Hindawi Publishing Corporation
Research Letters in Materials Science
Volume 2007, Article ID 37123, 5 pages
doi:10.1155/2007/37123

Research Letter

Processability of Pultrusion Using Natural Fiber and Thermoplastic Matrix

Tham Nguyen-Chung,[1] Klaus Friedrich,[2] and Günter Mennig[1]

[1] *Institut für Allgemeinen Maschinenbau und Kunststofftechnik, Technische Universität Chemnitz, 09107 Chemnitz, Germany*
[2] *Institut für Verbundwerkstoffe, Technische Universität Kaiserslautern, 67663 Kaiserslautern, Germany*

Correspondence should be addressed to Tham Nguyen-Chung, tham.nguyen.chung@mb.tu-chemnitz.de

Received 2 August 2007; Accepted 17 October 2007

Recommended by D. Chen

Fundamental mechanisms of the pultrusion process using commingled yarns of polypropylene matrix and discontinuous flax fiber to produce thermoplastic profiles were investigated in numerical and experimental manners. Essential issue is the fact that all natural fibers are discontinuous by nature, which may negatively influence the processability. The pultrusion process will be only successful if the pulling force exerted on the solidified pultrudates can be transmitted to the regions of unmelted commingled yarns by "bridging over" those melted regions within the die. This can be realized by applying a sufficient number of small yarn bundles of high compactness rather than a thicker single bundle of lower compactness as the raw material. Furthermore, the possibility of adding extra melt into the yarn bundles by side-fed extrusion has been investigated showing that the impregnation can be improved only for the outer layers of yarns, which is owed to the high viscosity of the thermoplastic melt and the limited length of the die.

1. INTRODUCTION

Pultrusion is a continuous process for manufacturing fiber reinforced profiles of constant cross-section. So far, the process has been mainly applied for thermosetting matrices as there are inherent difficulties associated with the thermoplastic matrices, such as high processing temperatures and high melt viscosities. However, with recent developments of preforms to facilitate impregnation, such as pre-impregnated tapes, commingled yarns, and powder-impregnated bundles [1, 2], thermoplastic pultrusion has gained a greater interest.

Figure 1 shows the heating die as an opening and steady-state thermodynamic system with reinforcing filaments and polymer melt fed separately (case A) or in term of commingled yarns (case B). The drag flow is a result of the movement of the filaments and the no-slip condition. Moreover, in case A the drag flow is superposed by the pressure-driven flow. With a proper design of the die inlet, the drag flow can compensate the pressure driven flow so that no sealing is needed. In case B no precaution is required since the melt is not pressurized.

The pultrusion process will be only stable if the force flux is properly transmitted from the outlet to the inlet, which is possible using continuous fibers like glass fiber rovings. Natural fibers are, however, not endless by nature. Their average length is normally shorter than the minimum length of the die which is required to ensure sufficient heat transfer and proper impregnation. So the only way to maintain the force flux between the outlet and inlet of the die is to make use of special textile technique to spin the separate fibers together into a bundle or to build extra core cords into the yarns. In this work, the fundamental mechanisms of the pultrusion of natural fibers reinforced thermoplastics were investigated on commingled yarns of polypropylene and flax fibers, whereby the possibility of adding extra melt by side-fed extrusion to improve impregnation was also considered. The objective of the paper is to find out a proper way to maintain the force transmission along the discontinuous yarns while at the same time ensuring a proper impregnation of the commingled yarns.

2. SIMULATION

Numerical simulation was carried out for a two-dimensional rectangular geometry, that is, effects caused by the side edges

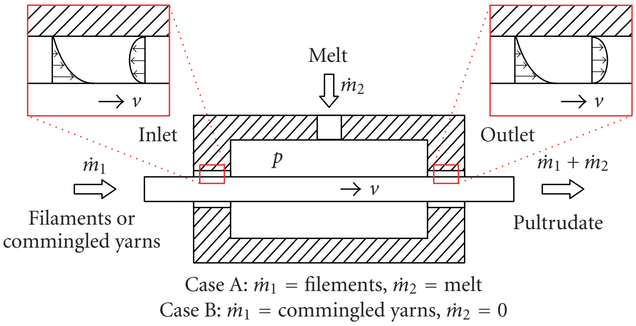

FIGURE 1: Heating die and the flow situation at the inlet and outlet.

