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A MODEL FOR THERMOPLASTIC MELT IMPREGNATION OF FIBER BUNDLES DURING CONSOLIDATION OF POWDER-IMPREGNATED CONTINUOUS FIBER COMPOSITES

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SUMMARY: Continuous fiber thermoplastic matrix composites are fabricated using a novel powder-impregnation process that combines vacuum assisted resin transfer molding (VARTM) with compression molding. The resulting composite has an average fiber volume fraction of 65%. A model has been developed for the consolidation phase during compression molding to predict the void fraction of the resulting composite. This model takes into account the fabric unit cell dimensions and material properties and assumes that the tow permeability remains constant. The model is compared to experimental values for void fraction for samples prepared using a range of consolidation pressures and dwell times.

KEYWORDS: thermoplastic matrix composites, particle deposition, compression molding, Vacuum Assisted Resin Transfer Molding (VARTM)

INTRODUCTION

Currently, there are many methods used to manufacture composites with a thermoplastic matrix. For applications that require continuous fiber reinforcement most methods usually consolidate a precursor material at elevated temperature and pressure in an autoclave or a heated hydraulic press. The precursor material used could be in the form commingled fabrics, pre-impregnated tapes, or thin films of resin in between layers of dry fibers. Commingled bundles combine reinforcing fibers (glass, carbon, aramid) in the same bundle as a matrix material (i.e. nylon, polypropylene, polyethylene, etc.) These bundles can then be knit, woven, or stitched to form the commingled fabric. Pre-impregnated tape consists of fabrics or collimated fibers pre-impregnated with resin. There are also some less traditional methods of manufacturing thermoplastic matrix

composites. These include melt impregnation and slurry deposition. In one melt impregnation process roving bundles are passed over a pin submerged in a polymer melt under tension to spread the bundle and impregnate it [1]. Slurry deposition processes can be generally classified two ways- wet and dry. Vodermeyer et. al. developed a version of wet particle deposition. They introduced particles in a continuous fiber preform by running bundles over a spreader and through an aqueous bath containing thermoplastic powder prior to forming the final preform [2]. Ye et al. made fiber preforms from bundles that were constructed by combining powder and fibers inside a thermoplastic sheath [3]. In an example of dry deposition, forced air is used to deposit thermoplastic particles onto the fiber bundle. Then the polymer/fiber combination is passed briefly through a furnace to soften the matrix material and adhere it to the bundles. Finally the powder-impregnated bundle is cooled and wound onto a spool and used as a precursor material [4]. The majority of the methods available today to manufacture thermoplastic matrix composite materials involve a great deal of labor to manufacture thick section structures. The process described in this paper can also be used with three dimensional or thick fabrics and is an attractive feature of this proposed manufacturing process.

MANUFACTURING PROCESS

The proposed manufacturing technique is a two step process. It uses a modified vacuum assisted resin transfer molding (VARTM) method in the first step to impregnate the fabrics with the thermoplastic powder. Then the matrix material is consolidated with the fabric by using the traditional compression molding or autoclave in the second step.

During the first step, the reinforcement fabric is placed on a rigid mold tooling surface surrounded by a rubber gasket. The gasket has a thickness equal to that of the fabric plus an additional height to leave a gap between the top plate of the mold and the fabric. There are vacuum vents in the bottom plate and an inlet for injection in the top plate. Once the fabric and gasket are in place and the mold is closed, a vacuum is applied. Once full vacuum is achieved, the inlet for injection is opened. The gap between the top of the fabric and the mold is flooded with the water/plastic powder slurry. The fabric is infiltrated with the water and powder mixture through the thickness due to the pressure gradient present across the fabric. A filter layer placed beneath the material layers helps build up a concentration of powder in the fabric because it allows only water to escape through the vacuum vents. After the total volume of the mixture has been infiltrated through the fabric, the impregnated fabric is removed from the mold and placed in an oven at low temperature to evaporate the water. At this point the macropores in the fabric between the bundles are filled with the powder while the micropores within the fiber bundles are completely dry. Fig. 1 is a schematic of the glass-fabric powder-impregnation process.

