

# **A MODEL FOR TOW IMPREGNATION AND CONSOLIDATION FOR PARTIALLY IMPREGNATED THERMOSET PREPREGS**

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## **ABSTRACT**

The formation and transport of voids in composite materials remains a key research area in composite manufacturing science. Knowledge of how voids, resin, and fiber reinforcement propagate throughout a composite material continuum from green state to cured state during an automated tape layup process is key to minimizing defects induced by void-initiated stress concentrations under applied loads for a wide variety of composite applications. This paper focuses on modeling resin flow in a deforming fiber tow during an automated process of partially impregnated thermoset prepreg composite material tapes. In this work, a tow unit cell based model has been presented that determines the consolidation and impregnation of a thermoset prepreg tape under an input pressure profile. A parametric study has been performed to characterize the behavior of varying tow speed and compaction forces on the degree of consolidation. Results indicate that increased tow consolidation is achieved with slower tow speeds and higher compaction forces although the relationship is not linear. The overall modeling of this project is motivated to address optimization of the “green state” composite properties and processing parameters to reduce or eliminate “cured state” defects, such as porosity and de-lamination. This work is partially funded by the Department of Energy under Award number DE-EE0001367.

## **1. INTRODUCTION AND BACKGROUND**

Knowledge of how voids form and propagate during processing is important for minimizing structural failures in composite materials. Voids initiate stress concentrations when subjected to loads. The overall goal of this research is to develop process models to predict defects (e.g. voids) from leading indicators during on-line consolidation processing of partially impregnated thermoset prepregs. These thermoset prepreg consists of continuous glass or carbon fibers that are partially covered with resins such as epoxies. They have pockets of dry fibers within the prepreg without the resin. The goal during the processing of such materials is to get the resin to cover all the fibers without any dry regions or voids. This is usually done in two steps. The ‘green state’ of the prepreg refers to when the tape is placed on the tool at room temperature introducing partial consolidation of the various layers. Step two is when the entire assembly is

placed in an autoclave or an oven and subjected to heat and pressure to completely consolidate and cure the composite. Figure 1 introduces a schematic of the step one of the on-line consolidation process that is the focus of this paper.

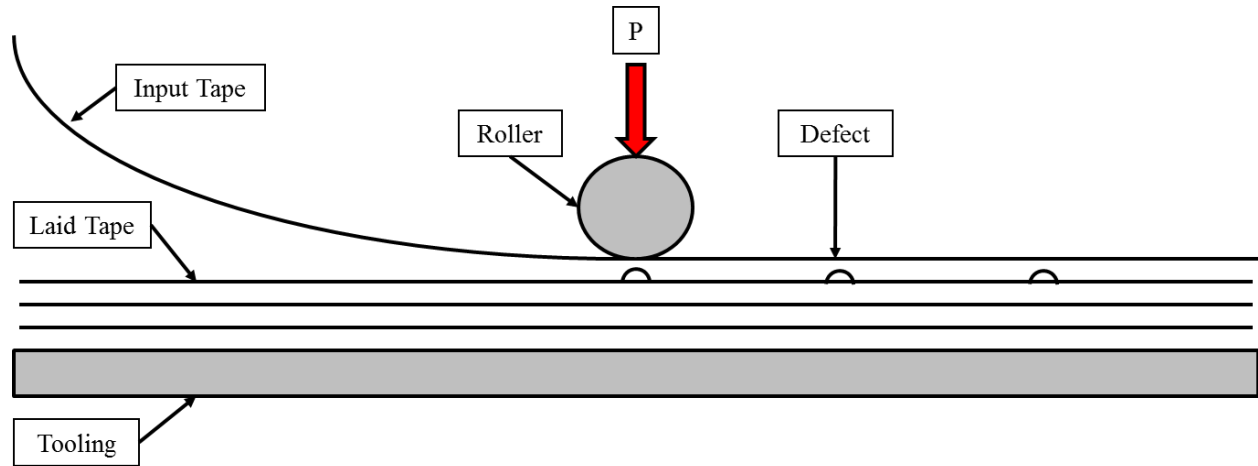


Figure 1. An on-line composite prepreg tape consolidation process schematic.

When a tape is placed over a substrate or on a previously placed tape layer as shown in Figure 1 using a consolidation roller, the resin within the partially impregnated prepreg will redistribute under the roller pressure reducing the regions of fibers uncovered by resin (voids). In this work we will model the resin impregnation as a function of roller pressure, speed and resin viscosity during the green state of the process. Two main phenomena of (i) resin impregnation in fiber tows and (ii) consolidation of fiber tows will be considered. The model will seek to determine the degree of resin impregnation of the tape due to the roller compaction. Presented results are based on sample values representative of typical carbon/epoxy prepreg tape system. This work is based on previous wet thermoplastic tape layup research as described in [1] and expansion of compression resin transfer molding (CRTM) work found in [2].