TABLE 1: Thermal and rheological properties of polypropylene and commingled yarns [4].

Property	Unit	Matrix (PP)	Commingled yarns
Density ρ	kg/cm ^{3}	900	1074
Specific heat c_p	J/kgK	1700	1600
Thermal conductivity k	W/mK	0.2	0.23
Reference temperature T_0	K	463	463
Zero-shear-rate viscosity $\eta_0(T_0)$	Pas	936.7	740000
Time constant λ	s	1.74	725
Power law index n	—	0.61	0.33
Arrhenius coefficient α	K	4330	4980

were neglected. First, it was considered that the yarns act like a continuous fiber bundle. The calculated temperature field will serve as an assumption for the temperature in further simulations. Although this may not meet the reality, it is a good way to deal with such unstable processes, where a coupled steady simulation would end up with no convergence. In a next step, the flow situation of the discontinuous yarns containing virtual cords was considered, whereby the velocities at the corresponding cord positions were fixed (Figure 2). In both cases, the commingled yarns were modeled as a fluid; and inertia, gravitation, and compressibility were neglected so that the equations of mass, momentum, and energy can be written as follows:

$$\nabla\cdot\vec{v} = 0,$$
$$-\nabla p + \nabla\cdot\underline{\tau} = 0, \qquad (1)$$
$$\rho c_p\left(\frac{\partial T}{\partial t} + \vec{v}\cdot\nabla T\right) = \nabla\cdot(k\nabla T) + \underline{\tau} : \underline{\dot{\gamma}},$$

where $t, \vec{v}, T, p, \underline{\tau}, \underline{\dot{\gamma}}, \rho, c_p$, and k denote time, velocity vector, temperature, hydrostatic pressure, extra-stress tensor, rate of deformation tensor, density, specific heat, and heat conductivity, respectively. The constitutive equation for a generalized Newtonian fluid is

$$\underline{\tau} = 2\eta(T,\dot{\gamma})\underline{\dot{\gamma}}, \quad \underline{\dot{\gamma}} = \frac{1}{2}(\nabla\vec{v} + \nabla\vec{v}^T). \qquad (2)$$

The viscosity was approximated using the Bird-Carreau model [3]:

$$\eta = \eta_0\left[1 + (\lambda\dot{\gamma})^2\right]^{(n-1)/2}, \quad \dot{\gamma} = \sqrt{2\underline{\dot{\gamma}} : \underline{\dot{\gamma}}}, \qquad (3)$$

where η_0 is the zero-shear-rate viscosity, n is the power law index, and λ is the time constant. The Arrhenius model was used for the viscosities at different temperatures:

$$\eta_i(T) = \eta_i(T_0)a_T(T), \quad a_T = \exp\left[\alpha\left(\frac{1}{T} - \frac{1}{T_0}\right)\right] \qquad (4)$$

with the reference temperature T_0 and the Arrhenius coefficient α. The thermal and rheological properties of polypropylene and commingled yarns in the melted state are listed in Table 1.

Finally, the situation using polypropylene as an extra melt to additionally impregnate the yarns was considered, whereby the flow in a porous media is governed by the Darcy law:

$$\vec{v} = -\frac{\underline{K}}{\eta_0}\nabla p. \qquad (5)$$

The longitudinal component K_{xx} of the permeability tensor $\underline{K}$ is calculated using the Kozeny-Carman model [5]:

$$K_{xx} = \frac{D_f^2}{16C}\frac{(1 - V_f)^3}{V_f^2}, \qquad (6)$$

where D_f is the fiber diameter, V_f is the fiber volume fraction (Table 2), and C is the Kozeny-Carman constant. The value of the Kozeny-Carman constant C was taken as 0.093 [6]. The transversal component K_{yy} was given by the Bruschke model [7]:

$$K_{yy} = \frac{l_e^2}{16}\left(\ln(l_e) - \frac{3}{4} + l_e^{-2} - \frac{l_e^{-4}}{4}\right)D_f^2 \quad \text{with } l_e^2 = \frac{1}{V_f}. \qquad (7)$$