During the second step of the process, the powder-impregnated fabric is placed in a rigid aluminum plate and frame mold for compression molding. Fig. 2 shows an example autoclave recipe that is divided into four phases and can be used for consolidation. In Phase 1, heat is applied to melt the powder while the mold is held at atmospheric pressure. Phase 2 starts once the powder is in the melt state and the pressure on the assembly is increased to facilitate the impregnation of the fiber bundles with the molten resin. During Phase 3 the temperature and pressure held constant to allow additional dwell time for saturation of fiber tows and consolidation of the composite. Finally, in Phase 4, the pressure is held constant while the

composite is cooled, solidifying the resin in place. The rigid mold is necessary for this step to ensure that the molten thermoplastic will not flow out of the sides of the fabric layers and instead be forced to flow into the fiber bundles.

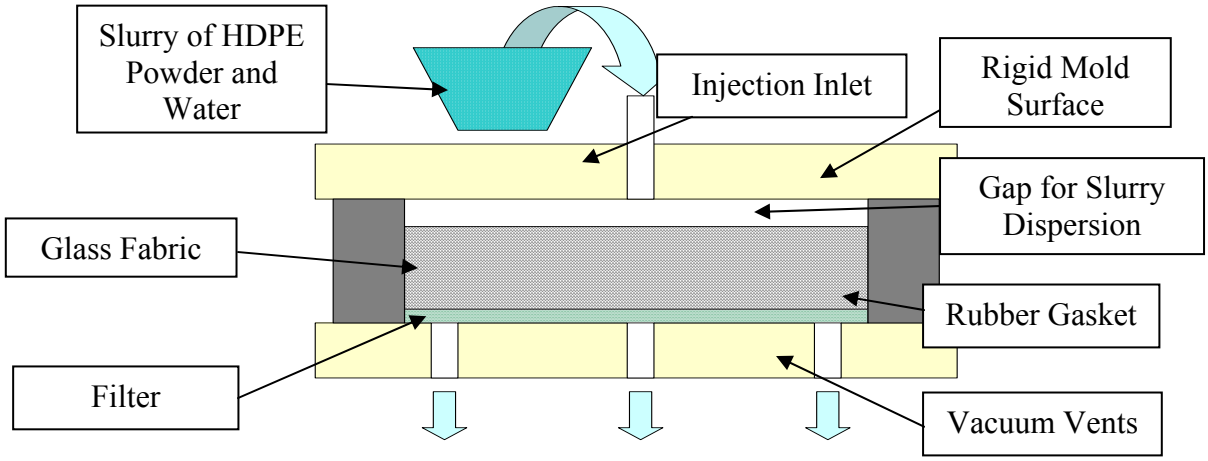

Fig. 1 Diagram of powder impregnation mold.

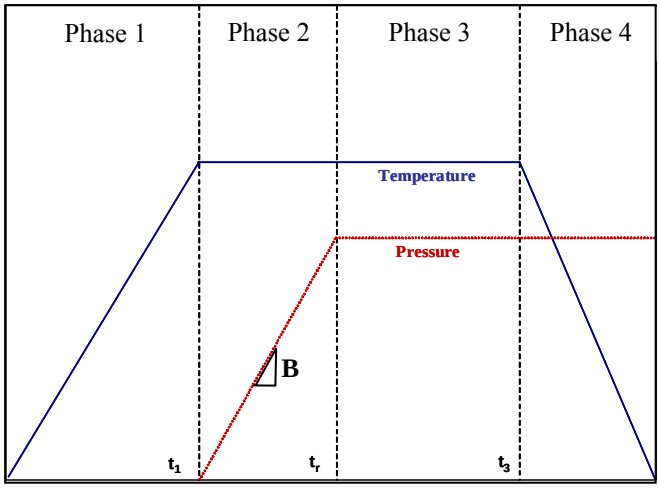

Fig. 2 Time, temperature, and pressure methodology used in the autoclave to consolidate the powder-impregnated composite.