## 2. METHODOLOGY

### 2.1 Introduction

It is of significant research interest to study the microscale resin impregnation that occurs when composite prepreg tapes are under compaction during processing. In this work, the goal is to determine parameters related to resin penetration into the partially impregnated fiber tow and quantify the remaining unfilled region of fiber tows as a function of resin viscosity, roller pressure and roller speed. This type of analysis is of practical research interest, because partially impregnated composite prepreps are a significant portion of available prepreps on the market and are frequently used for composite applications for their contained optimized net resin content. Figure 2 displays a sample of a partially impregnated unidirectional composite prepreg tape. Of particular interest is the distinction between the resin rich top and bottom surfaces (e.g. “shiny” surfaces of the tape within the figure) versus the fibrous dry center of the tape that must be impregnated during processing and cure.



Figure 2. Partially impregnated composite prepreg tape sample with resin rich top and bottom surfaces (e.g. “shiny” surfaces of the tape within the figure) versus the fibrous dry center.

Parameters that would be relevant for this process are tape thickness, tape displacement, average fiber volume fraction, average through thickness permeability, and compaction time. The model has been simplified and considers a single tow unit cell cross-section of tape that is directly located underneath the roller, with a unit cell consisting of a single dry tow surrounded by a resin shell. Figure 3 displays a schematic of the roller-tape system under consideration here, with a tow unit cell shown. The unit cell approach permits for an adaptable solution for a variety of tape architectures (e.g. woven materials) and boundary conditions (e.g. tape-tape, tape tooling); however, the scope of this work will be limited to the unidirectional tape system.

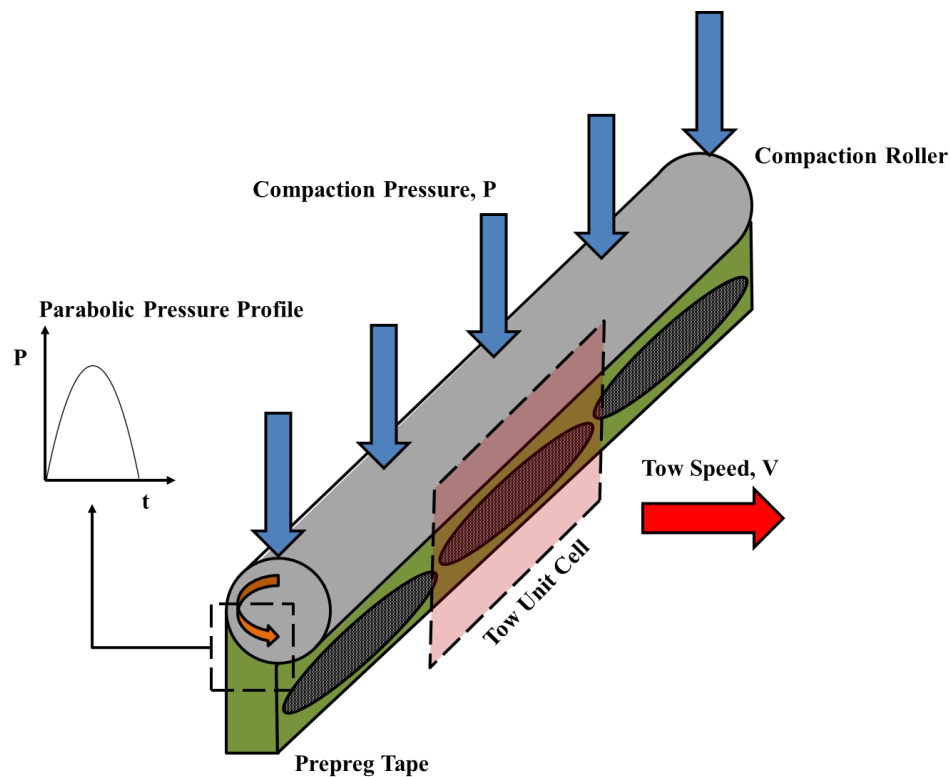


Figure 3. Schematic of a simplified roller – tape model with a tow unit cell.

As shown in Figure 3, the model assumes an ellipsoidal fiber tow unit cell impregnation can be approximated as a 1D rectangular fiber unit cell impregnation. A symmetric solution approach is taken to mitigate the ellipsoidal tow cross section assumption, such that data is calculated with respect to the top or bottom of the tape cross section to the centerline of the tape cross section. In addition, the model accounts for the pressure distribution from the roller by the application of a parabolic pressure profile as a function of time. This model is based on process modeling research conducted within compression resin transfer molding (CRTM) as described in [2]. Figure 4 displays a modeling schematic for the tape impregnation process model based on the single unit cell. The CRTM process model in [2] is based on three processing phases that can be adapted for the composite tape placement processes: Phase 1: A given amount of resin injected into a partially closed mold containing a dry fiber preform which in our process is the resin that is layered on top of the dry tape (fibers) prior to rolling; Phase 2: The closure of the mold until it is in contact with the fiber preform displacing all the resin into the preform, which in our process is the initial roller compaction; Phase 3: Further mold closure to the desired thickness of the part compacting the fiber preform and redistributing the resin, which in our process is the final roller compaction. These processing phases are established within the context of the tape placement processing described in this work.

