Pultrusion manufacturing process development by computational modelling and methods

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Abstract

This paper deals with the modelling and development of computational schemes to simulate pultrusion processes. Two different computational methods, finite differences and elements, are properly developed and critically analyzed. The methods are applied to a model, implemented with suitable boundary conditions, proposed in this paper. Simulations, compared with experimental data available in the literature, show a good agreement between theory and experiments.

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1. Introduction

Pultrusion is a continuous manufacturing process used to shape polymeric composite materials into parts with constant cross section. The reinforcement fibres, in the form of continuous strands (roving) or mats, are placed on creel racks; fibres are pulled through a guide plate and then impregnated passing by a resin bath. Uncured material crosses a preform plate system to be correctly shaped before entering into the curing die. The die entrance is characterized by a tapered shape to remove the resin excess and make the material inlet easier. A water cooling channel is placed in the first part of the die, to prevent premature material solidification; the heat for material polymerization is provided by heating platens placed on the top and bottom surfaces of the die. Outside the die, the cured composite material is pulled by a continuous pulling system (caterpillar or reciprocating pullers) and then a travelling cut-off saw cuts the part into desired length.

The interest in developing numerical models of complex manufacturing process is documented in [1]. In particular, in recent years, several researchers have performed numerical and experimental investigations on different problems related to the pultrusion process, focusing the attention on the analysis of the heat transfer and cure, on the pressure rise in the tapered zone of the die, and on problems related to impregnation of reinforcing fibers by thermoplastics.

Models for microscopic or macroscopic impregnation during the pultrusion process of thermoplastic matrix composites have been proposed in [2–4]. Suitable material models to close the conservation equation, based on Darcy’s
law for the flow through a porous media, for the pressure and velocity fields in a pultrusion die have been proposed by Raper et al. in [5] and by Gadam et al. in [6].

The importance of the analysis of heat transfer and cure in pultrusion is highly recognized to obtain a final product characterized by the desired mechanical properties or to realize a post-die shaping. A three dimensional model of heat transfer and cure is proposed in [7] and [8] by Chachad et al. Discretization requirements and the influence of process parameters have been studied in [9] using a bi-dimensional finite element model. Thermochemical aspects of the process have been investigated by Liu et al., who developed several numerical models, based on finite difference method, on finite element method, and on nodal control volume techniques [10–12]. Temperature dependent material properties and resin shrinkage were taken into account in [13] by Joshi et al., who in [14, 15] proposed an optimization procedure based on finite element/nodal control technique. In [16,17] finite element simulations of the resin flow in injection pultrusion were proposed. An experimental investigation on bent-pultrusion was performed by Britnell et al. in [18].

Two different three dimensional models, which take into account thermochemical aspects of the process are compared and critically analyzed in this paper. Both models consider heat transfer due to heating platens, convective boundary conditions, die-cooler at the die entrance, composite material anisotropy, mass transport effects, and the heat generated by the resin exothermic cure reaction. Section 2 deals with the theoretical formulation of the thermochemical model; FDM and FEM modelling, initial and boundary conditions are exposed in Section 3. Simulations are compared with experimental data and discussed in Section 4, while Section 5 deals with some concluding remarks and a brief analysis of research perspectives.

2. Thermochemical analysis

2.1. Heat transfer model

A correct modelling of the pultrusion process needs to take into account not only the processing composite material, but all tools related to heat transfer. Different equations for the energy balance of the forming die and the composite materials are required, taking into account the exothermic resin cure reaction and convective effect, related to the movement of the processing part.

The energy balance equation, for a generic domain, can be written as follows:

$$\frac{\partial}{\partial t} \int_V \rho u dV = - \int_S \phi_{\text{conv}} \cdot n dS - \int_S \phi_{\text{diff}} \cdot n dS + \int_V u_{\text{gen}} dV,$$

(2.1)

where $\rho$ is material density, $\phi_{\text{conv}}$ and $\phi_{\text{diff}}$ are the unitary convective and diffusive fluxes, respectively, $u$ is the specific internal energy, $u_{\text{gen}}$ is the rate of internal energy generation, $V$ is the control volume and $S$ is its boundary surface.