MATERIAL SELECTION

It is easy to build thickness with only a few layers of material with this two-step wet particle deposition process. In order for this process to work effectively, the matrix material used should be available in a powder form. Many thermoplastics such as polypropylene (PP), high density polyethylene (HDPE), and polyetheretherketone (PEEK) are already available in the powder form. The surface of the powder must be treated to increase wettability and dispersion in the carrying solvent which is usually water. This allows the mixture of water and powder to create

slurry of low viscosity, which can then be injected into the fabric preform. The matrix material used in the experiments is manufactured by Inhance®/Fluoro-Seal Ltd. It is an HDPE powder with an average particle diameter of 18 microns. Materials eligible for this process include polyolefins (i.e. HDPE, PP) and some engineering thermoplastics (i.e., PEEK).

Both 2D and 3D fabrics can be used as reinforcements however only a 2-D fabric is used for the work presented in this paper. The fabric used is a 3637 g/m^2 (108 oz/yd^2) biaxial glass fiber fabric manufactured by Vectorply™ Corporation. It is a non woven material and the fiber bundles are held in place by a network of polyester stitching with empty channels in between and parallel to the fiber bundles. In traditional LCM processes with thermosetting resins the channels would function as a pathway for resin to racetrack and impregnate the preform [5]. For the proposed process, these channels serve as a collection area for the matrix powder that will later be melted and forced into the neighboring fiber bundles in the second step of compression molding or autoclave processing. Any fabric that is used in this manufacturing process must have sufficient free space around the fiber bundle network to accumulate the powder so that once the fabric is compressed both the compressed macropores and micropores within in the fiber bundles will be filled with the matrix to the desired degree.

EXPERIMENTS

Experiments were conducted with the materials selected in which the step one of infiltrating the preform was identical. However different pressures and dwell times were used during consolidation in the autoclave. The experiments for step 2 used the recipe shown in Fig. 3. The five set of experiments consisted of a maximum pressure of 0.345 MPa with dwell times of 30 and 60 minutes respectively, maximum pressure of 0.690 MPa with dwell times of 30 and 60 minutes respectively and a maximum pressure of 1.379 MPa with dwell time of 30 minutes. To measure the void content at the end of the process, the sample from the fabricated composites can be sectioned and polished for examination under an optical microscope or scanning electron microscope (SEM). The images captured with the microscopes can be characterized to quantify the void content.

MODEL

Development of a process model will allow us to identify important material, geometric and process parameters and their influence on the manufacturing of the composite by the proposed process. This will require modeling of both steps- particle deposition and fiber bundle impregnation. This paper will focus on the mechanics of fiber bundle impregnation (micro-impregnation) with a highly viscous polymer melt upon application of pressure during consolidation (Phase 2 and 3 of step two). The effect of pressure, time, and temperature on the microstructure of the final composite form will be determined. Though it is desirable in most cases to have a fully impregnated laminate, at times a specific amount of porosity could be desirable [6]. The model will also be useful to determine optimal combination of time, temperature and pressure to obtain the desired porosity.

In this paper as we will model only the second step of the process at the unit cell level and assume that due to the repetitive nature of the preform one would expect a similar physics

throughout the preform. We will assume that initially all resin is in the macropores between the fiber tows and none has impregnated inside the fiber tows. The initial conditions will also require one to provide the resin content in the macropores and the geometry of the macropore and the fiber tow. The preform consists of 6 layers of unidirectional fiber- 3 in the 0° (or warp) direction and 3 in the 90° (or weft) direction. Each layer consists of two tows and one tow gap. The dimensions of the tows and gaps are found by examining the fabric under an optical microscope and taking measurements from the micrographs. An example of one is shown in Fig. 3**.**

