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THE IMPACT OF PROCESS PARAMETERS ON THE RESIDUAL STRESSES AND DISTORTIONS IN PULTRUSION

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1 General Introduction

Pultrusion is a manufacturing process for producing continuous lengths of fiber reinforced composite shapes with a constant cross section. While pultrusion machines vary in design, the process is basically the same. Fibers and resin materials are pulled through a heated forming die using a continuous pulling system and the cured profile is cut into its final length. A schematic view of the pultrusion process can be seen in Fig. 1.

In literature, thermo-chemical analyses of the pultrusion process have been carried out both numerically and experimentally [1-9] However, there is not much information about thermo-mechanical aspects of the pultrusion process such as the evolution of the mechanical properties, process induced stresses, strains and deformations. The residual stresses and distortions arise in the composite manufacturing processes due to a number of different mechanisms [10-13]: (i) the mismatch in the coefficient of thermal expansion (CTE) of the matrix material and the fibers, (ii) the chemical shrinkage of the matrix material, (iii) the tool-part interaction and (iv) the temperature and the degree of cure gradients through the composite thickness due to non-uniform curing. In [14], the development of the process induced residual stresses and strains together with the distortions were investigated using the cure hardening instantaneous linear elastic (CHILE) approach [13]. It was found that, tension stresses prevail for the inner region of the composite since the curing rate is higher here as compared to the outer regions where compression stresses are obtained at the end of the process. In [15], an integrated modelling of the pultrusion process of a NACA0018 blade profile was carried out by combining a 3D Eulerian thermo-chemical analysis and a 2D quasi-static plane strain mechanical analysis. The calculated residual stresses were transferred into the subsequent bending simulation of the pultruded blade profile and the internal stress distribution was evaluated. Apart from the pultrusion process, there are several studies in the literature regarding the mechanical modeling of the composite manufacturing processes such as resin
transfer molding (RTM), vacuum infusion, and autoclave [16-30].

In the present study the process induced residual stresses and distortions of a pultruded unidirectional (UD) glass/epoxy square profile are predicted. Two different case studies are carried out based on two different set temperatures of the heaters with varying pulling speeds from 10 cm/min to 60 cm/min. The spatial temperature and cure degree distributions are first obtained in the three dimensional (3D) thermo-chemical analysis (Eularian frame). After obtaining the temperature and degree of cure distribution in 3D, the process induced residual stresses and distortions in the transverse directions are evaluated in a 2D quasi-static mechanical analysis [14]. Using the proposed numerical model, the effects of the heating configuration and pulling speed on the process induced variations are investigated.

2 Numerical Implementation

2.1 Thermo-chemical Model

In the 3D thermo-chemical simulation of the pultrusion process, the energy equations given in Eq. 1 (for the composite part) and Eq. 2 (for the die) are solved.

\[ \rho \cdot C_p \left( \frac{\partial T}{\partial t} + u \cdot \frac{\partial T}{\partial x_3} \right) = k_{x_1,c} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2,c} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3,c} \frac{\partial^2 T}{\partial x_3^2} + q \]

\[ \rho_d \cdot C_p \cdot d \left( \frac{\partial T}{\partial t} = k_{x_1,d} \frac{\partial^2 T}{\partial x_1^2} + k_{x_2,d} \frac{\partial^2 T}{\partial x_2^2} + k_{x_3,d} \frac{\partial^2 T}{\partial x_3^2} \right) \]

where \( T \) is the temperature, \( t \) is the time, \( u \) is the pulling speed, \( \rho \) is the density, \( C_p \) is the specific heat and \( k_{x_1,c}, k_{x_2,c}, \) and \( k_{x_3,c} \) are the thermal conductivities along \( x_1, x_2 \) and \( x_3 \) directions, respectively. The subscripts \( c \) and \( d \) correspond to composite and die, respectively. Lumped material properties are used and assumed to be constant. The internal heat generation \( q \) [W/m^3] due to the exothermic reaction of the epoxy resin can be expressed as [7, 14]:

\[ q = (1-V_f) \rho_f H_v \left[ R_e(\alpha) \right]_e \]  

where \( V_f \) is the fiber volume fraction and \( H_v \) is the total heat of reaction, \( \rho \) is the density of epoxy resin, \( \alpha \) is the degree of cure and \( [R_e(\alpha)]_e \) is the effective cure rate, i.e. \( \frac{d\alpha}{dt} \), expressed as

\[ [R_e(\alpha)]_e = K_e \exp\left(-\frac{E}{R_T}\right) \cdot (1-\alpha)^n \cdot f(\alpha, T) \]  

where \( K_e \) is the pre-exponential constant, \( E \) is the activation energy, \( R \) is the universal gas constant and \( n \) is the order of reaction (kinetic exponent). \( K_e, E, H_v \) and \( n \) can be obtained by a curve fitting procedure applied to the experimental data evaluated using differential scanning calorimetry DSC [7]. \( f(\alpha, T) \) is the diffusion factor which accounts for the glass transition effect defined as [13]:

\[ f(\alpha, T) = \frac{1}{1 + \exp[C(\alpha - (\alpha_{c0} + \alpha_{CT} T))]} \]

where \( C \) is a diffusion constant, \( \alpha_{c0} \) is the critical degree of cure at \( T = 0 \) K and \( \alpha_{CT} \) is a constant for the increase in critical \( \alpha \) with \( T \) [13]. The relation of the effective resin kinetics equation can be expressed as [14]

\[ \frac{\partial \alpha}{\partial t} = \left[ R_e(\alpha) \right]_e - u \frac{\partial \alpha}{\partial x_3} \]

where it is the expression in Eq. 6 which is used in the numerical model.

For the thermo-chemical simulation of the pultrusion process, the control volume based finite difference (CV/FD) method [1, 4, 5, 31] is used in order to obtain the temperature and degree of cure distributions. MATLAB [32] is used for the implementation of the CV/FD. The nonlinear internal heat generation (Eq. 3) together with the resin kinetics equation (Eq. 4) are coupled with the energy equation (Eq. 1) in an explicit manner in order to obtain a straightforward and fast numerical procedure. The degree of cure is subsequently
updated explicitly for each control volume using Eq. 6 in its discretized form.

### 2.2 Thermo-chemical-mechanical Model

The effective mechanical properties of the composite are calculated by using the self consistent field micromechanics (SCFM) approach which is a well known technique in the literature [12, 14, 20]. For the instantaneous epoxy resin elastic modulus, the CHILE approach [13], which is a valid pseudo-viscoelastic approximation of the linear viscoelasticity [30], is used. This approach includes the cure hardening and also thermal softening [13] and is suitable for the pultrusion process since the shorter process times in pultrusion, as compared to the composite manufacturing processes such as RTM, Vacuum Infusion, Autoclave, only further support the use of the linear elastic CHILE approach. The glass transition temperature \( T_g \) is calculated using the Di Benedetto equation [13, 14]. An incremental linear elastic approach is implemented utilizing user-subroutines in ABAQUS [33] for the calculation of the residual stresses and distortions as used in [12, 14, 15].

### 3 Thermo-chemical Analysis

#### 3.1 Problem description

The 3D thermo-chemical simulation of the pultrusion of a square profile is carried out in a Eulerian frame. A UD glass/epoxy based composite and a steel die are used for the composite and the die block, respectively. Material properties and the resin kinetic parameters are listed in Table 1 and Table 2, respectively. The parameters used for the diffusion factor (Eq. 5) are given in Table 2. Only a quarter of the pultrusion domain, seen in Fig. 2, is modeled due to symmetry. At the symmetry surfaces adiabatic boundaries are defined in which no heat flow is allowed across the boundaries. The remaining exterior surfaces of the die are exposed to ambient temperature (27 °C) with a convective heat transfer coefficient of 10 W/(m² K) except for the surfaces located at the heating regions. Similarly, at the post die region, convective boundaries are defined for the exterior surfaces of the pultruded profile. The length of the post die region \( L_{conv} \) in Fig. 2 is determined to be approximately 13.7 m [14].