In particular the convective flux, which represents the internal energy variation related to material movement, is:

$$\int_S \phi_{\text{conv}} \cdot n dS = \int_S \rho u v \cdot n dS,$$

(2.2)

where $v$ is the velocity vector. The diffusive flux is given by:

$$\int_S \phi_{\text{diff}} \cdot n dS = \int_S q \cdot n dS,$$

(2.3)

where $q$ is the unitary heat flux through the control surface.

Taking into account Eqs. (2.2) and (2.3), the conservation equation (2.1) can be rewritten as follows:

$$\frac{\partial}{\partial t} \int_V \rho u dV = - \int_S \rho u v \cdot n dS - \int_S q \cdot n dS + \int_V u_{\text{gen}} dV.$$

(2.4)

Assuming that the material density $\rho$ is constant, specific internal energy is equal to the product between the heat capacity $c$ and the temperature $T$, and heat flux $q$ is the opposite of the product between material conductivity $k$ and temperature gradient. The balance equation (2.4) can be rewritten, in local form, as follows:

$$\rho c \frac{\partial T}{\partial t} + \rho c v \nabla T = k \nabla^2 T + u_{\text{gen}}.$$

(2.5)
The heat transfer equation for the heated die, in Cartesian coordinate system, can be finally written as follows:

$$
\rho_d c_{p,d} \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \left( k_{x,d} \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( k_{y,d} \frac{\partial T}{\partial y} \right) + \frac{\partial}{\partial z} \left( k_{z,d} \frac{\partial T}{\partial z} \right),
$$

(2.6)

where $\rho_d$ is the die material density, $c_{p,d}$ is the specific heat, and $k_{x,d}$, $k_{y,d}$, and $k_{z,d}$ are the thermal conductivities in $x$, $y$, and $z$ direction, respectively.

Taking into account that reinforcing fibres are wetted out and impregnated by the resin before entering the die, it is assumed that the resin does not flow, and then the heat transfer equation for the composite part can be written, assuming the $x$ axis as the pull direction, as follows:

$$
\rho_c c_{p,c} \left( \frac{\partial T}{\partial t} + v_x \frac{\partial T}{\partial x} \right) = k_{x,c} \frac{\partial}{\partial x} \left( \frac{\partial T}{\partial x} \right) + k_{y,c} \frac{\partial}{\partial y} \left( \frac{\partial T}{\partial y} \right) + k_{z,c} \frac{\partial}{\partial z} \left( \frac{\partial T}{\partial z} \right) + \rho_r v_r Q,
$$

(2.7)

where $\rho_r$ is the resin volume fraction ($\rho_r = 1 - \rho_f$, $\rho_f$ being the fiber volume fraction), $v_x$ is the pull speed, $\rho_c$ is the composite material density, $\rho_r$ is the resin density, $c_{p,c}$ is the specific heat, $k_{x,c}$, $k_{y,c}$, and $k_{z,c}$ are the thermal conductivities in the $x$, $y$, and $z$ directions respectively, and $Q$ is the specific heat generation rate due to resin exothermic cure reaction.

Composite material density, specific heat, and conductivities can be evaluated using the rule of mixtures.

2.2. Crystallization kinetics model

During the forming process, polymeric composite materials are subjected to the cure process, characterized by an exothermic reaction which causes an increase of material temperature. Several kinetic models can be found in the literature to describe the cure process; generally kinetic models relate the rate of resin reaction $R_r$ (equal to the first derivative of the degree of cure with respect to time) to temperature and degree of cure, according to the following equation:

$$
R_r(\alpha) = \frac{d\alpha}{dt} = K(T) f(\alpha),
$$

(2.8)

where $\alpha = \alpha(t)$ is the degree of cure (conversion fraction of the reactive species), $t$ is the reaction time, $K$ is a temperature dependent parameter, according to the Arrhenius equation:

$$
K(T) = K_0 \exp \left( \frac{-\Delta E}{RT} \right),
$$

(2.9)

and:

$$
f(\alpha) = (1 - \alpha)^n g(\alpha),
$$

(2.10)

where $K_0$ is a constant, $\Delta E$ is the activation energy, $T$ is the absolute temperature, $R$ is the gas universal constant, $n$ is the order of the reaction (kinetic exponent), and $g(\alpha) = 1 + C$, $C$ being the autocatalytic intensity; it is assumed, in the analysis developed in what follows, $g(\alpha) = 1$.

