NUMERICAL STUDY OF THE PULTRUSION PROCESS USING A PARABOLIC APPROACH

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Abstract. *Pultrusion is one of the manufacturing processes which are used for continuous fabrication of fiber-reinforced polymer composites of constant cross-section. In this process, dry fibrous reinforcement is pulled through a resin bath for resin impregnation. Composite materials possess superior properties such as high strength and stiffness to weight ratio, resistance to environmental deterioration, high electrical insulation and low assembly cost. Over the years, these materials have found increasingly wide applications in a variety of industries including the aerospace and chemical engineerin. More recently, pultruded products are gaining increasing market share in civil infrastructure applications due to their unique advantages over traditional steel and concrete materials. The pultrusion process can be understood as an advection-reaction-diffusion problem modeled by two partial differential equations, energy and a degree of cure transport equations. These equations are coupled by a reactive source-term from the resin curing exothermic reaction. At this work, a two-dimensional parabolic model using the finite element method was employed to determine the temperature and the degree of cure distributions inside the pultruded material with circular cross-section. This numerical study will allow to evaluate the effect of different parameters (such as: fiber volume, pulling speed and die-wall set point temperature) on the degree of cure of the final product composed by epoxy resin reinforced with carbon fiber. Therefore, an adequate knowledge of these characteristics contributes for a better pultrusion process design.*

Keywords*. Pultrusion, Finite Element Method, Epoxy Resin, Carbon Fiber.*

1. Introduction

Composite materials have been used in a variety of industrial as well as military applications. The very high strength-to-weight ratio for polymeric composite materials offers numerous design advantages from typical civil engineering uses to high performance aircraft and spacecraft applications. Composites exhibit excellent mechanical performance and good flexibility of design. They have increase service life and required less maintenance compared with conventional metals.

Pultrusion is a manufacturing method for composite materials with constant cross sections. It is a continuous process and therefore has a high productivity.

Conceptualized in the 1950's, pultrusion is one of several processes in which composite materials can be manufactured. Pultrusion appears to be deceivingly simple, and therefore for decades engineers have relied on the virtuosity and experience of pultruders to manufacture composites, rather than understanding the science, Chachad et al (1995). It is a greater competition and sophisticated manufacturing process.

A schematic representation of the pultrusion is shown in Fig. 1. Fibers are pulled from a creel through a resin bath and then it pass through a heated die. The die completes the impregnation of the fiber, controls the resin content and cures the material to its final shape as it passes through the die. This cured profile is then automatically cut to length.

Although the process appears to be simple, numerous process variables such as: pull speed, die temperature, quality of fiber/resin wet-out, thermal properties, die temperature and fiber volume profile can affect the pultruded product final quality.

Many previous works have analyzed the pultrusion process simulation. Chachad, Roux, Vaughan and Arafat (1995) used an unsteady three-dimensional model for the pultrusion of irregular shaped glass composites using a finite difference scheme and no comparison of predicted results and experimental dada was provided.

Gorthala, Roux and Vaughan (1994) reported a steady-state-two-dimensional numerical model for studying the manufacture of cylindrical composites by the energy, species, and momentum equations using a finite difference approach.

Suratno et al. (1998) performed a numerical simulation of carbon fibers reinforced with resin epoxy, using the finite element method. The fiber had different transversal and longitudinal thermal conductivity values but these results aren't compared with the isotropic case. The effect of the pulling speed in the temperature and degree of axial cure profiles of the composite material is also analyzed.

Ma and Chen (1993) developed a kinetic model for heat transfer to predict profiles of temperature and degree of cure in a pultruded glass-fiber composite of rectangular cross-section for a block-polyurethane resin by using a finite-difference method.

Wu and Joseph (1990) developed an unsteady-state model to describe transient conditions for start-up or change of operation conditions, which includes the degree of cure and temperature distributions in the material and die.