Two case studies are carried out based on the set temperature of the heaters which are given in Fig. 2. The set temperatures of the first case study (Case-1) are taken from [7] and for the second case (Case-2), the corresponding temperatures are taken from [6]. In each case study, six different pulling speeds, 10 cm/min, 20 cm/min, 30 cm/min, 40 cm/min, 50 cm/min and 60 cm/min, are used which indicates that the total pultrusion process time is approximately 146.4 min, 73.2 min, 48.8 min, 36.6 min, 29.3 min and 24.4 min, respectively based on the total length of the pultruded profile which is taken approximately as 14.6 m in the simulation.

#### 3.2 Results

The calculated temperature and degree of cure distributions at the interior region of the part (point A) are depicted in Fig. 3 for the two different heater configurations (Case-1 and Case-2 in Fig. 2) with

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**Table 1. Material Properties [7].**

<table>
<thead>
<tr>
<th>Material</th>
<th>( \rho ) (kg/m³)</th>
<th>( C_p ) (J/kg K)</th>
<th>( k_{x_1} ) (W/m K)</th>
<th>( k_{x_1}, k_{x_2} ) (W/m K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Composite (( V_f = 63.9% ))</td>
<td>2090.7</td>
<td>797.27</td>
<td>0.9053</td>
<td>0.5592</td>
</tr>
<tr>
<td>Die</td>
<td>7833</td>
<td>460</td>
<td>40</td>
<td>40</td>
</tr>
</tbody>
</table>

**Table 2. Epoxy Resin Kinetic Parameters [7, 14].**

<table>
<thead>
<tr>
<th>( H_v ) [kJ/kg]</th>
<th>( K_0 ) [1/s]</th>
<th>( E ) [kJ/mol]</th>
<th>( n )</th>
<th>( C )</th>
<th>( \alpha_{C0} )</th>
<th>( \alpha_{CT} ) [1/K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>324</td>
<td>192000</td>
<td>60</td>
<td>1.69</td>
<td>30</td>
<td>-1.5</td>
<td>0.0055</td>
</tr>
</tbody>
</table>
pulling speeds of 10, 30 and 50 cm/ min. It is seen that in general an increase in speed slows down the curing together with heating and cooling rates. A similar trend is also the case for the outer regions (e.g. point B). The corresponding temperature and cure degree values of point A and point B at the end of the process (at $x_3 \approx 14.6$ m) are shown in Fig. 4. As expected, the temperature values of the composite part at the end of the process increase because of the slower cooling rate and the degree of cure decrease owing to a slower curing rate with an increase in the pulling speed. For lower pulling speeds, higher degree of cure values are obtained at the end of the process in Case-2 as compared to the Case-1 due to the occurrence of the curing reaction under the third heater which has the highest set temperature of 200°C (Fig. 2).

The maximum composite temperatures are given in Table 3 for Case-1 and Case-2. It is seen that the maximum temperature of the outer region (point B) in Case-2 is higher than the one in Case-1 for all the pulling speed values. For the inner region (point A), the maximum temperature in Case-2 is higher for pulling speed of 20 and 30 cm/min. This provides higher curing rates and hence, higher degree of cure values are obtained for pulling speeds of 20 and 30 cm/min in Case-2 (Fig. 4) as compared to the Case-1 even though the set temperatures of the first two heaters in Case-2 (105.5 °C and 148.5 °C) are much lower than the ones in Case-1 (171 °C and 188 °C). However, for higher speeds, the degree of cure values in Case-2 at the end of the process drop much faster as compared to Case-1 with an increase in the pulling speed. This shows that the relatively high set temperature of the last heater in Case-2 is not sufficient to cure the product as in Case-1 for pulling speeds greater than 30 cm/min.

