ISOTHERMAL AND NON-ISOTHERMAL CONSOLIDATION OF UNIDIRECTIONAL CARBON FIBRE-THERMOPLASTIC TOWPREGS

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SUMMARY: The present work reports the utilisation of a recently proposed model for the consolidation of unidirectional thermoplastic towpregs in isothermal and non-isothermal conditions. A Finite Difference Method program was developed to simulate the towpreg consolidation in non-isothermal conditions. To assess which thermal condition shall be used in practice, pressure/displacement curves determined experimentally from compression tests carried out at constant speed are compared with those obtained from the isothermal and non-isothermal simulations. In spite of the good agreement between the experimental and theoretical data, the results have shown that the compression moulding of unidirectional towpregs can only be well predicted in isothermal conditions when slow closing speeds and long pre-heating times are used.

KEYWORDS: Modelling, Isothermal and non-isothermal consolidation, Compression moulding, Carbon fibres, Thermoplastic matrix composites, Towpregs, Powder coating process, Finite difference method (FDM).

INTRODUCTION

Thermoplastic matrices are advantageously used instead of thermosets to produce long fibre reinforced structural composites [1-4]. The impregnation of long fibres with thermoplastics brings difficulties that can be overcome through the production and use of powder coated towpregs [5-8].

Various studies on compression moulding and simulation of the processing cycle of unidirectional carbon/polycarbonate (C/PC) towpregs have been published recently [9, 10]. An isothermal model was established for the consolidation, relating the applied pressure to the operational (mould closing speed and temperature) and material variables (fibre/polymer content and packing arrangement, matrix viscosity and powder and fibre dimensions).

In this work, compression tests were carried out on the same C/PC towpreg preforms. The experimental data are compared with the previous isothermal model simulations and those of a new model that assumes non-isothermal consolidation. This model is based on a finite difference method (FDM) to determine the evolution of the temperature through the material thickness from the mould and towpreg initial temperatures and thermal properties.
THEORY

The powder coated thermoplastic matrix towpregs (Fig. 1a), as manufactured by the cost-effective dry coating technique [4-6, 9], can be seen as a lamina of an array of fibres with polymer particles attached. To produce a composite, a preform (Fig. 1b) consisting of a number of these lamina stacked together is compression moulded [4, 9, 10].

Fig.1: Powder coated thermoplastic matrix towpregs

The consolidation model is detailed elsewhere [9, 10]. Consequently, only its main aspects will be reviewed in this section. The model is based upon the following assumptions: i) the polymer and the fibres are incompressible; ii) the fibres in the lamina stack are regularly assembled (Figs. 1 and 2); iii) during consolidation, each polymer particle forms a bridge between the adjacent fibres (Fig. 2); iv) upon compression the bridges spread in the direction of the fibres and the pressure is considered constant through thickness; v) the mould closing speed is constant and no voids are left after consolidation and, vi) the polymer flow is considered laminar and steady, and the inertial forces are negligible.

It has been shown that the particle geometrical arrangement assumed in modelling affects the results of the simulations, and better results are obtained with a hexagonal arrangement (Figs. 1b) and 2) [10]. At constant speed, the press closing speed, $s_p$, coincides with the moulding thickness decreasing rate, $s_p = -\frac{d t}{h(t)} = -\dot{h}(t)$. The fibre, matrix and void volume contents are related to the instantaneous thickness $h(t)$ respectively by:

$$v_f = \frac{h_{fi} v_{fi}}{h(t)}; \quad v_m = \frac{h_{fi} (1 - v_{fi})}{h(t)}; \quad v_v = \frac{h(t) - h_{fi}}{h(t)} \quad (1)$$

where $h_{fi}$ and $v_{fi}$ are the final moulding thickness and fibre volume fraction, respectively.