The following boundary conditions were set (Figure 2): symmetry condition at the symmetry, slip wall along the yarn/air interface, no-slip along the die wall, constant velocity v_p along the so-called virtual cords which may be added into the cavity to simulate the solid areas within the melt, constant temperatures at the yarn and melt inlet. At the interface with the die and the air, heat flux densities were defined as follows:

$$q_d = \alpha_d(T - T_d), \quad q_a = \alpha_a(T - T_a) \qquad (8)$$

with constant die and air temperature T_d and T_a as well as constant heat transfer coefficients α_d and α_a. The die temperature T_d was kept constant. At the melt inlet, the velocity was either zero or constant, depending on whether extra melt was considered.

The governing equations together with the constitutive equation were solved numerically using the FE-code FIDAP [8]. A mixed formulation was applied for pressure and velocity. In transient case, the implicit backward Euler approach was used for the time integration.

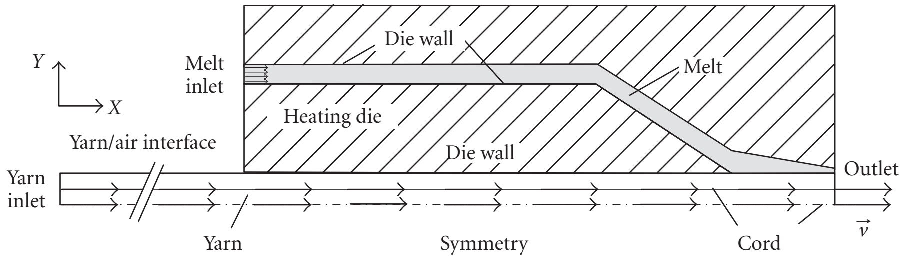

FIGURE 2: Model geometry and boundaries.

TABLE 2: Characterization of commingled yarns of polypropylene (PP) and flax [4].

Property	Unit	Value
Flax fiber volume fraction V_f	%	30
Density of PP ρ_{pp}	kg/m^3	900
Density of flax ρ_f	kg/m^3	1480
Diameter of PP fibers D_{PP}	μm	60
Diameter of flax fibers D_f	μm	60
Length of PP fibers L_{PP}	mm	40
Average length of flax fibers L_f	mm	40

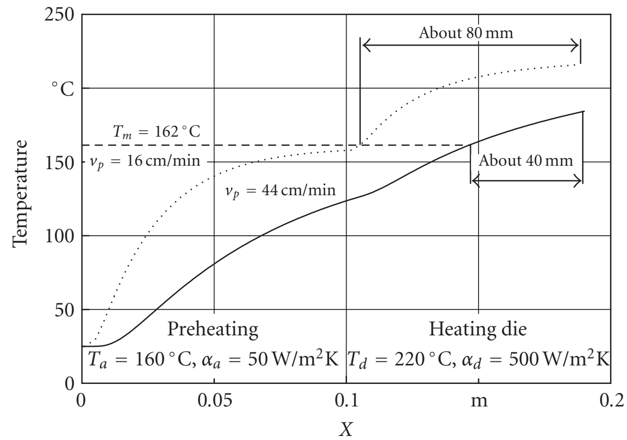

FIGURE 3: Temperature along the symmetry line.

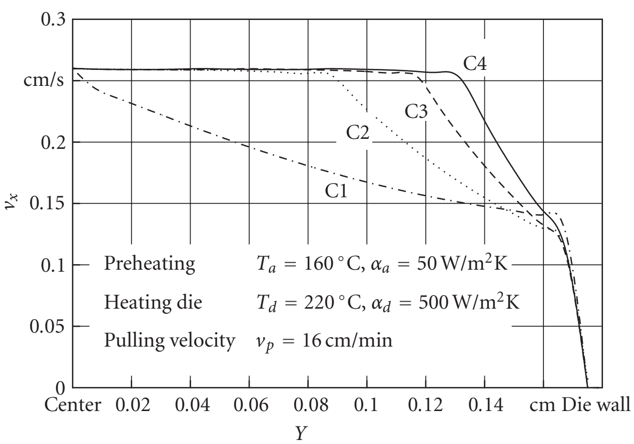

FIGURE 4: Longitudinal velocity along the outlet.