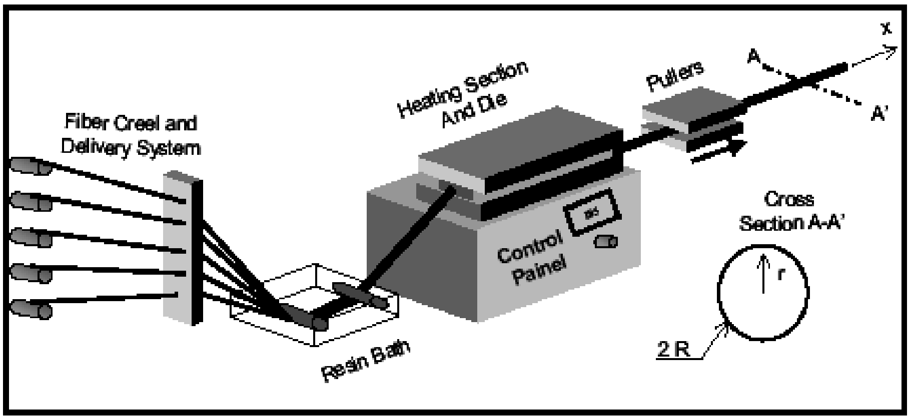


Figure 1. Schematic representation of the pultrusion process.

Han and Lee (1986) applied an empirical kinetic model with a prescribed wall temperature profile in the simulation. Pultrusion characteristics were studied with variables such as resin type, fiber type and the pulling speed.

At this context, the objective of the present work is to study numerically the pultrusion process using a parabolic approach to simulate the Shell Epon 9420/9470/537 epoxy resin curing, evaluating the degree of cure and temperature profiles under different conditions, such as: fiber volume, pulling speed and die-wall set-point temperature. Results showed that the pulling and the set-point temperature presented opposite effects. As the pulling speed value increases (or Tw decreases) the degree of cure is reduced. The fiber fraction volume (FV) influence indicated that lower FV values increases the cure rate. The knowledge of these effects can contribute for better operational conditions during the pultrusion process.

2. Mathematical formulation

The mathematical formulation is based in the following assumptions:
- the process is steady state two-dimensional;
- the material properties (density, specific heat, thermal conductivity) are constant at any resin-degree of cure;
- the composite material is isotropic;
- the effect of pressure on the heat of reaction is neglected.

Employing the conservation of energy principle, the two- dimensional steady-state energy equation for the non-moving parts of the pultruder (metal platens, insulators, heaters and the die) can be represented by Eq. (1).

$$uCp_b \frac{\partial T}{\partial x} - \nabla \cdot \left[k_b \nabla T \right] - \frac{\partial}{\partial x}\left(k_b \frac{\partial T}{\partial x} \right) = \dot{q} \tag{1}$$

with the boundary conditions:

at x = 0 (die entry) → T = Te (ambient temperature)

at $y^2 + z^2 = R^2$ (bar/die interface) T = Tset (control set point temperature)

where:

x – axial coordinate;
y, z – coordinate axis at the bar cross-section;
u – pulling speed;
T – absolute temperature;
ρ_b – bulk density;
Cp_b – bulk specific heat;
k_b – thermal conductivity;
q – volumetric heat rate due to the resin cure reaction.

$\nabla \cdot$ – divergence operator at the bar cross-section;
∇ – gradient operator at the bar cross-section.

The bulk composite material density (ρ_b) is calculated by a mass fraction method (Roux et al, 1998) expressed by:

$$\rho_b = \frac{1}{\frac{M_f}{\rho_f} + \frac{M_r}{\rho_r}} \quad \text{and} \quad M_f = \frac{FV}{FV + \left(\frac{\rho_r}{\rho_f} \left((1 - FV \right) \right)} \tag{2}$$

where M is the mass fraction and the subscripts f, r and b correspond to fibers, resin and bulk composite material, respectively.