Finally Eq. (2.8) is:

$$
R_r(\alpha) = \frac{d\alpha}{dt} = K_0 \exp \left( \frac{-\Delta E}{RT} \right) (1 - \alpha)^n.
$$

(2.11)

Taking into account that the degree of cure is defined as the ratio of the amount of heat evolved during the curing process up to time $t$ (indicated as $H(t)$), to the total heat of reaction (indicated as $H_{tr}$), yields:

$$
\alpha = \frac{H(t)}{H_{tr}},
$$

(2.12)

and

$$
R_r(\alpha) = \frac{d\alpha}{dt} = \frac{dH(t)}{H_{tr} dt}.
$$

(2.13)
Finally, the heat generation rate can be rewritten as follows:
\[ Q = \frac{dH(t)}{dt} = H_{tr} R_r(\alpha). \] (2.14)

\( H_{tr} \) and \( R_r \) can be experimentally evaluated using differential scanning calorimetry (DSC) measurements. In the pultrusion process, the concentration of the resin species into the forming die is governed by the following hyperbolic equation with nonlinear source:
\[ \frac{\partial \alpha}{\partial t} = R_r(\alpha) - v_x \frac{\partial \alpha}{\partial x}. \] (2.15)

3. Computational simulations

3.1. Finite difference simulations

The three dimensional temperature field in the die and the work piece can be evaluated solving two second-order parabolic partial differential equations (Eqs. (2.6) and (2.7)). Taking into account that a second order derivative can be written using a second order central finite difference formulation, and considering the time derivative, one finally has seven dependent variables for each equation. A fast and efficient procedure was obtained by decoupling the generative term in Eq. (2.7), as follows:

\[
\rho c_{\text{p},c} \left[ \left( \frac{\partial T}{\partial t} + v_x \frac{\partial T}{\partial x} \right) \right]^{n+1} = \left[ k_{x,c} \frac{\partial^2 T}{\partial x^2} + k_{y,c} \frac{\partial^2 T}{\partial y^2} + k_{z,c} \frac{\partial^2 T}{\partial z^2} \right]^{n+1} + [\rho_r V_r Q]^n
\] (3.1)

\[ \left[ \frac{\partial \alpha}{\partial t} \right]^{n+1} = \left[ R_r(\alpha) - v_x \frac{\partial \alpha}{\partial x} \right]^n. \] (3.2)

The efficiency of the scheme was improved by splitting the solution algorithm, i.e. by splitting the system of algebraic equations, into a number of sub-steps equal to the number of considered dimensions and decoupling the generative term.

In this way, in each sub steps, only three terms, associated with a particular coordinate direction, are treated implicitly. As a consequence, implicit terms can be grouped adjacent to the main diagonal and the solution can be obtained by the Thomas algorithm. The splitting technique adopted was the Alternating Direction Implicit (A.D.I.) method (Peaceman and Rachford 1955). The A.D.I. scheme, however, is characterized, for three dimensional problems, by computational economy, spatially second order accuracy, and only conditional stability. An extended recent bibliography on finite differences and elements methods can be found [19–23], as well as in the bibliography therein cited.

Finite difference formulations of Eq. (3.1) are written as follows:
\[ T_{i,j,k}^{n+1} - T_{i,j,k}^n = -c \left( T_{i,j,k}^{n+1} - T_{i-1,j,k}^{n+1} \right) + r_x \left( T_{i-1,j,k}^{n+1} - 2T_{i,j,k}^{n+1} + T_{i+1,j,k}^{n+1} \right) \]
\[ + r_y \left( T_{i,j-1,k}^{n+1} - 2T_{i,j,k}^{n+1} + T_{i,j+1,k}^{n+1} \right) + r_z \left( T_{i,j,k-1}^{n+1} - 2T_{i,j,k}^{n+1} + T_{i,j,k+1}^{n+1} \right) + q^n, \] (3.3)

where:
\[ r_x = \frac{k_{x,c} \Delta t}{\rho c_{\text{p},c} \Delta x^2}; \quad r_y = \frac{k_{y,c} \Delta t}{\rho c_{\text{p},c} \Delta y^2}; \quad r_z = \frac{k_{z,c} \Delta t}{\rho c_{\text{p},c} \Delta z^2}; \quad c = \frac{\Delta t}{\Delta x} v_x; \quad q = \frac{\rho_r V_r Q}{\rho c_{\text{p},c}}. \] (3.4)