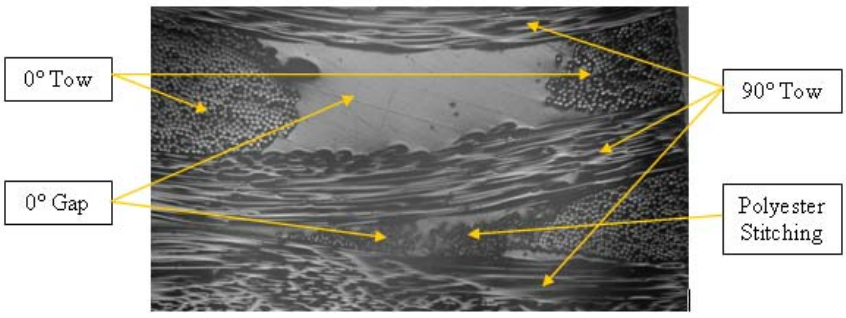

Fig. 3 Optical micrograph of 2D fabric used to measure elements of unit cell.

The left-hand side of Fig.4 shows how the idealized tows are arranged in the preform. Recall that it is assumed that during the power impregnation process the tow gaps are entirely filled with the HDPE powder. This assumption can be relaxed after the step 1 of this process is modeled. The unit cell for this preform can be created by taking advantage of the repetitive nature of the preform and the symmetry present. Thus the unit cell will consists of only one half of a 0° tow gap and one half of a 0° tow as shown in Fig. 4. The grey rectangle represents the tow gap while the orange rectangle filled with white circles (fibers) represents the tow. The tow gap, H_g, has an initial value of H_{g0} though this decreases as the resin flows into the tow between the fibers.

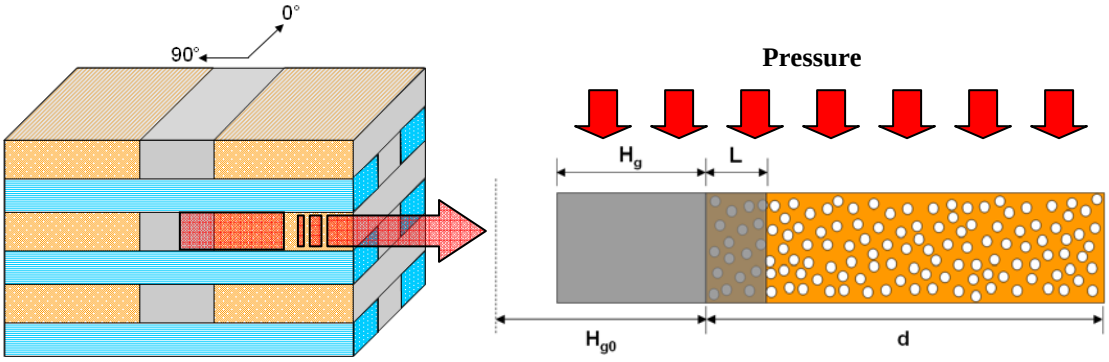

Fig. 4 Unit cell for the model- 1/2 Tow Gap and 1/2 Tow from the Idealized Preform Geometry.

Analysis

As simplified in Fig. 4, one can consider the flow to be one dimensional in which the resin flows from the macropore into the fiber tows. In reality, the macropore shrinks under the compression pressure as the resin impregnates the fiber tows. In our model we assume that as the resin leaves

the macropore, the volume of macropore reduces accordingly and hence there will be no voids remaining in the macropores.

One can use Darcy's Law to describe the relationship between the average fluid velocity and the resin impregnation into the fiber tow in one dimension as follows.