The thermal properties of fiber and resin used in computation of the temperature and degree of cure profiles are tabulated in Table. 1. The bulk thermal conductivity (k_b) and the bulk specific heat (Cp_b) of the composite were obtained using also the mass fraction method, Eqs. 3 and 4:

$$k_b = \frac{1}{\frac{M_f}{k_f} + \frac{M_r}{k_r}} \tag{3}$$

$$Cp_b = (FV)Cp_f + (1 - FV)Cp_r \tag{4}$$

where FV is fiber fraction volume.

Table 1. Thermal properties of AS4/Epon 9470/537 composite material (Valliappan et al., 1996)

Material	Density (g cm^{-3})	Specific heat (J g^{-1} K^{-1})	Thermal conductivity (W m^{-1}K^{-1})
Fiber	1.79	712	11.6
Matrix	1.26	1255	0.2

The volumetric heat rate (q) due to the resin-cure reaction is related to the degree of cure (α) by the Eq. 5:

$$\dot{q} = \rho r (1 - FV) \Delta H \frac{D\alpha}{Dt} \tag{5}$$

with ΔH = total heat of reaction per unit mass of resin.

The degree of cure is defined as the ratio between the energy liberated by the reaction until an instant of time (t) and the total energy liberated in whole cure reaction. The degree of cure variation with the time is calculated by:

$$\frac{D\alpha}{Dt} = \left[A\,exp\left(-\frac{E}{RT} \right) \right] (1 - \alpha)^m \tag{6}$$

where:
A – pre-exponential constant;
E – activation energy;
R – universal gas constant;
m – order of the reaction, and

$$\frac{D(\;)}{Dt} = u \frac{\partial(\;)}{\partial x} = \text{ Steady-state one-dimensional substantial derivative.} \tag{6}$$

In Table 2 are listed the kinetic parameters of the resin used on this work.

Table 2. Kinetic parameters of AS4/Epon 9470/537 composite material (Valliappan et al., 1996)

Parameter	Value
Pre-exponential constant, K_0 (s^{-1})	$19.14 \cdot 10^4$
Activation energy, EA (J mol^{-1})	$60.5 \cdot 103$
Heat reaction, ΔH (J g^{-1})	323.7
Order of reaction, n	1.69

3. Solution methodology

For the convective-diffusive problems simulated by Eq. (1) the Péclét number is defined as:

$$Pe = (\rho C_p / k) \; u \; R \quad (7)$$

When the Péclét number is high, the pultrusion process shown in Fig. 1 can be solved using a parabolic model and the Eq. (1) is simplified by neglecting the axial conduction term, as follow:

$$u \; C_p \; \partial T/\partial x - \nabla \cdot \left[k \nabla T \right] = \dot{q} \quad (8)$$

In this parabolic approximation, Eq. (6) and Eq. (8) were discretized in the pultruded bar cross-section (Fig. 2) by using the Galerkin finite element and the axial direction (pulling axis) was discretized by a similar time marching technique.

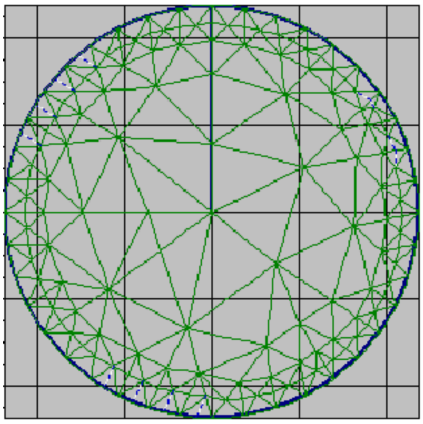


Figure 2. Computational grid at an intermediate process solution.

The heat transfer process and the resin cure kinetics equations in this parabolic approach are solved by a commercial code named PDEase2D. At the present work the Galerkin Finite Element Method of weighted residuals with quadratic basis to convert continuous partial differential equations into discrete nodal equations is used. This method insures highly accurate results and rapid convergence. An adaptive scheme was used with successive mesh refinement in the more intense gradient regions, using an unstructured mesh with triangular elements of six nodes and second-degree interpolation polynomials. The algebraic equations were solved by Conjugated Gradient and Newton-Raphson iterative methods.