Splitting Eq. (3.3) into the three directions of the coordinate system and assuming the time step for each sub-step as \( \Delta t / 3 \), sub-steps, associated to the \( x \), \( y \), and \( z \) direction, respectively, can be written as follows:
\[ -(c + r_x) T_{i-1,j,k}^{n+1/3} + (1 + c + 2r_x) T_{i,j,k}^{n+1/3} - r_x T_{i+1,j,k}^{n+1/3} = T_{i,j,k}^n \left( 1 - 2r_y - 2r_z \right) + r_y \left( T_{i,j+1,k}^n + T_{i,j-1,k}^n \right) \]
\[ + r_z \left( T_{i,j,k-1}^n + T_{i,j,k+1}^n \right) + \frac{1}{3} q^n; \] (3.5)
in steady state condition: 

\(-r_y T_{i-j-1,k} + (2r_y + 1) T_{i,j,k}^{n+2/3} - r_y T_{i,j+1,k}^{n+2/3} = (c + r_x) T_{i-1,j,k}^{n+1/3} + (1 - c - 2r_x) T_{i,j,k}^{n+1/3} + r_x T_{i+1,j,k}^{n+1/3} + r_z \left(T_{i,j,k-1}^{n+1} - 2T_{i,j,k}^{n+1} + T_{i,j,k+1}^{n+1}\right) + \frac{1}{3} q^n; \)

\(-r_z T_{i,j,k}^{n+1} + (1 + 2r_z) T_{i,j,k}^{n+1} - r_z T_{i,j,k+1}^{n+1} = (r_x + c) T_{i-1,j,k}^{n+1/3} - (c + 2r_x) T_{i,j,k}^{n+1/3} + r_x T_{i,j+1,k}^{n+1/3} + r_y T_{i,j-1,k}^{n+2/3} + (1 - 2r_y) T_{i,j,k}^{n+2/3} + r_y \left(T_{i,j-1,k}^{n+2} + T_{i,j+1,k}^{n+2}\right) + \frac{1}{3} q^n. \)

A similar analysis can be easily obtained also for Eq. (2.6).

The degree of cure was evaluated solving Eq. (2.15) using the upwind method, as follows:

\[ \alpha_{i,j,k}^{n+1} = [R_r(\alpha)]^n \Delta t + \left(1 - v_x \frac{\Delta t}{\Delta x}\right) \alpha_{i,j,k}^n + v_x \frac{\Delta t}{\Delta x} \alpha_{i-1,j,k}^n. \]

### 3.2. Finite element simulations

The pultrusion process is characterized by the non-linear aspect related to the coupling between the energy equation (2.7) and the species equation (2.15). As a result, even assuming material properties as constants, the finite element matrix is asymmetric and cannot be solved by several commercial finite element packages. The ANSYS finite element code, used for the present investigation, allows us to solve non-linear problems; however, taking into account that the rate of cure reaction is a function of the temperature and the temperature field can be strongly influenced by the generative term due to the exothermic reaction, a direct implementation of the generative term is not possible.

In [10–15] an approach to problem computation, based on a finite-element/nodal-control-volume technique, has been proposed using both finite element and finite difference methods. Finite difference grids and nodal control volumes have been constructed on the finite element mesh; the convective and the generative terms have been decoupled from the energy equation and evaluated at the previous time step.

