Research on the melt impregnation of continuous carbon fiber reinforced nylon 66 composites

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Abstract. Impregnation mold of continuous carbon fiber reinforced thermoplastic composites was designed and built in the article. Based on the theory of fluid mechanics and Darcy's law, a model of the melt impregnation was also established. The influences of fiber bundle width and impregnation pins’ diameter on the impregnation degree were studied by numerical simulation. Continuous carbon fiber reinforced nylon 66 composites were prepared. The effects of coated angle and impregnation mold temperature on the mechanical properties of the composites were also described. The agreement between the experimental data and prediction by the model was found to be satisfactory.

1. Introduction

Demanding for advanced materials in aerospace and automobile industry applications has led to the emerge of composites. Composites were a kind of heterogeneous material, which consisted of two or more substances of different physical or chemical properties. In the 1960s, the short fiber reinforced thermoplastic composites had realized industrial production. However, the composites can not be applied in the force bearing component and structural use. Therefore, continuous fiber reinforced thermoplastic composites (CFRP) were developed, which was regarded as a novel material consisted of carbon or glass fibers and polymer matrix. It was proved that thermoplastic composites were superior to thermosetting counterpart for the superior mechanical properties in practical application, such as short cycle-time, high strength, high elastic modulus, light weight and so on. However, the thermoplastic composites were too difficult to be impregnated because of the high viscosity. The traditional impregnation processes of manufacturing continuous fiber reinforced thermoplastic composites were powder impregnation, solvent impregnation as well as melt impregnation [1]. Among these methods, the process which had got some attention was melt impregnation process. During the impregnation process, the fiber bundles were pulled through the guide pins, which were under the molten
thermoplastic. Due to the high viscosity of the molten thermoplastic, it was difficult for the molten polymer completely penetrate into the fiber bundle. To solve the problem, many studies had been done to research how to reduce the viscosity and realize the better impregnation. Gaymans [2] studied that the polypropylene impregnated the glass fiber roving with pins and the results showed that the diameter of the pin, tension and melt viscosity were the key factors during the impregnation. Peltonen [3] studied the effects of the temperature, time, pressure, the length of the contact surface, tension force and MFR-value of PP on the impregnation. Nygard [4] pointed out that using the radial slit die, which included different vibration techniques and a silt die, was better than the cross-head impregnation mold. Meanwhile, they found that with the use of the radial slit die, the high volume fraction was obtained and the fiber-matrix content was controlled. Kumar [5] prepared the PP/LGF reinforced particles by melt impregnation device designed by themselves. Asai [6] used the screw to take the molten resin into the mold, then take the use of impregnation unit by specific design to achieve the full impregnation of fiber. Hatano [7] manufactured continuous fiber reinforced thermoplastic composites with the use of a combination of pull extrusion molding and weaving techniques. Cogswell [8] studied the impregnation technology of thermoplastic composites and the impregnation effect was best when the viscosity of melt was less than 30 Pa·s. Törmälä [3] discussed the effect of temperature on the impregnation degree by the experiment of glass fiber reinforced polypropylene, the results showed that the impregnation degree of the polymer was improved by 20% when the temperature of the polymer increased from 180℃ to 230℃. Wevers [2] studied the effect of traction on impregnation effect by the experiment of continuous glass fiber reinforced polypropylene. It was shown that the impregnation degree increased with the increase of traction force. Kim [9] came to the conclusion from the experiment of carbon fiber reinforced polyetheretherketone that the impregnation effect improved with the increase of pressure.

In this paper, a new impregnation model was developed not only discussed the variety between the carbon fiber roving and the pins but also analyzed the influence on the mechanical properties of composites, process parameters and structure parameters on the impregnation efficiency.

2. Materials and equipment
Carbon fiber (T700SC) with Number of Filament being 12000 was supplied by Toray Industries, Inc. Tokyo, Japan. Nylon 66 (A-1) was supplied by Rhodia polyamide (Shanghai) Co., Ltd. Twin-screw extruder (KS20) with diameter of single screw being 20mm was supplied by Kunshan Kesun Plastic & Rubber Machinery Co., Ltd. Injection molding machine (PT-130) was supplied by L.K. Group, Hong Kong, China.