3. RESULTS

3.1. Continuous yarns

Figure 3 shows exemplarily the temperature along the symmetry line in case the yarns were considered as being continuous. At a low pulling velocity $v_p = 16$ cm/min, the section containing the melting yarns, that is, where their temperatures are above the melting point T_m of the polypropylene matrix ($T_m = 162$°C), reaches a length of approximately 80 mm, which means that a proper melting of the yarns throughout the commingled yarns is generally possible. The main problem is therefore to find a proper way to maintain the force transmission along the yarns, while enabling a proper melting of the yarns. This can be done by increasing the pulling speed above 44 cm/min to reduce the melting section below the average length of the natural fibers ($L_f \approx 40$ mm) so that it is possible to transmit the pulling force along the symmetry of the die.

3.2. Discontinuous yarns

When the commingled yarns are partially melted, they cannot be assumed as being continuous. However, virtual cords may be added along the unmelted area of the yarns to enforce the transmission of the pulling velocity. In the following cases, either one virtual cord was added along the symmetry line (C1) or up to three other ones are distributed equidistantly over half of the die thickness (C2 to C4). Along the cords, the velocity is kept equal to the pulling velocity.

In Figure 4 the corresponding longitudinal velocity at the die outlet is shown respectively. It can be seen clearly that with one cord at the symmetry line (C1), the longitudinal velocity obtained is far away from this ideal situation of a uniform velocity across the thickness. As the number of the cords increases, the velocity profiles at the outlet show a wider region of constant velocity in the middle of the die. With four extra cords (C4) a good shaping of the finished product may be expected, since the velocity profile at the outlet is not so much different from a typical velocity profile in an extrusion die for a shear-thinning polymer. Since significant improvement of the velocity profile can be still achieved as the cord number over half of the cavity thickness was increased from 2 to 3, it is recommendable that at least 5 cords should be used for the whole cavity thickness to ensure a good shape of the pultrudate.

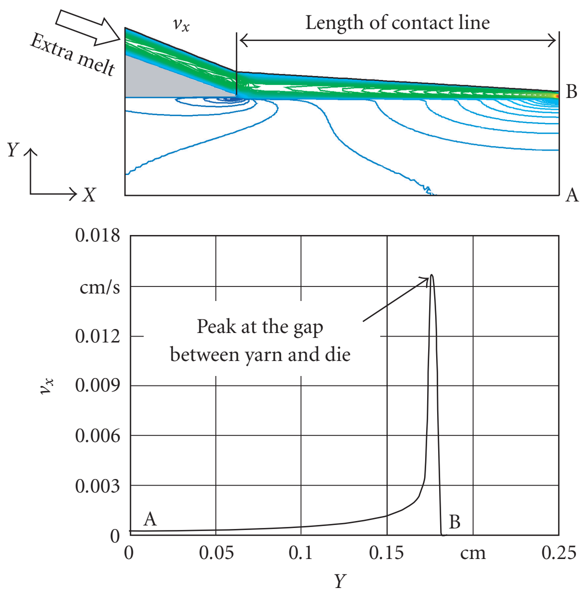

FIGURE 5: Longitudinal velocity in the die (top) and along the outlet (bottom).