The pressure gradient ($d p/d x$) required to cause flow in the fibre direction is obtained by application of the continuity, Darcy and Carman-Kozeny equations to the polymer bridge [9, 10, 11-14]. Considering that $p(x) = 0$ at the ends of the polymer bridge ($x = \pm L(t)/2$), the pressure applied over the full bridge length ($L$) is determined by [9, 10]:

$$p = -\frac{C h(t)}{[h(t) - h_{fi} v_{fi}]^6} \quad (2)$$

where $C$ is a constant dependent on the fibre/polymer arrangement

$$C = \frac{64 \eta k_{xx} (h_{fi})^5 (v_{fi})^4 (1 - v_{fi}) (r_p)^6}{27 (r_f)^6} \quad (3)$$

where
η polymer viscosity

\( r_f, r_p \) fibre and polymer particle radii, respectively

k\(_{xx}\) Carman-Kozeny constant, \( k_{xx} = 0.7 \) (according to Gutowski and al. [12])

Fig. 2: Initial polymer bridge for the hexagonal arrangement; \( L_0 \) and \( L \) are the initial and final polymer bridge lengths, respectively

The instantaneous polymer shear rate in the bridge is calculated using the concept of hydraulic radius of the circular cross-section [9, 10, 13].

**Isothermal Consolidation**

Assuming an isothermal process, the instantaneous pressure developed during compression can be analytically determined if the fibre and particle polymer radii, and the final moulding thickness and fibre volume fraction are known [9, 10]. At each instant of consolidation, all material layers are at the mould temperature. The evolution of pressure during compression is directly determined from Eqn. 2, considering that the instantaneous thickness decreasing rate equals the press closing speed.

An Arrhenius power law was used to relate the polymer viscosity, \( \eta \), at each mould temperature

\[
\eta = m_o \cdot e^{\left( \frac{\Delta E}{R \cdot T} \right)} \times (\dot{\gamma})^{n-1}
\]  

(4)

where \( T \) is the mould absolute temperature, \( R \) the gas constant and the following polymer constants were determined experimentally:

- \( m_o \) polymer consistency when \( T \) approaches infinity (Pa.s\(^n\))
- \( \Delta E \) polymer flow activation energy
- \( \dot{\gamma} \) polymer shear rate (s\(^{-1}\))

**Non-Isothermal Consolidation**

In non-isothermal conditions, the temperature varies through the thickness. As the polymer viscosity depends on temperature, the material layers deform at different rates. Thus, to calculate the instantaneous pressure, it is necessary to determine numerically the temperature gradient along the laminate \( z \) co-ordinate axis (Fig. 3). A program was developed to that end. Considering as negligible the generation and transfer of heat by viscous dissipation and convection, the instantaneous temperature at a point of co-ordinate \( z \) is calculated using finite differences to solve the simplified equation of energy:

\[
\frac{\partial T}{\partial t} = \alpha \frac{\partial^2 T}{\partial z^2}
\]  

(5)
where \( \alpha \) is the material thermal diffusivity,

\[
\alpha = \frac{k}{\rho C_p}
\]  

(6)

Considering that the origin of the co-ordinate axes is located at the centre of the laminate, the following boundary conditions are considered (Fig. 3):

\[
T_{\text{mould}} = \text{const} \quad \text{and} \quad \text{at } z = \pm \frac{h_{\text{tot}}}{2} \quad T = T_{\text{mould}}
\]  

(7)

\[\text{Fig. 3: Towpreg laminate used in the non-isothermal model}\]

Fig. 4 shows the flowchart used in the program. As can be seen, the determination of the temperature gradient is made in two steps using small increments of time (\( \Delta t \)): a) in the first step, the material, consisting of layers of equal thickness, is pre-heated without pressure, and the temperature gradient through the material thickness is calculated at the beginning of compression; b) in the second step and during consolidation at constant press closing speed, \( s_p \), the layers are considered to have different viscosity and thermal properties, and to deform at different rates.

It can be shown that the total number of layers, \( n \), and the laminate initial thickness, \( h_{o}^{\text{tot}} \), are determined by:

\[
n = \text{integer} \left( \frac{h_{fi}^{t} - 2 r_f}{h_{f}^{(j)}} \right)
\]  

(8)

\[
h_{f}^{(j)} = r_f \sqrt{\frac{\pi \sqrt{3}}{2 v_{fi}}}
\]  

(9)

\[
h_{o}^{\text{tot}} = n \times h_{o}^{(j)} + 2 r_f
\]  

(10)

\[
h_{o}^{(j)} = \frac{3}{2} (r_f + r_p)
\]  

