![Figure 7: Contour map of the axial tensile strain in the nearest and next nearest neighbouring fibres to a fibre-break in. The resin matrix is not shown.](image)
The elasto-plasticity of the matrix has been incorporated into a FE model and led to a significant decrease in strain concentration factor in the fibres in comparison with the non-yield (elastic) matrix case which is considered to be 1.17 [9]. As the elasto-plastic matrix at the vicinity of a broken fibre absorbs energy to deform plastically, it reduces the stress concentration on the intact adjacent fibres of a broken fibre [22]. This reduces the probability of failure of the adjacent fibres and eventually of the UD composite material.

4. STATISTICAL MODEL

The statistical model predicts the progression of fibre fractures in a layer of UD composite. The model consisted of 20 unidirectional carbon fibres of the size of the ineffective length, 0.161 mm, embedded in TGDDM/DDS resin matrix. Each fibre adopted a failure strain randomly from a normal distribution generated by the Box-Muller method [23]. The mean strain of a fibre of an ineffective length was obtained by extrapolating the data for longer lengths [24]. In the model procedure, the weakest fibre fails when the applied strain exceeds its failure strain. Following a fibre fracture, an overload is shed equally among its six intact nearest neighbouring fibres according to the SCF. With two adjacent fibres broken, the overload is transferred onto their eight adjacent fibres and so on. The number of fibres affected by the failures continuously changes with the occurrence of new fibre-breaks. Thus, the local intensified strain is updated after each fibre-break for all surrounding fibres. The procedure is repeated until an unstable crack occurs and the fibres start to fail successively without an increase in the applied strain, and thus the layer fails. The applied strain at this stage is taken as the failure strain of the composite layer. The programme can be ended when 2-3% of the fibres in a layer have failed [13]. Figure 8 shows two examples of fibre failure sequence in typical composite layers. Initially, stable groups of broken fibres form in the layer that withstand the overload imposed by the broken fibres, which then become unstable at higher applied strains.

![Figure 8: The sequence of fibre failure (numbered) in two examples of a fibre composite layer with different random fibre failure distributions. White and shaded fibres represent intact and broken fibres, respectively.](image)

Therefore, the failure strain of the layer is determined by the weakest fibre when failure begins. Table 4 shows the mean failure strain of the composite layer and the average size of the largest stable group of broken fibres.

In the next stage, a fixed number of the single layers are stacked together to form a piece of UD composite of a known length. However, in this preliminary study, the interaction between the single layers is not considered. The failure is assumed to occur when random fibre breaks have weakened one layer so that it can no longer withstand
the applied load. The average failure strain of a known size of composite, $\bar{\varepsilon}_{\text{composite}}$, can be obtained by:

$$\bar{\varepsilon}_{\text{composite}} = \bar{\varepsilon}_{y} - z_{l}\sigma_{l}$$  \hspace{1cm} (9)

where $\bar{\varepsilon}_{y}$ is the average failure strain of the layers, $z_{l}$ is the probability of layer failure and $\sigma_{l}$ is the standard deviation of failure strain in the layers.

Table 4: The layer failure strain and the size of stable group of broken fibres (the layer thickness is 0.161 mm).

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean failure strain of single fibre (%)</td>
<td>2.47 ± 0.49</td>
</tr>
<tr>
<td>Layer strain at the first fibre failure (%)</td>
<td>1.42 ± 0.25</td>
</tr>
<tr>
<td>Layer failure strain (%)</td>
<td>1.83 ± 0.16</td>
</tr>
<tr>
<td>Mean size of stable group of broken fibres</td>
<td>1.5 ± 0.9</td>
</tr>
</tbody>
</table>

It was assumed that a specific number of single layers with a random distribution of failure strains are stacked to make a fibre composite. Figure 9 shows the change in failure strain of the 3D composite with logarithmic length. The failure strain becomes independent of length above a few centimetres. In this preliminary study, it is assumed that all neighbouring fibres around fractured fibres share an equal overload regardless of their absolute distances from the crack. The composite failure strain predicted in this preliminary study is greater than typical experimental values (nearly 1.12%) for a given length of composite[13]. This may be associated with the choice of ineffective length. It has been demonstrated that a slight increase in the layer thickness decreases the composite failure strain and results in a better agreement with experiment. Interaction between layers may also be significant and has not been taken into account in this analysis.

![Figure 9: The predicted failure strain for the UD composite with TGDDM/DDS matrix as a function of length.](image)

5. CONCLUSIONS
A multiscale modelling technique for predicting the thermomechanical and engineering properties of a fibre-reinforced composite system has been described. Group Interaction
Modelling has been used to predict full stress-strain curves for a multifunctional amine cured epoxy resin matrix. The development of this technique to cope with a two component, branching polymer is a significant step forward in modelling terms. The model predicts the non-linear dependence of a variety of thermomechanical and engineering properties as a function of temperature.

The tensile failure strain of a 3D fibre composite has been predicted using a Monte Carlo simulation. The effect of shear yielding in the resin matrix at the vicinity of fibre fractures is considered through the introduction of SCF values. The predicted failure results fall better in a range of real composite failure strains compared to a non-yield matrix. However, a better agreement with the experiment requires a detailed study of the influence of the ineffective length of the fibres.

REFERENCES
20. Nedele M. R., University of Bristol, Bristol, 1996, PhD.