The variation of the maximal through thickness cure degree difference ($\Delta \alpha_{\text{max}}$) during the process between points A and B is shown in Fig. 5. It is seen that $\Delta \alpha_{\text{max}}$ begins decreasing after a pulling speed of 30 cm/min for both cases due to the decrease in curing together with heating and cooling rates. As earlier mentioned, the through thickness cure degree gradients can result in non-uniform curing and hence the internal constraints arise [12].
Table 3. The maximum composite temperature values (°C) for different pulling speeds.

<table>
<thead>
<tr>
<th>Speed (cm/min)</th>
<th>Case-1</th>
<th>Case-2</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Point A</td>
<td>Point B</td>
</tr>
<tr>
<td><strong>10</strong></td>
<td>213.3</td>
<td>190.7</td>
</tr>
<tr>
<td><strong>20</strong></td>
<td>216.7</td>
<td>191.9</td>
</tr>
<tr>
<td><strong>30</strong></td>
<td>214.7</td>
<td>188.4</td>
</tr>
<tr>
<td><strong>40</strong></td>
<td>206.7</td>
<td>182.9</td>
</tr>
<tr>
<td><strong>50</strong></td>
<td>195.0</td>
<td>180.0</td>
</tr>
</tbody>
</table>

Fig. 3. Temperature (top) and degree of cure (bottom) distributions at point A (center of the composite) for Case-1 and Case-2 with different pulling speeds.

Fig. 4. Temperature and degree of cure values for point A and point B at the end of the process.

Fig. 5. Variation of the maximum through thickness cure degree difference between points A and B for different pulling speed values.
4 Thermo-chemical-mechanical Analysis

4.1 Problem description

The evolution of the process induced strains and stresses in transverse directions are predicted using a 2D plane-strain model. Since the cross sectional dimensions (0.0254 m) are much smaller than the total length of the pultruded profile (~14.6 m) in the pulling direction, a plane strain assumption is made for the residual stress analysis [14]. In this model, the cross section of the part is assumed to move along the pulling direction of the process while tracking the corresponding temperature and degree of cure profiles calculated in the 3D thermo-chemical simulation. In other words, a 3D Eulerian thermo-chemical model is coupled with a 2D plane strain Lagrangian mechanical model (see Fig. 6) [14]. In this 2D model, a realistic mechanical contact formulation at the die-part interface, which allows separation of the composite from the rigid die surface due to the shrinkage of the part, is defined.

The corresponding stresses, strains and displacements are calculated based on the temperature and the cure distributions together with the corresponding \( T_g \) of the cross section by using the quadratic plane-strain elements in ABAQUS.

It should be noted that the resin CTE in rubbery state \((T_g > T)\) is known to be approximately 2.5 times of the CTE in glassy state \((T_g < T)\) [10, 26, 27]. The constants for the CHILÉ model and the Di Benedetto equation together with the mechanical properties of the epoxy resin and the fiber are taken from [14]. In the present study, the total volumetric shrinkage of the epoxy resin is assumed to be 6% [14].

4.2 Results

The displacement evolutions of the point B (Fig. 2), which is dominated by the combination of the thermal expansion and the chemical shrinkage, in the \( x_2 \)-direction (U2) are depicted in Fig. 7. Note that the results should be seen together with the results obtained in the thermo-chemical analysis in Section 3.2. It is seen from Fig. 7 that the separation between the part and the die at point B occurs only for the pulling speed of 10 cm/min and for higher speeds there is no separation observed at the interface. The locations of the glass transition \((T_g)\) points are also indicated as triangular markers in Fig. 7. It is seen that \( T_g \) shifts along the pulling direction as the speed is increased. After \( T_g \), the increase in magnitude of the displacement becomes smaller due to the change in CTE of the epoxy resin. The shift in \( T_g \) with an increase in the pulling speed provides a larger rubbery region (from beginning of the process to \( T_g \)) having a CTE of 2.5 times of the CTE in the glassy state. However, it should be born in mind that at the same time the curing and cooling rates decrease (Fig. 3). The final displacements for point B at the end of the process are shown in Fig. 8. It is seen that similar trend is obtained for the residual displacement (U2) at point B in both heater configurations (Case-1 and Case-2). The magnitude of the displacement increases up to 30 cm/min and starts decreasing after that speed due to the drop in curing and cooling rates (Fig. 3).
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Fig. 7. Displacement evolutions for point B in the $x_2$-direction $U_2$ (left). The corresponding zoomed plot of $U_2$ for axial distance between 0-1.2 m (right).