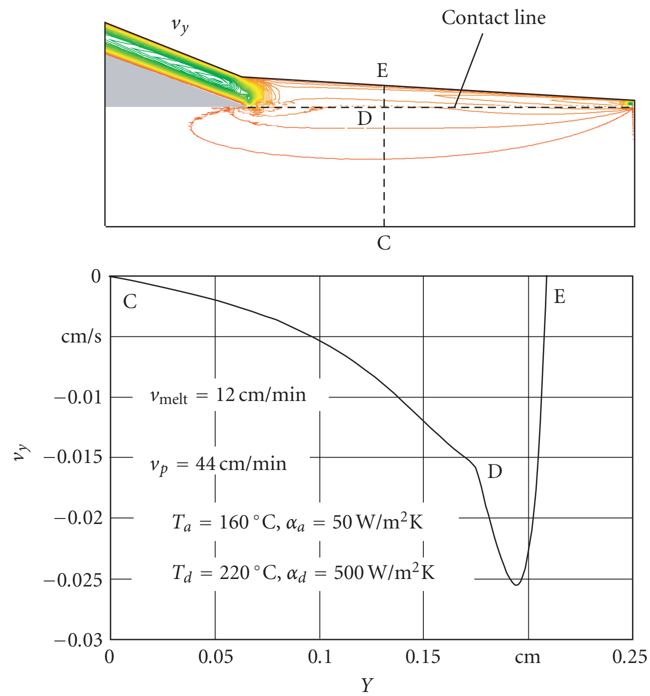

FIGURE 6: Transversal velocity in the die (top) and along the line CDE (bottom).

3.3. *Extra melt*

When extra melt was inserted to improve the impregnation of the yarns which were modeled as a porous media, the movement of the yarns was only considered in the temperature calculation, but not in the flow simulation. Figures 5 and 6 show contour plots of the velocity parallel and across the flow direction within the contact region yarn/melt. Based on the transversal velocity v_y at a representative cross-section (e.g., along the line CDE) as shown in Figure 6, the time needed for the extra melt to fully impregnate the whole thickness of the yarns was estimated. In current case, the time required for a full impregnation is much longer than the time the yarns stay in the contact region, which means that only the outer layers of the commingled yarns can be properly impregnated. In order to improve the impregnation of the inner layers, the length of the contact region should be significantly increased. Moreover, the contact region should end at a certain distance away from the outlet to avoid such a peak of the longitudinal velocity v_x occurred at the gap between the yarns and the die wall (Figure 5, bottom), which may negatively influence the shape of the finished product.

4. EXPERIMENTS

Experiments were conducted on commingled yarns of polypropylene and flax fibers [9], which were spun by the Thüringisches Institut für Textil- und Kunststoff-Forschung (Rudolstadt, Germany). Rectangular profiles of 3.5 mm × 10 mm were manufactured using the self-designed facility at the Institut für Verbundwerkstoffe (Kaiserslautern, Germany) (Figure 7). Several bundles of yarn were stored in a creel stand (Figure 8) and then guided into the preheating chamber, where it was heated to a temperature close to the melting temperature of the polymeric fibers. Directly after

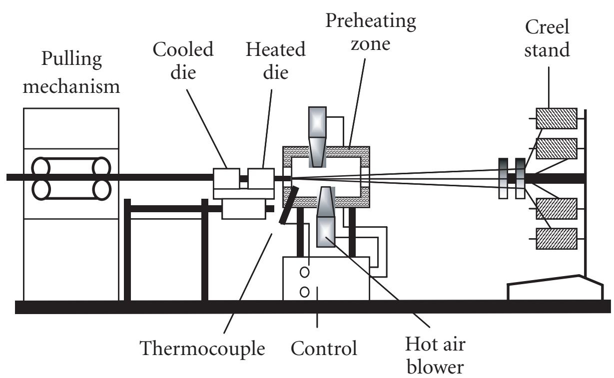

FIGURE 7: Schematic of the pultrusion facility.

preheating, the yarn bundles were pulled through the heating die where the polymer is supposed to melt and impregnate the flax fibers before being consolidated in the cooling die. Table 3 shows some of the process conditions which led to successful experiments.

Some successful examples of the finished profiles can be seen in Figure 9, whereby 80 bundles of the yarns have been used to fill the die cavity. Assuming that the cavity was totally filled and each bundle is of rectangular geometry, the edge length of each bundle can be calculated, which is approximately 0.66 mm. In other words, 5 to 6 yarn bundles were needed for a die thickness of 3.5 mm to ensure the success of the pultrusion process, which verified well the theoretical requirements for the number of yarns as revealed by the numerical simulation.

This way, it can be shown that the pultrusion process is stable when a sufficient number of yarn bundles of high

Table 3: Pultrusion parameters using commingled yarns of polypropylene and flax.