The solution in the axial direction is obtained applying a Crank-Nicolson scheme where the cubic term in the Taylor series expansion is determined by a three-step approach. The z-axis step was controlled imposing this cubic term less than an pre-determined error limit.

4. Results

Pantaleão et al. (2000) presented a comparison between the elliptic Taylor-Galerkin and the parabolic results for the pultrusion process of thermosetting composite with circular cross-section. Both methodologies showed a good agreement for the pultrusion pulling speed typical values. Authors also reported a comparison with the experimental data provided by Suratno et al. (1998) and their numerical results as illustrated in Fig. 3a and Fig. 3b for the temperature and degree of cure profiles at the bar cross-section centerline. It was concluded that the parabolic scheme has some advantages because it requires smaller computational processing time and is easier to be implemented than the elliptic model.

Due to these results, at the present work a parabolic approach, Eq.(8), was used to study the effect of different controlling parameters in the pultrusion process of a bar with circular cross-section. The bar diameter was maintained constant for all numerical simulations and the carbon fiber and resin properties were presented in Table 1 and Table. 2.

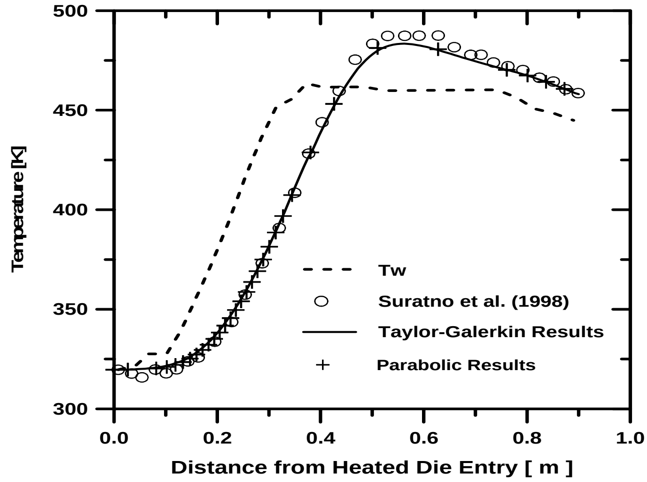


Figure 3a. Results for the axial temperature profile (u = 0.1/60 m/s and FV = 0.7)

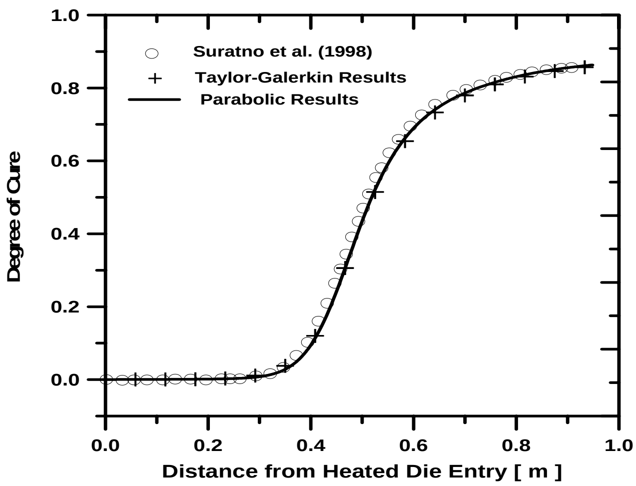
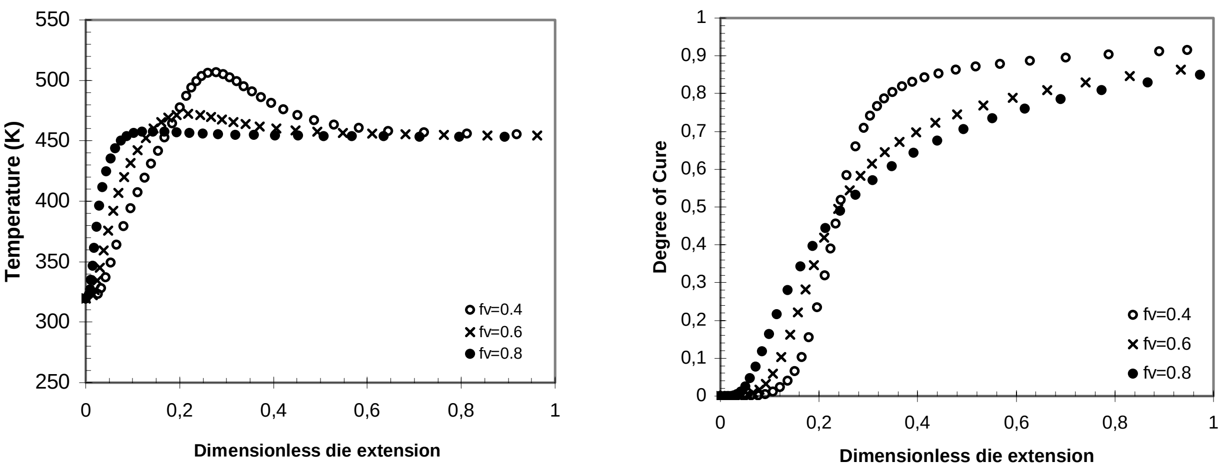