3. Establishment and numerical simulation of impregnation model
3.1. Establishment of impregnation model
The impregnation process of carbon fiber bundle was displayed in Figure 1. There were a series of parallel pins in the impregnation device. The carbon fiber bundle can be fully impregnated by the molten polymer due to the tension action when the carbon fiber bundle pulled out of the impregnation device. After fully impregnated, the carbon fiber bundle pulled through an exit mold of a diameter of 3mm. The fiber passed through a series of parallel pins, the impregnation would occur on the contact surface, as shown in Figure 2. The exit mold could control the fiber content. Impregnation was accomplished by the polymeric matrix penetrate into surrounding fiber bundles.
According to the previous research [10, 11], the melting rate of polymer that penetrated into the fiber bundle was considered as the most important factors in the process of melt impregnation. The rate that the molten polymer penetrated into the fibers had been analyzed by Darcy’s law [12], as shown in the equation (1), which was an empirical relation from experiments on water flow through sand beds. We assumed that Darcy’s law could be used in the vertical flow of the thermoplastic composites,

\[ V_h = -\frac{K}{\mu} \frac{dP}{dh} \]  

(1)

Where \( V_h \) is the vertical velocity of the thermoplastic composites, \( P \) is the applied pressure, \( h \) is the designates the vertical position. The mass flow rate through the vertical position \( h \) is,

\[ m = \rho (-V_h) W \epsilon \]  

(2)

And \( K \) is the permeability coefficient of the fiber structure, which can be calculated by the Kozeny-Carman equation [13],

\[ K = \frac{D_i^3 \phi^3}{16k (1-\phi)^2} \]  

(3)
Where $D_f$ is the fiber diameter, $\phi$ is the porosity of the carbon fiber bundle and $k$ is a constant related to polymer viscosity, fiber structure and fiber type, which can be calculated from the following equation [3],

$$k = \frac{2\phi^3}{(1-\phi) \left\{ \ln \left( \frac{1}{1-\phi} \right) \left[ \frac{1-(1-\phi)^2}{1-(1-\phi)^2} \right] \right\}}$$

Equations (1) and (2) are combined to get the following equation,

$$\frac{m}{\rho W \varepsilon} = \frac{K}{\mu} \frac{dP}{dh}$$

The pressure at the $h=h_0$ is the applied pressure, $P_0$. The following equation is obtained by the integration of equation (5) from $h_0$ to $h_f$,

$$\frac{m}{\rho W \varepsilon} (h_f-h_0) = \frac{K}{\mu} (P_a - P_0) \frac{dh_f}{dt}$$

Where $h_f$ is the location of the thermoplastic composites where the pressure is the atmospheric pressure $P_a$. The mass flow rate can be expressed in term of $dh_f/dt$ as,

$$m = \rho W \varepsilon \left( -\frac{dh_f}{dt} \right)$$

Equations (6) and (7) are combined to get the following equation,

$$-(h_f-h_0) \frac{dh_f}{dt} = \frac{K}{\mu} (P_a - P_0) dt$$

The following equation is obtained by the integration of equation (8) along with the initial condition ($h=h_0$ at $t=0$),

$$t = \frac{\mu h_0^2}{2K (P_0 - P_a)} \left[ 1 + \left( \frac{h_f}{h_0} \right)^2 - 2 \frac{h_f}{h_0} \right]$$

The impregnation degree $D_{imp}$ can be defined as the ratio between the number of impregnated fibers and the total number of fibers. If we assume that the fibers are uniformly distributed, the impregnation degree can be expressed as,

$$D_{imp} = \frac{\text{area impregnated}}{\text{total area}} = \frac{Wh_f - Wh_0}{Wh_0} = 1 - \frac{h_f}{h_0}$$
Substituting equation (10) into equation (9). The impregnation degree is obtained as a function of
time, applied pressure, viscosity, initial bundle vertical and permeability,
\[ t = \frac{\mu h_0^2}{2K(P_0 - P_a)} \left[ 1 + \left(1 - D_{imp}^2\right)^2 - 2\left(1 - D_{imp}\right) \right] \] (11)

We assumed that the cross-sectional is the rectangular shape, the total cross-sectional area equal to
the cross-sectional area of the mono-filaments and the cross-sectional area impregnated by
thermoplastics composites. The relationship between the impregnation thickness \( h_0 \) and the total
number of carbon fibers \( n_f \) and the diameter of the carbon fiber \( D_f \) and the fiber bundle width \( W \) and
the porosity \( \phi \) can be calculated from geometric considerations. This is shown schematically in Figure
2 and expressed mathematically in equation (12),
\[ h_0 = \frac{\pi n_f D_f^2}{4 W (1 - \phi)} \] (12)