(11)

where, \( h_{fi}^{t} \), \( h_{o}^{(j)} \) and \( h_{f}^{(j)} \) are the total laminate thickness at the end of consolidation and the initial and final thicknesses of layer \( j \), respectively.
During compression, the polymer viscosity is determined at each layer, using Eqn. 4 and the temperature calculated at its middle-plan at each instant, \( t \). Thus, considering Eqns 2 and 3 and a constant pressure across the thickness, the instantaneous deformation rate of each layer \( j, \dot{\mathbf{h}}^{(j)} \), is calculated by solving the system of \( n \) equations:

\[
\dot{\mathbf{h}}^{(j)} = \frac{\eta^{(j+1)}}{\eta^{(j)}} \left[ \mathbf{h}^{(j)} - h_f v_f \right]^{6} \mathbf{h}^{(j+1)}
\]

\[
\sum_{j=1}^{n} \dot{\mathbf{h}}^{(j)} = s_p
\]

where the closing speed is equal to the sum of the deformation rates of all layers in the last equation and \( \eta^{(j)} \) and \( h^{(j)} \) are the viscosity and layer thickness at the instant, \( t \).

From the layer deformation rate, \( \dot{\mathbf{h}}^{(j)} \), the position of the layers in the \( z \)-axis can be determined for the next instant, \( t_{i+1} = t_i + \Delta t \). The conductivity and specific heat, \( k^{(j)} \) and \( C_p^{(j)} \), are then calculated as:

\[
\frac{1}{k^{(j)}} = \frac{v_f^{(j)}}{k_f} + \frac{v_p^{(j)}}{k_p} + \frac{v_v^{(j)}}{k_v}
\]
where $k_f$, $k_p$, $k_v$ are the conductivities of the fibre, polymer and air, and $v_f^{(j)}$, $v_p^{(j)}$, $v_v^{(j)}$ their volume fractions in layer $j$. At each layer, Eqn. 1 is used to determine the volume fraction of the components. The specific heat is

$$C_p^{(j)} = C_{pf} w_f^{(j)} + C_{pp} w_p^{(j)} + C_{pv} w_v^{(j)}$$  \hspace{1cm} (14)

where $C_{pf}$, $C_{pp}$, $C_{pv}$ are the specific heats of the fibre, polymer and air, and $w_f^{(j)}$, $w_p^{(j)}$, $w_v^{(j)}$ their mass fractions in layer $j$. In each layer, the mass fraction of the components is determined from the volume fractions calculated by Eqn. 1 using their densities.

**EXPERIMENTAL WORK**

**Characterisation of materials**

A polycarbonate powder (Bayer Makrolon 2458) and PAN carbon fibres (Amoco Thornel T300/12/NT) were used to produce the C/PC towpregs in a dry powder coating unit [4, 5]. The dimension and size distributions of the PC powder were determined by using the image analysis and sieving techniques. The PC powder particles, that are irregular in shape, have an average diameter of 156 $\mu$m.

The shear rate viscosity of the polycarbonate was measured with a TA Instruments Weissenberg Rheogoniometer. The viscosity data at four temperatures (220, 240, 260 and 280 °C) and shear rates up to 10 s$^{-1}$ were used to calculate the parameters of Eqn. 5,

$$m_o = 46.3 \text{ nPa.s}^n, \quad \frac{\Delta E}{R} = 12860 \text{ K} \quad \text{and} \quad n = 0.953.$$

The average diameter of the PAN carbon fibres, 7.2 $\mu$m, was determined by a laser diffraction technique. The fibre volume fraction was calculated as 25.3 $\pm$ 2.04 %.

**Consolidation tests**

An instrumented 800 kN SATIM press with two independently heated plates was used to produce laminates at temperatures ranging from 220 to 280 °C. The pressure and plate displacement were continuously monitored and recorded in a x-t recorder. The towpreg laminate was pre-heated in the mould during 10 min. The consolidation conditions are summarised in Table 1.

| **Table 1:** Typical processing conditions used for the consolidation of the laminates |
|---|---|---|---|---|
| **PROPERTY** | **Units** | **Sample reference** |
| Temperature (T) | °C | A | B | C | D |
| Press closing speed ($s_p$) | mm.s$^{-1}$ | 0.035 | 0.045 | 0.05 | 0.10 |
| Viscosity ($\eta$) | Pa.s | 8810 | 2460 | 1000 | 570 |
| Average shear rate ($\dot{\gamma}$) | s$^{-1}$ | 55 | 60 | 72 | 120 |
Modelling studies

Isothermal Conditions

Figure 5 compares the normalised pressure data with the results of the isothermal model, considering the fibre/polymer hexagonal and square arrangements [9].