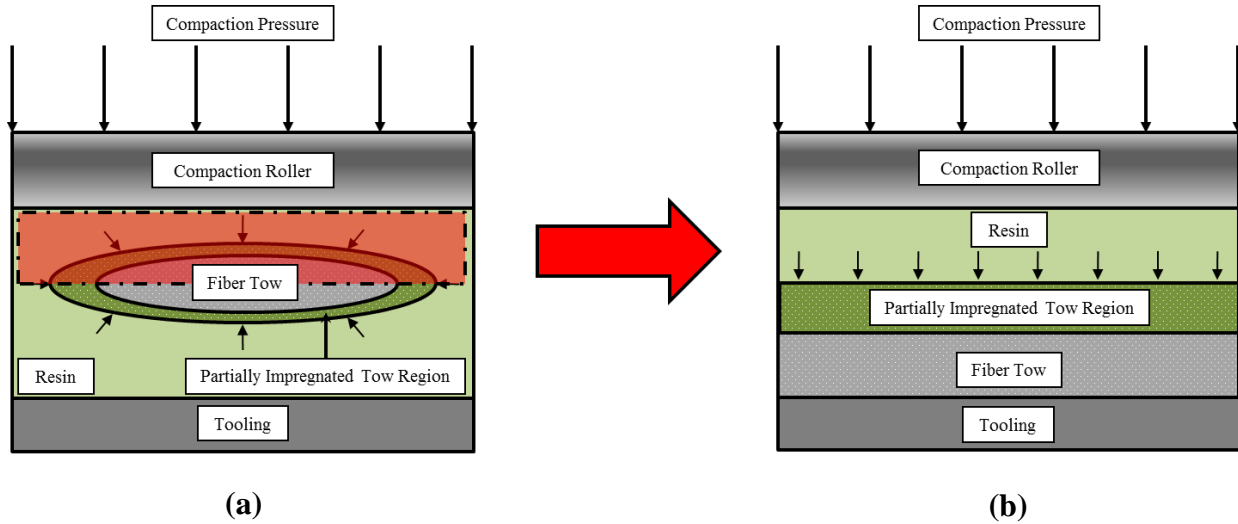


Figure 4. Schematic and simplification of tape impregnation unit cell schematic: (a) original fiber tow unit cell model; (b) modified unit cell model.

### 3. THEORY

#### 3.1 Assumptions and Simplifications

Many assumptions are made to simplify the modeling of the tape impregnation process. The tape deformation during phase 1 is neglected. Also, a linear pressure gradient assumed in region of the tow impregnated with the resin before the initiation of phase 2. The magnitude of this initial pressure is small and the depth of penetration is only a fraction of the initial preform thickness so linear pressure assumption is justifiable. The flow is modeled as one-dimensional flow through thickness. This is a reasonable assumption as the resin covers the entire surface of the tape on the outside as seen from Figure 2 and the roller moves in the thickness direction during compaction.

The dry part of the preform is assumed to be stiff, due to the high tow speeds involved. In addition, the lubricated part of the impregnated tow visco-elasticity will be neglected. This is because the fiber volume fraction is assumed to be dependent only on the applied stress (applied to preform deformation). The following constitutive equation was cited by [[3] to be suitable for describing the relationship between applied stress and fiber volume fraction,

$$v_f = v_{fo} + (v_{fmax} - v_{fo}) \tanh^n \left( \frac{p_{pref}}{p_{prefmax}m} \right) \quad [1]$$

where  $v_f$  is the fiber volume fraction,  $v_{fo}$  is initial fiber volume fraction,  $v_{fmax}$  is the maximum fiber volume fraction,  $p_{pref}$  is the applied tow pressure (e.g. compaction stress exhibited by the fiber tow),  $p_{prefmax}$  is the maximum applied tow pressure, and  $m, n$  are empirical fitting parameters. Figure 5 displays sample compaction pressure curves based on equation [[1].