$$\bar{u} = \frac{K}{\mu}\frac{P_r}{L} = \phi\frac{dL}{dt} \tag{1}$$

Here K is fiber tow permeability across the fibers, μ is the viscosity of the resin at the hold temperature, ϕ is the porosity of the fiber tow, P_r is the resin pressure in the macropore and L is the location of the resin front inside the tow at a given instant in time. In step 2 the applied pressure P_a, is balanced by the stress taken on by the fiber network and resin pressure [7]. However, in this case we are not accounting for tow deformation, thus eliminating the fiber stress term, σ_f, and assuming that the applied pressure is balanced entirely by resin pressure. Finally, the expression for resin pressure, P_r:

$$P_r = \frac{P_a}{v_r} \tag{2}$$

where v_r is the resin volume fraction. Using Eqns. (1) and (2) we obtain the following ordinary differential equation that describes the impregnation of the resin into the fiber tow with time:

$$\frac{dL}{dt} = \frac{K}{\mu\phi}\frac{P_a}{v_r}\frac{1}{L} \tag{3}$$

Now we need to derive an expression for v_r in terms of the geometry of the tow, the tow gap and porosity. The volume of each phase present in the system – resin (V_r), fiber (V_f), and void (V_v) - at any time is as follows: $V_r=H_g+\varphi L$, $V_f=d(1-\varphi)$, and $V_v=(d-L)\varphi$. H_g, d, and L are shown in Fig. 4. The volume fraction of resin can be derived using the equations for V_r, V_f, and V_v:

$$v_r = \frac{V_r}{V_r + V_f + V_v} = \frac{H_g + \phi L}{H_g + d} \tag{4}$$

assuming there is not much change in the width of the tows. Finally, substituting this expression back into Eqn. (3) we find the differential equation for flow front velocity:

$$\frac{dL}{dt} = \frac{K}{\mu\phi}\frac{P_a}{L}\left(\frac{H_g + d}{H_g + L\phi}\right) \tag{5}$$

It is assumed that the permeability, K, remains constant throughout consolidation. The gap size, H_g, changes with respect to time but the *volume* of resin will always remain a constant. When L is equal to 0, H_g is equal to the initial dimension of the tow gap H_{g0}. We find a relation between resin front position within the tow, L_r, at the end of Phase 2 when the time is t_r.

$$\frac{H_{g0}(H_{g0}+d)}{\phi^2}\ln\left\{\frac{H_{g0}+d}{H_{g0}+d-L_r\phi}\right\} - \frac{H_{g0}}{\phi}L_r = \frac{KB}{\mu\phi}\frac{t_r^2}{2} \tag{6}$$

In the next phase of autoclave processing, P_a is held constant over a dwell period to promote bundle impregnation. we find the expression to describe the flow front progression, L, with respect to time in phase 3 is:

$$\frac{H_{g0}(H_{g0}+d)}{\phi^2}\ln\left\{\frac{H_{g0}+d-L_r\phi}{H_{g0}+d-L\phi}\right\}+\frac{H_{g0}}{\phi}(L_r-L)=\frac{P_aK}{\mu\phi}(t-t_r) \qquad (7)$$

where L_r is found using Eqn. (6). Material properties and constants can be found in Table 1.

Table 1 Material properties and measured geometric parameters for the fiber tow

Material Property [units]	**Value**	**Material Property [units]**	**Value**
Tow Permeability: K [m^2] at 0.345 MPa	$2.5 * 10^{-13}$	Molten HDPE Viscosity: μ [Pa·s]	2750
Tow Permeability: K [m^2] at 0.690 MPa	$2 * 10^{-13}$	Bundle Porosity: φ	.38
Tow Permeability: K [m^2] at 1.379 MPa	$1.5 * 10^{-13}$	H_{g0} [m]	0.000735
		d [m]	0.002561

RESULTS

The model was compared to the available experimental data for void fraction within a fiber bundle after being pressurized at 0.345 MPa and 0.69 MPa for 30 and 60 minutes and at 1.379 MPa for 30 minutes using a ramp rate of 804 Pa/s. The theoretical void fraction from the simplified model can be calculated using Eqns. (6) and (7) to find the location of the impregnation of resin into the fiber tow, L at 30 and 60 minutes and using the value of L in the expression below one can find the void fraction inside a tow:

$$v_v=\frac{V_v}{V_r+V_f+V_v}=\frac{(d-L)\phi}{H_{g0}+d} \qquad (8)$$

The minimum possible void fraction can be calculated based on the known dimensions of the tow and tow gap. As stated before, the only resin available to fill the tow is the resin in the tow gap. Thus, the maximum possible position of the flow front (L_{max}) can be found by dividing H_{g0} by the porosity of the tow. The experimental parameter values presented in Table 1 were used to find L_{max} which can be substituted for L in Eqn. (8) to find the minimum possible void fraction equal to 0.0723 under any pressure as there is no more resin available in the macropore to impregnate the tows. These will change based on the geometry of the unit cell and if step 1 of the process can completely fill the gap. The experimental data is compared to the model predictions in Table 2.