![Graph showing experimental and theoretical pressure data as function of material instantaneous thickness.]

Fig. 5: Experimental and theoretical pressure data as function of the material instantaneous thickness (from [10]).

Although the fitting is quite good, the isothermal simulations provide little information about the influence of the pre-heating time and the closing speed. For example, pressures greater than expected were obtained at the initial stages of compression. This situation certainly results from the lower temperature at the internal layers of the laminate. On the other hand, upon increasing $s_p$, higher pressures are developed because of the slow heat transfer that limits the temperature that the material internal layers can reach.

Non-Isothermal Conditions

Figure 6 shows the evolution of the temperature across thickness during the consolidation of the C/PC towpreg for three different pre-heating times (5, 10 and 20 min).

![Graph showing variation of temperature across thickness for different pre-heating times.]

Fig. 6: Variation of the temperature across thickness for different pre-heating times at various compression times $t_c=0, 2, 4$ and $6$ min.
The plots were calculated considering the towpreg and mould initial temperatures as 25°C and 260 °C, respectively, and the following properties:

<table>
<thead>
<tr>
<th></th>
<th>Thermal conductivity (W/m.K)</th>
<th>Density (kg/m³)</th>
<th>Specific heat (kJ/kg·K)</th>
<th>Radius (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fibre</td>
<td>8.5</td>
<td>1760</td>
<td>0.925</td>
<td>3.6</td>
</tr>
<tr>
<td>Polymer</td>
<td>0.25</td>
<td>1200</td>
<td>1.8</td>
<td>78</td>
</tr>
<tr>
<td>Air</td>
<td>0.025</td>
<td>1.1</td>
<td>1.005</td>
<td>-</td>
</tr>
</tbody>
</table>

\[
v_{fi}=25\%, \ h_{f_i}^{tot}=2 \text{ mm}, \ s_p=0.05 \text{ mm/s}, \ m_o=46.3 \text{ nPa.s}^n, \ \frac{\Delta E}{R} =12860 \text{ K}, \ n=0.953,
\]

As shown in Fig. 6, the temperature across thickness has a significant dependence on the pre-heating time. For these C/PC towregs, it is only possible to reach an almost constant temperature across thickness after 20 min of pre-heating. If 5 minutes are used instead, the polycarbonate at the centre of the laminate remains solid during most of the compression.

In Figure 7, the pressure data obtained from the consolidation tests at 260 °C are compared with the simulations using the isothermal and non-isothermal models. It is apparent that the non-isothermal simulations are closer to the experimental results in special at the initial stages of compression. However, the difference is not very significant, which probably means that the 10 minutes pre-heating used in the compression tests were sufficient to guarantee a quasi-isothermal situation.

![Graph showing pressure data comparison](image)

**Fig. 7: Comparison between the experimental data and the isothermal and non-isothermal simulations at 260°C**

As Fig. 8 shows, at the beginning of the compression, the layer in contact with the mould (layer 1) decreases in thickness much rapidly than the middle-plan laminate layer (layer 84). However, as the top and bottom layers reduce their thicknesses more rapidly, they become more difficult to deform. This explains the tendency, shown in Fig. 8, of all layers displaying similar deformation rates at later stages of compression in spite of relatively large differences in temperature as shown in Figure 6.
CONCLUSIONS

Models to simulate the consolidation of powder coated towpregs in isothermal and non-isothermal conditions were developed in this work and compared with experimental data. The results showed that the isothermal model simulations could only predict successfully the material consolidation if the towpregs were previously pre-heated at the mould temperature. The consolidation speed affects the accuracy of the simulation when the isothermal model is used. The comparison between experimental and simulation results showed that better accuracy is achieved when a non-isothermal model is used. Therefore, the optimisation of the production rate by fine-tuning of the pre-heating time must be based on a model of this type. Also, a model like the one developed in this work may help to achieve process optimisation by studying the consolidation during cooling.

REFERENCES