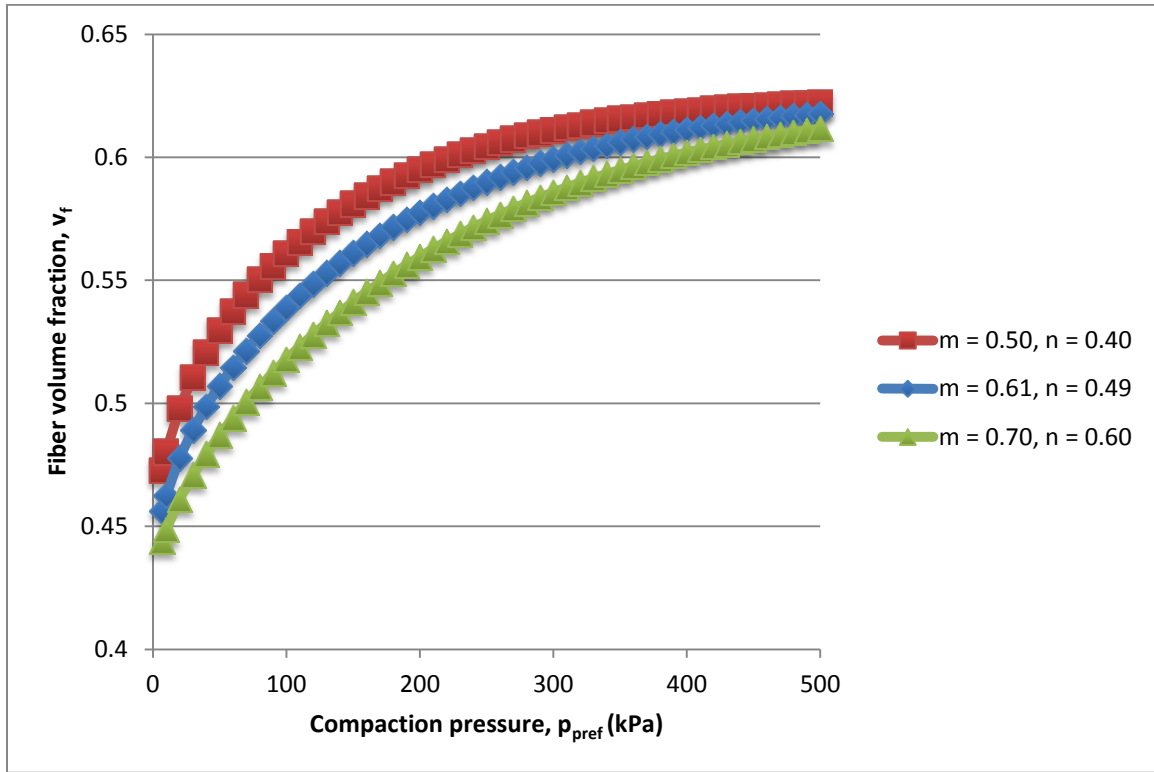


Figure 5. Compaction pressure curves based on equation [[1].

Based on initial experimental work, it was found that the ideal empirical fitting parameters are at  $m = 0.61$  and  $n = 0.49$  to correlate with experimental results relating preform deformation and fiber volume fraction to applied pressure. Values were taken here with  $v_{fo} = 0.425$ ,  $v_{fmax} = 0.625$ , and  $P_{prefmax} = 500$  kPa. In addition, the Kozeny-Carman constitutive equation is assumed to describe the relationship of through thickness fiber permeability change with the fiber volume fraction of the tape as cited in [[2],[4]],

$$K_{zz}(v_f) = ko \frac{(1 - v_f)^3}{v_f^2} \quad [2]$$

where  $K_{zz}$  is through thickness (e.g. z-direction) permeability,  $v_f$  is the fiber volume fraction, and  $ko$  is the permeability coefficient. Here  $ko = 5.8\text{E-}13 \text{ m}^2$ . The resin flow is assumed to follow Darcy's Law that describes the relationship between the flow rate and the pressure drop. Since the Reynolds number,  $Re \ll 1$ , the flow can be considered "quasi-steady" and the simulation can solve the steady state problem at each time step.

### 3.2 Governing Equations

The primary physics that can be used to describe the resin flow through the porous tape unit cell region that is used within this work is Darcy's Law coupled with mass conservation of fluid elements under compaction. As noted in [2], once can find that at each time step,

$$\nabla \left( -\frac{\mathbf{K}}{\eta} \cdot \nabla p \right) = -\frac{\partial \epsilon}{\partial t} \quad [3]$$

where  $\epsilon$  is the volumetric strain,  $\mathbf{K}$  is the tow fabric permeability,  $\eta$  is the resin viscosity,  $t$  is the process time, and  $p$  is the resin fluid pressure. Note it is assumed that the dry regions of the tow are much stiffer than the wetted regions of the tow and thereby do not influence the resin flow under the roller compaction force. Also, one can relate the fluid resin pressure ( $p$ ) to the applied compaction pressure from the roller ( $p_{ap}$ ) and the experienced compaction stress on the tow ( $p_{pref}$ ) using the Terzaghi relation as found in [1].

$$p_{pref} = p_{ap} - p \quad [4]$$

To simplify equation [3], once can rewrite the volumetric strain rate on the right hand side as follows,

$$\frac{\partial \epsilon}{\partial t} = \frac{\left( \frac{\partial \Delta V}{\partial t} \right)}{\Delta V} \quad [5]$$

with  $\Delta V$  as an elementary volume. Since this model accounts for one-dimensional deformation of the tow elements, one can express the right hand side of equation [5] with the following,

$$\frac{\left( \frac{\partial \Delta V}{\partial t} \right)}{\Delta V} = \frac{\frac{\partial h}{\partial t}}{h_0} \quad [6]$$

where  $h$  is the tow element thickness and  $h_0$  is the original thickness. Note that equation [6] serves as a linearized strain description of the tow. Since the volume of reinforcement within the tow is constant, the thickness change can be written with respect to fiber volume fraction.