Figure 3b. Results for the axial degree of cure profile (u = 0.1/60 m/s and FV = 0.7)

Figure 4 illustrates the effect of fiber fraction volume (FV) in the temperature and degree of cure profiles at the pultruded bar centerline.

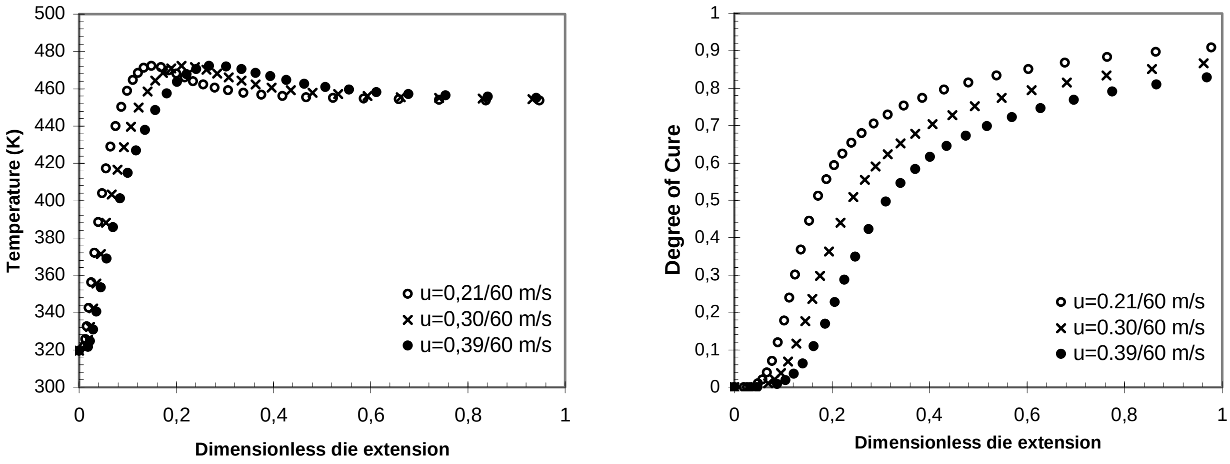


(a) (b)

Figure 04 - Temperature profiles (a) and degree of cure (b) at the die centerline for different fiber fraction volumes.