The total coated time \( t \) has relations with the coated angle \( \theta \), the number of the pin \( N \), the pin
diameter \( D \), the traction speed \( U \). This is expressed mathematically in equation (13),
\[ t = \frac{\theta D}{2U} \] (13)

Bates [10] had studied the melt pressure while a glass fiber pulled over a pin in polyamide 66 or
polypropylene polymers melt pool. The results showed that the melt pressure is,
\[ P_0 = \frac{2T}{WD} \] (14)

Equations (11) to (14) are combined to get the following equation,
\[ t = \frac{\theta D}{U} = \frac{\mu}{K \left( \frac{2T}{WD} - P_a \right)} \left[ \frac{\pi n_f D_f^2}{4 W (1 - \phi)} \right]^2 \left[ 1 + \left(1 - D_{imp}^2\right)^2 - 2\left(1 - D_{imp}\right) \right] \] (15)

The configuration of pins is shown in Figure 3.

\[ \theta \]
\[ D \]
\[ L \]

**Figure 3.** The configuration of pins

According to geometric knowledge, we get the relationship among the coated angle \( \theta \), the pin
diameter \( D \) and the center distance between the two pins \( L \), written as,
\[ \theta = 2 \arcsin \left( \frac{D}{L} \right) \]  

(16)

Substituting equation (16) into equation (15), the following equation can get,

\[ D_{\text{imp}} = \frac{4W}{\pi n_j D_j^2} \sqrt{\frac{2KD}{\mu U}} \left( \frac{2T}{WD} - P_a \right) \arcsin \left( \frac{D}{L} \right) \]  

(17)

Equation (17) provides a relationship between the impregnation degree and the process variables in the impregnation process, namely, physical parameters, process parameters and structure parameters.

Equations (1) and (17) showed that the impregnation degree is determined by numbers of factors, including the permeability coefficient of fiber structure, the fiber bundle width, the pin diameter, the distance between two pins, the line speed of the fiber, the fiber pretension and melt viscosity. Among these factors, the permeability coefficient and Kozeny-Carman constant can be calculate based on the porosity and the melt viscosity, which regarded as constants for a Newton fluid. Therefore, in the following section, the influence of other impregnation parameters would be mainly discussed.

To quantitatively analyzed the influence of the line speed of fiber, a T700 carbon fiber through a high-viscosity melt was impregnated. The melt viscosity was assumed to be 50Pa·s, the fiber bundle width was 6mm, the pretension was 20N, the pin diameter was 8mm. These parameters were used for the calculation below unless otherwise specified.

3.2. Numerical simulation analysis

3.2.1. Effect of fiber bundle width on impregnation degree

![Figure 4. Degree of impregnation as a function of the fiber line speed and the fiber bundle width.](image)

Figure 4 was the impregnation degree calculated as a function of the line speed of the fiber at different fiber bundle width of 4mm, 8mm and 12mm. For a single curve, such as the 4mm one, the impregnation degree decreased sharply at a low speed, reached the steady state at about 0.1m/s. The
Figure showed when the line speed of the fiber was greater than 0.1 m/s, the degree of impregnation had a little changes. This can be explained as the melt flow. There are two factors affected the impregnation process, the amount of the melt and the impregnation duration. With the increase of the line speed of the fiber, the amount of the melt flowing into the fiber bundle decreased, while the duration shortening of impregnation process. For a low line speed of the fiber, more and more polymer penetrated into the fiber bundle, resulting in a high degree of impregnation due to the sufficient time. On the contrary, the duration of impregnation decreased sharply resulting in a relatively low degree of impregnation, but in the actual production, the relationship between the performance of the product, the production efficiency and the line speed of the fiber should be considered. As a result, in the premise of ensuring the performance of the product and the production efficiency, the maximum speed could be chosen.

With the increase of the fiber width, the distance between the mono-filament fibers became wider, resulting in an enhancement of the impregnation process. The degree of the impregnation increased, while the steady state was almost unchanged.

3.2.2. Effect of tension on impregnation degree

Figure 5. Degree of impregnation as a function of the fiber line speed and the fiber tension.