$$h = \frac{h_0 v_{fo}}{v_f} \quad [7]$$

With this relationship, equation [5] can be written as the following result.

$$\frac{\partial \epsilon}{\partial t} = -\frac{v_{fo}}{v_f^2} \frac{\partial v_f}{\partial t} \quad [8]$$

Note one can rewrite the derivative of the fiber volume of fraction be a function of compaction pressure by using the chain rule.

$$\frac{\partial v_f}{\partial t} = \frac{\partial v_f}{\partial p_{pref}} \frac{\partial p_{pref}}{\partial t} \quad [9]$$

With this relationship, one can rewrite equation [8] as the following result.

$$\frac{\partial \epsilon}{\partial t} = -\frac{v_{fo}}{v_f^2} \frac{\partial v_f}{\partial p_{pref}} \frac{\partial p_{pref}}{\partial t} \quad [10]$$

Using a similar chain rule approach, one can arrive at an expression for the change in the tow permeability in the through thickness direction with relation to tow fiber volume fraction and compaction pressure.

$$\frac{\partial K_{zz}}{\partial z} = \frac{\partial K_{zz}}{\partial v_f} \frac{\partial v_f}{\partial p_{pref}} \frac{\partial p_{pref}}{\partial t} \quad [11]$$

Finally, for one dimensional through thickness resin flow (e.g. z-direction) in the fiber tow driven by the roller compaction pressure, one can rewrite equation [3] using equations [10] and [11] to arrive at the following governing equation.

$$\frac{1}{\eta} \frac{\partial K_{zz}}{\partial v_f} \frac{\partial v_f}{\partial p_{pref}} \frac{\partial p_{pref}}{\partial z} \frac{\partial p}{\partial z} + \frac{K_{zz}}{\eta} \frac{\partial^2 p}{\partial z^2} = -\frac{v_{fo}}{v_f^2} \frac{\partial v_f}{\partial p_{pref}} \frac{\partial p_{pref}}{\partial t} \quad [12]$$

Equation [12] is a modified version of conventional Darcy's law that yields the resin pressure throughout the thickness of the tow as a function of fiber volume fraction, permeability, thickness, and time. To calculate the resin flow front progression with time through the depth of the tow, an expression can be determined based on the averaged velocity via Darcy's Law at the flow front position,

$$\frac{\partial L}{\partial t} = -\frac{(K_{zz})_{z=L}}{\eta \phi_{z=L}} \frac{\partial p}{\partial z_{z=L}} \quad [13]$$

where L is the flow front position or the resin penetration depth into the tow and  $\phi$  is the porosity of the tow defined as  $\phi = (1 - v_f)$ . Note the subscript  $z=L$  implies values at the flow front position. Equations [12] and [13] are thus the two primary equations that the model solves to describe the physics of the tow unit cell roller compaction.

### 3.3 Non-Dimensional Analysis and Numerical Solution

To solve the nonlinear governing equations, both equations [12] and [13] are solved numerically throughout the preform thickness and process time domains to generate the state of the tow after compaction, as detailed in [2]. The model generates information related to compaction of the tow unit cell and the dry tow space impregnated given a particular pressure profile. This allows one to perform parametric studies to evaluate the optimal processing parameters (e.g. compaction pressure profiles, tow speeds, etc.) to ensure nominal consolidation and impregnation of the tape during roller processing. To aid in this type of analysis, the governing equations have been adapted into a non-dimensional form. For this analysis, one can make the following non-dimensionalizations of dependent and independent variables of the governing equations,

$$\hat{L} = \frac{L}{H} \quad [14]$$

$$\hat{K}_{zz} = \frac{K_{zz}}{k_0} \quad [15]$$

$$\hat{z} = \frac{z}{H} \quad [16]$$

$$\hat{t} = \frac{t}{t_c} \quad [17]$$

$$\hat{p} = 1 - \hat{p}_{pref} = \frac{p}{p_{ap}} \quad [18]$$

With  $H$  as the original tow thickness,  $t_c$  as the characteristic time, and other variables defined previously. With these non-dimensionalized variables, one can rewrite equation [12] in non-dimensional form.

$$\frac{k_0 p_{ap} t_c}{v_{fo} \eta H^2} v_f^2 \left( -\frac{\partial \hat{K}_{zz}}{\partial v_f} \frac{\partial v_f}{\partial \hat{p}_{pref}} \left( \frac{\partial \hat{p}}{\partial \hat{z}} \right)^2 + \hat{K}_{zz} \frac{\partial^2 \hat{p}}{\partial \hat{z}^2} \right) = \frac{\partial \hat{p}}{\partial \hat{t}} \frac{\partial v_f}{\partial \hat{p}_{pref}} \quad [19]$$