In this paper a relatively simpler and more correct procedure is proposed, since the convective term is included into the energy equation and only the generative term has been decoupled from the energy equation. According to the developed procedure the steady state temperature field is initially calculated, taking into account the imposed boundary conditions, assuming the degree of cure as null and the composite temperature as the temperature of the resin bath in each node. In each point of the composite material, an overestimated value of the reaction rate can be evaluated using Eq. (3.7) and then the degree of cure is adjourned according to the following equation, obtained from Eq. (2.15) in steady state condition:

\[ v_x \frac{\partial \alpha}{\partial x} = R_r(\alpha). \]

The values of degree of cure, obtained by the above method, are then used to evaluate a relatively more accurate value of the reaction rate and the new values of the heat generation are applied to evaluate the temperature field at the second next step of iteration. The above procedure is repeated, using results of each load step to evaluate reaction rate, degree of cure, and heat generation to be used in the successive step, until the temperature difference, obtained in each node between two consecutive iterations, satisfies a previously defined convergence criterion.

### 3.3. Initial and boundary conditions

The following initial and boundary conditions have been used to solve the above formulated boundary value problem:

- The initial degree of cure $\alpha$ of the composite is assumed as null, its initial temperature $T_{\text{com}}$ is equal to the resin bath temperature $T_{\text{resin}}$, and initial temperature of the die $T_{\text{die}}$ is assumed as the room temperature $T_{\text{room}}$:

\[ T_{\text{com}}(x, y, z, t = 0) = T_{\text{resin}}, \]
\[ T_{\text{die}}(x, y, z, t = 0) = T_{\text{room}}, \]
\[ \alpha(x, y, z, t = 0) = 0. \]
The temperature and the degree of cure of the processing material in the cross section of the die entrance is constrained to the above values:

\[ T_{\text{com}}(x = 0, y, z, t) = T_{\text{resin}}, \]
\[ \alpha(x = 0, y, z, t) = 0. \] (3.11)

– The composite cross section, at the die exit, is modelled as adiabatic:

\[ \frac{\partial T_{\text{com}}(x, y, z, t)}{\partial x} \bigg|_{x=L} = 0; \] (3.12)

– Adiabatic conditions are imposed also in symmetry sections.

– Constant temperatures \( T_{\text{imp}} \) are imposed in opportune nodes to simulate the heating platens and the die-cooler:

\[ T_{\text{die}}(x, y, z, t) = T_{\text{imp}}. \] (3.13)

– Convective boundary conditions are imposed on external die surfaces:

\[ q_{\text{cond}} = -h_{\text{conv}}(T_{\text{die}} - T_{\text{room}}), \] (3.14)

where \( q_{\text{cond}} \) is the convective flux and \( h_{\text{conv}} \) is the convective coefficient.

– Taking into account the relatively low pull speed, at the interface between the die and the part, it is assumed that the conductive heat flow is orthogonal to the die axis:

\[ k_{y,c} \frac{\partial T_{\text{com}}}{\partial y} \bigg|_{c} = k_{y,d} \frac{\partial T_{\text{die}}}{\partial y} \bigg|_{d}, \]
\[ k_{z,c} \frac{\partial T_{\text{com}}}{\partial y} \bigg|_{c} = k_{z,d} \frac{\partial T_{\text{die}}}{\partial y} \bigg|_{d}. \] (3.15)

4. Results and discussion

The pultrusion of a work piece with a C cross section has been simulated to validate the proposed finite difference and finite element models by comparing numerical results with results in Ref. [15]. Fig. 1(a), (b), and (c) show, respectively, the geometry of the model and its spatial discretization in the cross section for the finite difference and finite element model. Only half a model has been considered for symmetry reasons.

Die length \( L \), width \( W \), and height \( H \) are 915 mm, 72 mm, and 72 mm, respectively. Composite section dimensions are shown in Fig. 1(c). The die is heated by six platens, whose dimensions are 255 mm (length) and 72 mm (width), where three platens are placed on the top surface and the remaining on the bottom surface. The platen’s temperature is assumed as indicated in Table 1 (\( T_1 \), \( T_2 \), and \( T_3 \) refer to the heating platens on the top of the die, from die entrance to die exit; \( T_4 \), \( T_5 \), and \( T_6 \) refer to the heating platens on the bottom of the die, in the same order). Consecutive platens are spaced by 30 mm and the empty zone is subjected to convective boundary conditions. Between the die entrance
and the first platen, along a distance of 90 mm, a water cooling channel is placed to avoid resin premature gelation. Die cooler temperature is assumed as 50 °C. The part enters into the die at the temperature of 30 °C. Pull speed is 2.299 mm/s. Room temperature is assumed as 30 °C and the convective coefficient is considered as 10 W/mm² K. Symmetry sections are modelled using adiabatic conditions. The properties of the considered materials and the resin kinetic parameters are listed in Tables 2 and 3, respectively.