Figure 5 showed the influence of different tensions on impregnation degree. The calculation was done by the different tensions of 10N, 20N and 30N, while fixing other parameters mentioned above. Increasing the value of tension, degree of impregnation was improved, which could be proved in the experimental result reported by Peltonen [3] and Gaymans [2]. The starting value of the steady state was unchanged. It was indicated that the value of the stable state did not depend on the fiber bundle and the tension. Also, with the increase of fiber tension, the pressure between the fiber bundle and the pins increased according to equations (1) and (2), which could also enhance the degree of impregnation according to the experimental results reported by Kim [9].
3.2.3. **Effect of pin diameter on impregnation degree**

The degree of impregnation as a function of the fiber line speed and the fiber diameter.

The influence of different pin diameter on the impregnation degree was shown in Figure 6. The calculation was done by the different pin diameters of 4mm, 8mm and 12mm. By increasing the value of the pin diameter, the contact length of the fiber bundle and the pins increased resulting in enhancing the degree of the impregnation, which could be proved in the experimental results reported by Peltonen [14]. The amount of polymer impregnated with one pin was limited as we have seen above. Several pins were often used to strengthen the impregnation. According to the model in this paper, the impregnation degree of each pin and the total impregnation degree of multiple pins could be calculated.

The analyses discussed above was only based on the ideal boundary conditions. For a real case, there existed a minimum pressure between the fiber bundle and the pins. When the pressure was lower than the atmospheric pressure \( P_0 \), the penetration could not be realized and the equation (15) would have little practical effect, so the impregnation mold should meet the following condition,

\[
\frac{2T}{WD} - P_a > 0
\]  

Consequently, it is necessary to optimize the fiber bundle width in practical application.

4. **Results and discussion**

4.1. **Effect of coated angle on the mechanical properties of composites**

By the melt impregnation mold and impregnation mechanism, continuous carbon fiber reinforced nylon 66 prepreg was prepared. The effect of configuration of impregnation pins on coated angle was studied by theoretical analysis. The effect of coated angle on the mechanical properties of fiber reinforced thermoplastic composites was also researched experimentally and compared with the theory of melt impregnation.

4.1.1. **Effect of configuration of impregnation pins on coated angle.** According to the different configurations of impregnation pins, the total coated angle and average coated angle could be obtained. The analysis results were shown in Table 1 with different the configuration of impregnation pins in Figure 7.
Table 1. Effect of configuration of impregnation pins on coated angle

| Configuration of impregnation pins | a  | b  | c  | d  | e  | f  |
|-----------------------------------|----|----|----|----|----|----|
| Total coated angle/°              | 0  | 43 | 58 | 92 | 276| 416|
| Average coated angle/°            | 0  | 11 | 19 | 31 | 40 | 60 |

Figure 7. The configuration of impregnation pins

Under the action of a certain tension, the fiber bundles would repeatedly collapse when it passed through the impregnation pins so that the fiber bundles could be dispersed on the surface of impregnation pins. The larger the coated angle between fiber bundles and impregnation pins, the greater the dispersion effect. When the coated angle was zero, there would be a fiber accumulation phenomenon, which resulted in a poor impregnation effect.

4.1.2. Effect of coating angle on mechanical properties of composites

Figure 8. The influence of coating angle on tensile properties

Figure 8 was the influence of coated angle on tensile properties of composites. It can be seen from the figure that the tensile properties of carbon fiber reinforced nylon 66 composites improved with the increase of the coated angle. When the coated angle raised from 0° to 11°, the tensile strength increased.
from 107MPa to 130MPa, which improved by 21.5%. When the coated angle was increased to 60°, the corresponding tensile strength was 214MPa, which improved by 100%.

Figure 9. The influence of coated angle on flexural properties

According to the Figure 9, it can be found that the flexural properties of carbon fiber reinforced nylon 66 composites improved with the increase of the coated angle. When the coated angle raised from 0° to 11°, the bending strength increased from 107MPa to 130MPa, which improved by 10.5%. When the coated angle was increased to 60°, the bending strength was 290MPa, which improved by 91%.

With the increase of the value of coated angle and the configuration changes of impregnation pins, the fiber bundles across the surface of the impregnation pins were dispersed, which improved the mechanical properties of composites.

4.1.3. Theoretical model validation of coated angle. The line speed of the fiber was 0.09m/s and the pin diameter was 8mm in the experiment.

Figure 10. The influence of the coated angle for the degree of impregnation and tensile properties.
As shown in Figure 10, the influence of the coated angle on the degree of impregnation and tensile properties was described. It can be seen that the degree of impregnation increased with high coated angle. Corresponding, there was also a continual increase in tensile properties as the degree of impregnation. Through the comparison between theoretical analysis and contrast test, it was proved that the theoretical analysis and theoretical model had rationality.