Note one can choose a value of  $t_c$  such that the coefficient of the left hand side of the previous equation is made unity. With that, one can write  $t_c$  as the following result.

$$t_c = \frac{v_{fo} \eta H^2}{k_0 p_{ap}} \quad [20]$$

One can now rewrite equation [12] into a simplified non-dimensional form using equations [4], [19] and [20],

$$v_f^2 \left( -\hat{K}'_{zz} \left( \frac{\partial \hat{p}}{\partial \hat{z}} \right)^2 + \hat{K}_{zz} \frac{\partial^2 \hat{p}}{\partial \hat{z}^2} \right) = \frac{\partial \hat{p}}{\partial \hat{t}} \quad [21]$$

with the following definitions:



$$\hat{K}'_{zz} = \frac{\partial \hat{K}_{zz}}{\partial v_f} = \frac{-3(1 - v_f)^2 v_f - 2(1 - v_f)^3}{v_f^3} \quad [22]$$

$$v'_f = \frac{\partial v_f}{\partial \hat{p}_{pref}} = \frac{(v_{fmax} - v_{f0})np_{ap}}{mp_{max}} \left( 1 + \tanh^2 \left( \frac{\hat{p}_{pref} p_{ap}}{mp_{max}} \right) \right) \left( \tanh \left( \frac{\hat{p}_{pref} p_{ap}}{mp_{max}} \right) \right)^{n-1} \quad [23]$$

Substitutions of the non-dimensionalized variables found in equations [14]-[18] coupled with equations [20] can yield a non-dimensionalized version of equation [13] to solve for the flow front position.

$$\frac{\partial \hat{L}}{\partial \hat{t}} = - \frac{(\hat{K}_{zz})_{z=L}}{\phi_{z=L}} \frac{\partial \hat{p}}{\partial \hat{z}_{z=L}} \quad [24]$$

The solver iterates with time and for each time step solves equation [21] for the resin pressure throughout the thickness domain of the tow. Note one can treat the tow impregnation problem as a steady state problem at each time step, since the problem is considered “quasi-steady” as mentioned previously. When this resin pressure is found, equation [24] advances the resin flow front with the next time step with the generated pressure information from the previous time step. For every time step increment, a new thickness mesh is generated with modified mesh spacing to reflect the change in tow deformation from the application of the compaction roller. This process of iterating and solving both equations for pressure and flow front position continues until the end of the process time is reached. Details about the numerical analysis execution can be found in [2].

Unique to this work is the ability to supply arbitrary pressure profiles as functions of time. Previous work [2] has been limited to the application of constant pressure during processing for this type of analysis. Here, the roller tends to apply pressure in a parabolic or sinusoidal manner onto a given section of tape. Later results are based on a sinusoidal pressure profile of the following form.

$$P(t) = P_0 \sin(\omega t) = P_0 \sin \left( \frac{V}{R} t \right) \quad [25]$$

Here,  $P_0$  is the initial compaction pressure,  $\omega$  is the angular velocity of the roller,  $V$  is the tow speed of the roller, and  $R$  is the radius of the roller. Depending on the time scales involved, one may apply desired pressure profile to attain more accurate results. For example, high tow speed processing (e.g.  $V = 0.15$  m/s) may be able to attain realistic results with the application of a constant pressure for a short time window that a section of tape is under the roller versus a parabolic pressure profile that may be more accurate for slower processing tow speeds and times (e.g.  $V = 0.015$  m/s). The results that are generated and are of engineering interest from this analysis are the net displacement of the tow in the thickness direction, the wetted tow region distance through the tow thickness, the dry region distance through the tow thickness, the wetted and dry tow region fiber volume fraction and through thickness permeability. These results can be used to predict if processing conditions are optimum to ensure minimized defects by reducing the dry tow region sufficiently to eventually impregnate the remaining region during vacuum compaction and curing.

## 4. RESULTS

The presented model was run under sample parameters to evaluate its viability for realistic tape system geometries. Figure 6 shows a tape unit cell geometry schematic that serves as a sample representative tape cross section. Note values presented here are arbitrary but representative of sample tape placement system geometries. The schematic highlights an initial dry tow or void region characterized by the initial height,  $a_i$  and an initial overall tape height,  $h_i$ . It is important to note that the model is initialized with  $L_i$ , which is the initial impregnation depth into the dry tow. For this work,  $L_i$  is takes to be 1% of  $h_i$ .

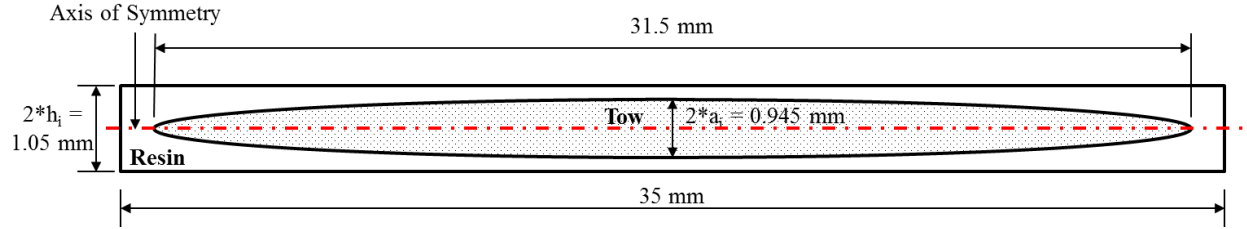


Figure 6. Tow unit cell initial sample geometry schematic.