A comparison between cure profiles in points A and B, indicated in Fig. 1(b) and (c), obtained using FDM and FEM proposed models, with data from Ref. [15] is shown in Fig. 2. Temperature peak, mean degree of cure in the exit section, and standard deviation of the degree of cure, in the same section, are compared in Table 4. As evidenced, very good agreement has been found with data from the literature.

In particular, relatively more accurate values, for temperature peak and mean degree of cure, have been found using the finite element model, which has provided an overestimated value of the standard degree of cure. The finite difference model seems to provide underestimated values for all the considered parameters. These (negligible, according to the authors’ opinion) differences are influenced by the model spatial discretization in the pull direction (node numbers in the pull direction are 101 and 1947, for the FD and FE model, respectively), as well as in the cross section. The relatively high Peclet number, indeed, affects the accuracy of the solution; however, it can be considered acceptable, taking into account the reduced computation time (a Peclet number smaller than two needs too much computational time to be used in iterative optimization procedures). It is interesting to underline that, for the finite difference model, the evaluation of the mean value of the degree of cure and of its standard deviation in the final cross section of the material provides values (0.8896 and 0.0044, respectively), which are relatively closer to the respective values in Ref. [15], using the same points.

5. Conclusions and perspectives

In this paper two different approaches to the simulations of thermochemical aspects of the pultrusion manufacturing process have been proposed, compared and discussed. Governing equations, boundary conditions, numerical (finite difference and finite element) methods and procedures are described. Numerical results, in particular, temperature peak, mean degree of cure in the final cross section and its standard deviation, and cure profiles along pull direction have been compared with data provided by the previous literature. Very good agreement was found. Taking into account the obtained results, it can be concluded that:
Table 4
Simulation results

<table>
<thead>
<tr>
<th>Data</th>
<th>Temperature peak (°C)</th>
<th>Mean degree of cure</th>
<th>Standard deviation of degree of cure</th>
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</thead>
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<tr>
<td>Ref. [15]</td>
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<td>0.0045</td>
</tr>
<tr>
<td>Finite difference model</td>
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<td>Finite element model</td>
<td>217.27</td>
<td>0.8913</td>
<td>0.0049</td>
</tr>
</tbody>
</table>

Fig. 2. Cure profiles.

i. A remarkable accuracy has been achieved using the proposed finite element method; which requires also less computational time, with respect to the finite difference model, using the same value of the Peclet number (the used FE code imposes the respect of the conditions related to the Peclet number).

ii. The finite difference procedure is numerically stable and accurate also for values of the Peclet number larger than its limit value; however other performed simulations evidenced a non-acceptable increase of computational time for relatively more correct Peclet numbers.

iii. Both models can be used as good tools to investigate the influence of process parameters, such as temperature and location of the heating platens, temperature of the die cooler and of the resin bath, and pull speed, on temperature and degree of cure profiles and distributions. Cure profiles can be investigated also for parts characterized by different resin volume fraction, as well as by different section.

iv. Finite element modelling appeared to be relatively more appropriate to model parts characterized by irregular or curve cross sections, taking into account the major flexibility of the node’s disposition into the cross section of composite and die.

v. Implementation of non-linear aspects (related to variations in material properties and to radiative effects), alternative heat sources (microwave assisted pultrusion), and post-die shaping results are easier using the finite element method, taking into account that reduced variation of the model, due to the software capability used, is needed.

vi. Taking into account the accuracy and the stability achieved by the finite difference model, as well as the relatively easier implementation in automatic procedures, it can be also used for process optimization, instead of more expensive experimental investigation.

References