Fig. 8. The residual displacement values for point B (at the end of the process) in the $x_2$-direction ($U_2$).

The evolution of the process induced transverse normal stresses in the $x_1$-direction ($S_{11}$) are shown in Fig. 9 and Fig. 10. A similar trend for the stress development during the process is obtained as in [14] such that tension stresses prevail at the inner region (point A) and compression stresses occur at the outer region (point B) while upholding the self static equilibrium in which there is no applied external load. It is seen that after $T_g$, there is a sharp drop in the stress levels for point A which is in tension (Fig. 9), on the other hand, a sharp increase in stress levels is observed after $T_g$ for point B which is in compression (Fig. 10) because of the change in CTE of the epoxy resin. This affects the stress development such that the stresses are built-up with a slower rate as the speed increases. The variations in the stress levels around the $T_g$ locations (drop in the tensile stress levels for point A and increase in the compression stress levels for point B) are owing to the non-uniform $T_g$ distribution over the cross-section such that at lower speeds, point B changes its state from rubbery to glassy first and for the higher speeds glass transition occurs first at point A which changes the behavior and the position of the internal constraint.

For the second heater configuration (Case-2), relatively lower stress levels are obtained for pulling speed greater than 30 cm/min as compared to Case-1 because the composite is cured and cooled down slower in Case-2 for higher speeds. This can be seen from Fig. 11 in which the residual stresses ($S_{11}$) for point A and point B at the end of the process are depicted. It is seen that the stress levels start decreasing with a pulling speed greater than 20 cm/min and 30 cm/min for point A and point B, respectively. Since there is static equilibrium, the changes in the residual stress are in the opposite direction for point A and point B, e.g. if the tension stress level decreases for point A, the corresponding compression stress level for point B also decreases (Fig. 11).
The undeformed contour plots of S11 for Case-2 are depicted in Fig. 12 with a pulling speed of 20 cm/min and 60 cm/min. It is seen that the pattern of the stress distribution varies with the pulling speed. The position of the maximum compression stress remains the same (i.e. point B), however the position of the maximum tension stress (originally at point A) moves towards point B.

It should be noted that the characteristics of the process induced stresses and distortions highly depend on the competition between the cross sectional temperature, degree of cure and $T_g$ developments together with the instantaneous stiffness evolution of the composite.

5 Conclusions

In this work, a 3D thermo-chemical analysis in Eulerian frame was coupled with a 2D quasi-static mechanical analysis in a Lagrangian frame for the pultrusion process simulation of a UD glass/epoxy square profile (25.4 mm × 25.4 mm). In the mechanical analysis of the process, 2D quadratic plane strain elements were used for the calculation of the process induced stresses and distortions [14]. The effects of the two different heater configurations (Case-1 and Case-2 in Fig. 2) with a varying pulling speed between 10-60 cm/min on the process induced variations were investigated.
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Fig. 12. Undeformed contour plots of the residual stress at the end of the process for pulling speed of 20 cm/min (a) and 60 cm/min (b).

It was found that the pulling speed, which is one of the most significant process parameters, has a nonlinear effect on the residual stresses and distortions for different heater configurations. The possible reasons are the changes in the internal constraints, non-uniform cross sectional curing rates and glass transitions. The residual stress (Fig. 11) and distortion (Fig. 8) levels together with the maximal through thickness cure gradient (Fig. 5) in Case-1 were found to be relatively high as compared to Case-2 at the end of the process for pulling speeds greater than 30 cm/min. The cross sectional stress distribution is changed with an increase in the pulling speed (Fig. 12) which shows that the internal dynamics are shifted towards the outer regions. Therefore, it is not trivial to obtain the optimum process parameters to get the desired manufacturing process conditions.

References