Figure 7 and Figure 8 display results of a parametric study performed with the presented model and the sample geometric inputs from Figure 6. The goal of the parametric study is to evaluate the degree of compaction and impregnation of the tow unit cell after the roller compaction cycle is completed. Here, the percentage reduction of the dry fiber region and percentage tow compaction results are presented with respect to tow speed for different cases of initial compaction force. The results are presented in a non-dimensionalized manner for base comparisons between cases, with the void size non-dimensionalized with respect to the initial void size ( $a_i$ ) and the tape size non-dimensionalized with respect to the initial tape height ( $h_i$ ). Note, compaction forces are reported using their initial value of  $P_0$  but are applied in a sinusoidal manner as shown in equation [25]. Non-dimensionalized compaction time is taken with respect to the final compaction time,  $t_f$ .

The results show similar trends for both the percentage reduction of the dry fiber region and percentage tow compaction. The results indicate that as tow speed increases, the degree of compaction is reduced. This is due to the fact that a faster tow speed leads to a shorter time for consolidation and resin impregnation, thus leading to a lower degree of compaction after completion of the roller compaction cycle. This model allows for one to evaluate how tow speed can influence the degree of compaction and impregnation. Impregnation effects are characterized by reduction of the dry fiber region due to resin impregnation and fiber compaction. In addition, the model shows that by increasing the initial compaction force, one can achieve improved compaction overall. It is important to note that results become linear for higher tow speeds, but deviate non-linearly as tow speed decreases, with a threshold speed of approximately 0.1 m/s for this parametric study.

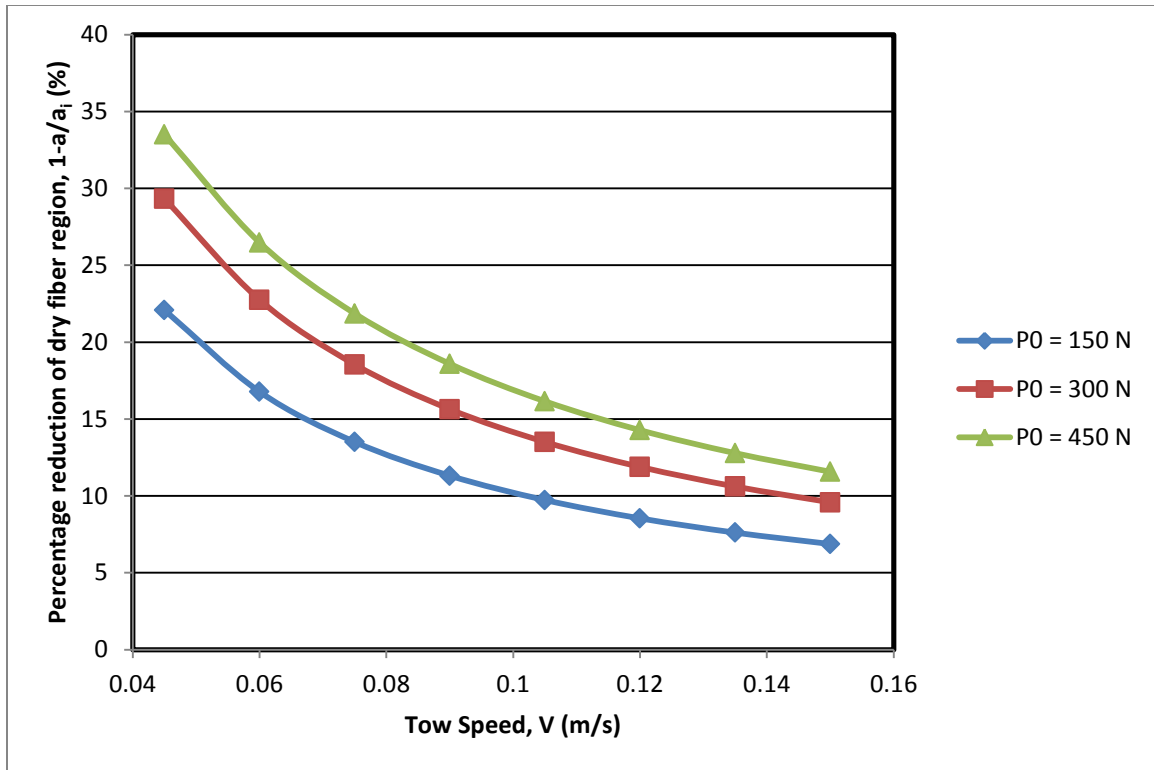


Figure 7. Percentage reduction of dry fiber region at  $t/t_f = 1$ .