4.2. Effect of impregnation mold temperature on mechanical properties of composites

To study the influence of the temperature of impregnation mold on the mechanical properties of carbon fiber reinforced nylon 66 composites (CF/PA66), screw speed with 75r/min, the feed rate with 12r/min, traction speed with 5.4r/min and pressure maintaining time with 30s were set in the experiment. Five groups of different temperature of twin-screw (area 1 to area 4) and impregnation mold (area 5 to area 7) were selected, as shown in Table 2:

| Area 1 | Area 2 | Area 3 | Area 4 | Area 5 | Area 6 | Area 7 |
|--------|--------|--------|--------|--------|--------|--------|
| 1      | 250    | 255    | 260    | 265    | 270    | 270    |
| 2      | 250    | 260    | 270    | 275    | 280    | 280    |
| 3      | 260    | 270    | 275    | 285    | 290    | 290    |
| 4      | 260    | 280    | 290    | 295    | 300    | 300    |
| 5      | 260    | 280    | 290    | 300    | 310    | 310    |

Table 2. The temperature of twin screw and impregnation mold/℃.

Figure 11 illustrated the influence of impregnation mold temperature on the tensile properties of composites. The figure revealed that the tensile properties of the carbon fiber reinforced nylon 66 increased with the enhance of the temperature of the impregnation mold firstly and then tend to be stable. When the mold temperature increased from 270℃ to 290℃, the tensile strength increased from 136MPa to 214MPa, which improved by 57%. When the impregnation mold temperature was 300℃ and 310℃, its corresponding tensile strength was 213MPa and 215MPa respectively, tensile strength almost no changes. The tensile strength of the composites reached the maximum and it was in a stable state when the temperature of the impregnation mold was 290℃, just as shown in Figure 11.
The influence of impregnation mold temperature on the flexural properties of composites was shown in Figure 12. The figure showed that the bending strength of the carbon fiber reinforced nylon 66 increased with the rose of the temperature of the impregnation mold and then fluctuation within a certain range. The bending strength increased from 241 MPa to 317 MPa, which improved by 48% when the impregnation mold temperature enhanced from 270°C to 300°C. When the impregnation mold temperature was 310°C, the corresponding bending strength was 310 MPa, bending strength almost no changes. It can be known from Figure 8 that the bending strength of the composites reached the maximum when the impregnation mold temperature was 300°C.

![Figure 12](image)

**Figure 12.** The influence of the impregnation mold temperature on flexural properties.

Figure 13 revealed the influence of impregnation mold temperature on the impact properties of the composites. As shown in the Figure 13, the impact strength of carbon fiber reinforced nylon 66 increased firstly and then decreased with the rose of the temperature of impregnation mold and finally tended to be gentle. When the mold temperature increased from 270°C to 290°C, the impact strength increased from 35.6 KJ/m² to 46.5 KJ/m², which improved by 30.6%. Continue to improve the impregnation mold temperature, the impact strength increased on the contrary. It can be concluded from Figure 13 that the impact strength of the composites reached the maximum when the impregnation mold temperature was 290°C.
Figure 13. The influence of the impregnation mold temperature on impact properties.

Using differential scanning calorimetry measured nylon 66, the melting point was 262.576°C, so the melt impregnation mold temperature should be higher than the melting point of nylon 66. The comprehensive compared the effects of the impregnation mold temperature on tensile properties, flexural properties and impact properties of composites, the optimum temperature of melt impregnation mold should be between 290°C and 300°C.

5. Conclusions
This paper studied the process of multi-rollers impregnation of fiber reinforced composites and a model was developed to estimate the degree of the impregnation when the fiber symmetrically pulled over a stable pin. Many factors were took into consideration, including the pin diameter, the fiber line speed, the fiber bundle width, fiber pretension, polymer viscosity and the permeability coefficient of fiber structure. The results showed that the maximum impregnation degree was not related to the fiber pretension and the pin diameter but highly depended on the fiber bundle width. The optimum mold temperature was in the range of 290°C~300°C, while continuous carbon fiber reinforced nylon 66 composites obtained by melt impregnation method had better mechanical properties. The results had satisfactorily corroborated by experiment.

Acknowledgements
The authors would like to acknowledge the support of the Special Fund of the Central University Research of China (XK1518).

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