Note that there is a small increase in experimentally measured void content between the 0.690 MPa/60 minute sample and the 1.379 MPa/30 minute sample. This phenomenon could be due to the fiber bundles becoming overly compressed before the resin has a chance to impregnate the bundle entirely. Mayer et al. observed a similar result on their work [8]**.** No data point was taken

for the 1.379 MPa/60 minute sample (hence the empty slot in Table 2). The model can predict lower value for void fraction if there was sufficient resin available in between the tows but as for our case, irrespective of the pressure of time, once the resin in the tows is utilized, the resin front cannot process any further.

Table 2 Experimentally measured and model predicted values of void fraction.
(* See discussion for explanation)

Pressure [MPa]	**Model**	**Experimental**	**Model**	**Experimental**
	30 minutes		**60 minutes**	
0.345	0.16	0.17 ± 0.05	0.11	0.14 ± 0.01
0.690	0.12	0.06 ± 0.06	0.07*	0.036 ± 0.01
1.379	0.07*	0.05 ± 0.016	-	-

DISCUSSION

As noted before, based on the geometry of the unit cell of the fiber preform the minimum possible void fraction is 0.0723. This is considering a very simplified version of the unit cell with resin only being contributed by the resin gap. Note that in some cases the experimental values for void fraction are lower than 0.0723. This could be due to many reasons. First, in reality gaps that are perpendicular to the tows being considered in the unit cell could be contributing to the impregnation of the tow. Also, when more than one layer of material is stacked in the powder impregnation mold there is some caking of power in between the layers. It is not removed prior to compression molding and may contribute to tow impregnation as well. While taking the limitation of the resin gap into consideration the model in cases 0.690 MPa/60 minutes and 1.379 MPa/30 minutes would be halted at 0.0723, prior to the completion of the dwell time. If the restriction in regards to the resin gap size is removed (thus considering resin from other sources besides just the gap for tow impregnation) and the model is allowed to run through to the full dwell time then the void fraction will 0.065 which is a little closer to the experimental value

The permeability values used in the model calculations for each applied pressure are listed in Table 1. The permeability was assumed to be constant from the start of the compression through to the end of the dwell times and for all applied pressure. In reality, the permeability of the tow most likely decreases as pressure is increased and will be less for the higher applied pressures (0.690 and 1.379 MPa). In order to check our assumption we can determine a value for the permeability of the by substituting the experimentally measured void fraction in the model for 30 minutes of dwell time for the three different pressures. The permeability values that will give us exact match between experiments at 30 minutes for the three different pressures used in the model to predict the void fraction for the 60 minutes cases for the 0.345 and 0.690 MPa applied pressures for which we have experimental data. This calculation yields a void fraction of 0.1766 for 0.345 MPa at 60 minutes (the experimental value was 0.1364) and a void fraction of 0.0641 for 0.690 MPa at 60 minutes (the experimental values was 0.036). Thus our simple model with many assumptions is able to predict void fraction within acceptable values, considering the uncertainty on permeability and the assumptions made to describe the geometry and the flow.

CONCLUSIONS

A model for the flow of highly viscous, molten thermoplastic resin into a compressed tow has been developed. In this model it was assumed that permeability remained constant and deformation of the fiber bed was not taken into account. A semi-experimental value for the permeability of the tow was calculated using the model and the data generated from the SEM images. When this permeability was then applied to the model the void fraction predicted for one case was slightly more accurate than the original model prediction with an estimate for the permeability of the tow.

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