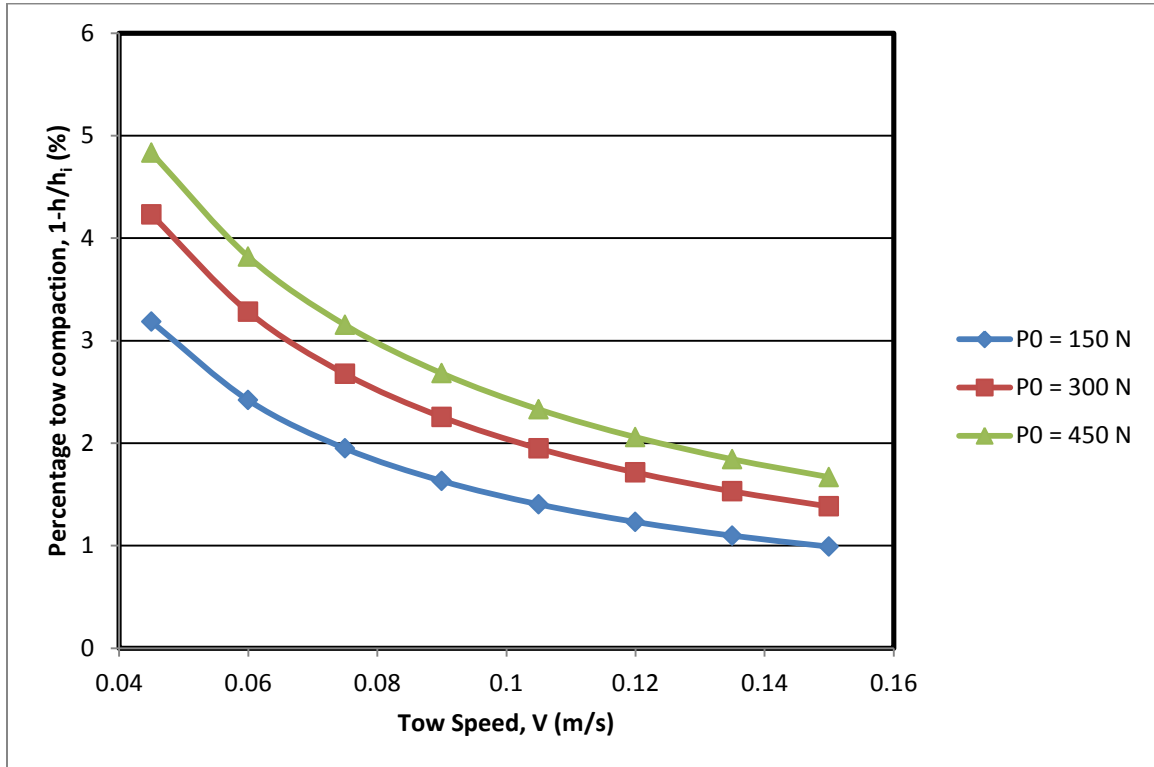


Figure 8. Percentage tow compaction at  $t/t_f = 1$ .

This threshold of linearity could be important for process engineering, as compaction and impregnation behaviors in the non-linear threshold of tow speed and compaction force may experience characteristic non-linear behaviors (e.g. visco-elasticity) when compared to processing at higher tow speeds within the linear region. Of further interest is that the effects of improved dry fiber and tow compaction with increased initial compaction force are reduced as one processes at higher tow speeds. This is characterized by the decreased spacing in between the different initial compaction force plots as tow speed is increased. This is important process as increase in compaction force by the roller may yield only marginal improvements in tape compaction and impregnation. The model allows one to explore the optimization of processing parameters to achieve the desired tape consolidation and impregnation state. Further parametric studies that can be performed are how changes in processing viscosity and fiber tow stiffness (e.g.  $m, n$  parameters from equation [[1]]) are related to tow consolidation and impregnation as functions of compaction force and tow speed.

## 5. CONCLUSIONS

A process model has been presented that predicts the consolidation, impregnation, and tape dimensions of a partially impregnated thermoset prepreg tape during roller compaction. The model considers geometric, material, and process parameters with the goal of exploring the influence of process parameters on the final consolidated composite before curing. A parametric study has been performed to explore how changes in compaction force and tow speed impact the compaction and impregnation of the partially impregnated composite prepreg tape. The model is found to predict by how much a tow is consolidated and impregnated given an input pressure profile (e.g. sinusoidal pressure profile). Experimental verification and validation is necessary to ensure model outputs are consistent with real-life observations. The model will be compared to thermoset prepreg on-line consolidation processes to ensure validity. Future work will seek to compare model accuracy and modifications will be made to optimize the model for thermoset composite material systems. Current work is exploring the implementation this model for not only roller processing but also vacuum bag compaction with a temperature ramp for the curing cycle.

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