Process number	Preheating temperature [°C]	Heating die temperature [°C]	Pulling speed [cm/min]	Bundles number	Cooling die temperature [°C]
1	155	210	12	80	25
2	155	220	12	80	25
3	160	220	24	80	25
4	160	230	24	80	25
5	160	240	24	80	25
6	166	240	36	80	25
7	166	250	36	80	25

Figure 8: Creel stand.

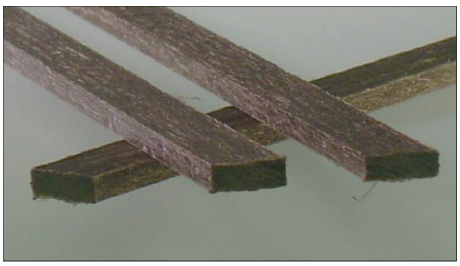

Figure 9: Pultrusion profiles.

compactness are used instead of a single bundle of less compactness. The reason might be that due to the compactness some spots of the compact bundles remain unmelted, which behave like core cords so that a throughout force transmission is possible. Moreover, the compact bundles are less sensitive to break and also the force required to pull the compact bundles is smaller since there is no need to compress the bundle into the die.

5. CONCLUSIONS

The simulations and experiments have revealed some of the fundamental issues about the pultrusion of discontinuously fiber reinforced thermoplastics. In general, it is possible to produce pultrusion profiles by using a sufficient number of compact bundles of commingled yarns as raw material. The requirement for the minimal number of yarn bundles to ensure a good shape of the finished pultrudate may be theoretically calculated using numerical simulation. Extra melt inserted into the yarn bundles by side-fed extrusion to improve the impregnation effects only the outer layers of yarns, which is owed to the high viscosity of the thermoplastic melt and the limited length of the die.

ACKNOWLEDGMENTS

The authors would like to thank Dr.-Ing. T. Reußmann at the Thüringisches Institut für Textil- und Kunststoff-Forschung (Rudolstadt, Germany) for supplying polypropylene/flax yarns. Financial support by the Deutsche Forschungsgemeinschaft (Bonn, Germany) is gratefully acknowledged. The present paper is based on a presentation at ICAPP 2007, the 2nd International Conference on Advances in Petrochemicals and Polymers, held in Bangkok, Thailand on 25 - 28 June 2007.

REFERENCES

[1] G. Bechtold, S. Wiedmer, and K. Friedrich, "Pultrusion of thermoplastic composites—new developments and modelling studies," *Journal of Thermoplastic Composite Materials*, vol. 15, no. 5, pp. 443–465, 2002.

[2] W. J. Tomlinson and J. R. Holland, "Pultrusion and properties of unidirectional glass fibre-polypropylene matrix composites," *Journal of Materials Science Letters*, vol. 13, no. 9, pp. 675–677, 1994.

[3] R. B. Bird, R. C. Armstrong, and O. Hassager, *Dynamics of Polymer Liquids*, vol. 1, John Wiley & Sons, New York, NY, USA, 1977.

[4] T. Reußmann, *Entwicklung eines Verfahrens zur Herstellung von Langfasergranulat mit Naturfaserverstärklung*, Ph.D. thesis, Technische Universität Chemnitz, Germany, 2002.

[5] P. C. Carman, "Flow through granular beds," *Transaction of the American Institute of Chemical Engineers*, vol. 15, pp. 150–166, 1937.

[6] S. Voorakaranam, B. Joseph, and J. L. Kardos, "Modeling and control of an injection pultrusion process," *Journal of Composite Material*, vol. 33, no. 13, pp. 1173–1203, 1999.

[7] M. V. Bruschke and S. G. Advani, "Flow of generalized Newtonian fluids across a periodic array of cylinders," *Journal of Rheology*, vol. 37, no. 3, pp. 479–498, 1993.

[8] FIDAP User's Manual, *Fluent Inc.*, Lebanon, Ohio, USA, 1998.

[9] I. Angelov, S. Wiedmer, M. Evstatiev, K. Friedrich, and G. Mennig, "Pultrusion of a flax/polypropylene yarn," *Composites Part A: Applied Science and Manufacturing*, vol. 38, no. 5, pp. 1431–1438, 2007.