It is noted that higher FV values reduces the maximum temperature value at the die entry, Fig. 4a. For FV = 0.8, the temperature peak is less intense and smoothed, exceeding the die wall temperature due to resin-cure exothermic reaction. At the die final extension, the FV influence can be neglected. Figure 4b shows that the degree of cure exhibits a different behavior. At the die entry, the composite material has a higher degree of cure (α) for lower FV values. As FV increases, an inversion of this effect occurs, with greater α values at the die exit. Although it appears a good solution to reduce the FV values to enhance the degree of cure rate, this procedure can compromise the mechanical resistance of the thermosetting composite material.

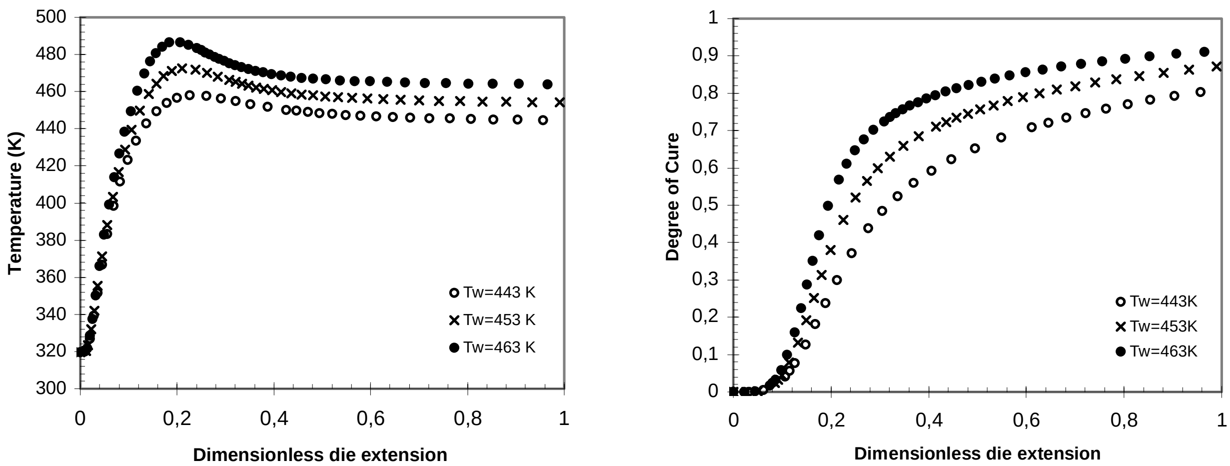
The effect of the pulling speed is indicated in Fig. 05 for three different values. The temperature distribution at the circular bar centerline is shifted to the die entry as the pulling speed (u) decreases, but the maximum values are the same ones. The degree of cure, Fig. 5b, also presents a delay as the u values increases. Therefore, low pulling speeds values (corresponding to greater residence time) reach higher degree of cure values as it was expected.



(a) (b)

Figure 05 - Temperature profiles (a) and degree of cure (b) at centerline for different pulling speeds.

Numerical simulations were carried using three different die-wall set point temperatures (Tw) as shown in Fig. 6a and 6b, for temperature and degree of cure, respectively. As the Tw value increases, the temperature peak also increases, resulting in higher temperature values at the circular bar centerline. The increase in the set point temperature also causes a lower time residence to achieve the same degree of cure level. So, it would be required a larger die extension when lower Tw values are imposed.



(a) (b)

Figure 06 - Temperature profiles (a) and degree of cure (b) at centerline for different set-point temperatures.

4. Conclusions

At the present work a parabolic approximation was employed to numerically simulate the pultrusion process of a composite material composed by epoxy resin reinforced with carbon fiber. The influence of fiber fraction volume (FV), pulling speed (u) and die-wall set-point temperature (Tw) on the temperature and degree of cure was obtained. Results showed that the pulling and the set-point temperature presented opposite effects. So, as the u value increases (or Tw decreases) the degree of cure is reduced. A trade-off solution, analyzing the operational conditions (energy consumption to the pulling and die-heating, for example) can be investigated to achieve a better cost-effective pultrusion process.

5. Acknowledges

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6. References

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