Long fiber thermoplastic pellets of glass fiber/polypropylene from pultrusion process

Ponlapath Tipboonsri, Voraya Wattanahitsiri and Anin Memon*  
Department of Industrial Engineering, Faculty of Engineering, Rajamangala University of Technology Thanyaburi, Pathumthani 12110, Thailand  
*E-mail: anin.m@en.rmutt.ac.th

Abstract. Long fiber thermoplastic pellets (LFTP) are composite materials that consist of thermoplastic and discontinuous reinforcement fibers with an aspect ratio more than critical aspect ratio. LFTP are high modulus, high strength and excellent impact resistance. Normally, commercial LFTP were produced by hot melt impregnation process or combinding between pultrusion and extrusion process. In this research, Thermoplastic pultrusion process was used for production of LFTP. The thermoplastic pultrusion process is uncomplicated process and machine less than hot melt impregnation process. The glass fibers were used in LFTP as a reinforcement and polypropylene (PP) fibers were used as a matrix of LFTP. General full factorial design of experiment was used to determine the optimal condition for un-impregnation on specimen from pultrusion process of LFTP. The study focused on three factors including the filling ratio, the molding temperature and pulling speed. The specimens were investigated on impregnation of resin in LFTP by microscope. The results show that un-impregnation decreased with an increase of filling ratio. Un-impregnation decreased with an increase of molding temperature. Finally, un-impregnation had increased with an increase of pulling speed. However, the void content has a little effect to molding temperature and pulling speed, or they have not almost effect. From statistical analysis, the optimal condition for the pultrusion process of LFTP was filling ratio of 104.73 %, molding temperature of 230 °C and pulling speed of 10 cm/min, which was given the lowest un-impregnation value of 5.85 %

1. Introduction  
Currently, there are many composite material applications, and there is a tendency to increase in the future for automotive, aviation and industrial parts. Composite material is combined of 2 phase as reinforcement and matrix. Matrix is bonded between reinforced material and matrix, and it transfers force to reinforced material [1]. Normally, matrix can be classified into 3 types included polymer, metal and ceramic [2]. Reinforcement can be classified of many patterns: particle pattern such as powder and flake, fiber pattern such as short fiber, long fiber and continuous fiber [3]. The difference fiber length has affected with different properties of composite. Longer fiber has affected to better properties, but production is more difficult than shorter fiber. Fabrication of composites has many processes, it depends on matrix and reinforcement such as injection molding, compression molding, filament winding, hand lay-up, vacuum foaming and pultrusion process, etc [4].

Injection molding of composite material reinforced by fiber, can easily be fabricated by using a short fiber. The using of short fiber in composite, it has lower mechanical properties to compare with a long fiber. Short fiber pellets can be easily mixed and injected, with a better fiber dispersion. The using of
long fiber in injection molding has a problem of production, because long fiber is difficult for mixing and dispersion of products. Currently, the long fiber reinforced plastic pellets (LFRP) are developed for injection molding by pellets form, with a diameter of 2-4 mm and a length of 6-12 mm [5, 6]. The fabrication of long fiber thermoplastic pellets (LFTP) and continuous fiber can be pulled through the impregnation die. Molten plastic is extruded from extruder to impregnation die, and impregnate to reinforced fiber. After that it is cooled and cut with a required length. This process is called the hot melt impregnation process. The hot melt impregnation process often found a problem of impregnation of thermoplastic resin in fiber, because thermoplastic melt has a high viscosity and difficult for impregnation. In addition, the extruder is used in the hot melt impregnation process [5] that make a high cost of machine and production.

Therefore, there are interested to develop the process of long fiber thermoplastic pellets (LFTP) by using the pultrusion process, which has higher impregnated quality and lower cost than ordinary process. The pultrusion process, continuous fibers and thermoplastic matrix fibers are pulled through a constant cross-section hot die. Thermoplastic is melted and impregnated with a reinforced fiber, after that they are pulled out of die become continuous composite material. From the literature reviews, the pultrusion process can be used to fabricate a continuous composite material, which has impregnation of resin in fiber around 80-93% [7]. The pultrusion parameters consist of filling ratio, molding temperature and pulling speed [7, 8]. This research presents the effects of filling ratio, molding speed, and pulling speed on the quality of long fiber thermoplastic pellets. The glass fibers were used in LFTP due to high mechanical properties, continuous fiber form and low cost. Polypropylene fibers (PP) were used as a matrix of LFTP due to high mechanical, low density, low water absorption, low process temperature and low cost.

2. Materials and methods

2.1. Materials

Glass fibers (GF) were paralleled with PP fibers. The glass fiber was selected as the main reinforcing fiber (1200 tex with a density of 2.620 g/cm³). Polypropylene yarns (133 tex with a density of 0.946 g/cm³) were used as matrix fibers in long fiber pellets, which have melting temperature at 178.8 °C from differential scanning calorimetry (DSC) result as shown in figure 1 (a), and they have degradation temperature around 320 °C from thermal gravimetric analysis (TGA) result as shown in figure 1 (b). The volume fractions of fiber and matrix were defined by adjusting the filling ratio as presented in equation (1).

![Figure 1.](image) (a) DSC curve of polypropylene and (b) TGA curve of polypropylene.
2.2. Methods

Long fiber thermoplastic pellets were fabricated by thermoplastic pultrusion process as shown in figure 2. This research was finding the filling ratio, molding temperature and pulling speed that make to best impregnation quality, or un-impregnation is nearly 0 %. The pultrusion parameters consisted of filling ratio, molding temperature and pulling speed. The preliminary, TGA and DSC results, molding temperatures of 200, 210, 220 and 230 °C were selected to design by DOE (Design of experiment). The pulling speeds were varied as 6, 10, 14, 18 and 22 Hz of motor frequency or 10, 20, 30, 40 and 50 cm/min, respectively. The filling ratios were varied as 100.73 and 104.73 %, and volume fractions of polypropylene and glass fiber are shown in table 1. The pulling speed and filling ratio were varied as shown in table 2. The molding die consist of heater 6 zones and the molding temperatures are controlled at zone 3 and 4 as shown in figure 2. Meanwhile, heater at zone 1 and 2 are set below the melting point of PP, zone 5 and 6 are set below the melting point as well for cooling down the part. The volume fractions of fiber and matrix were defined by adjusting the filling ratio [7] as presented in equation (1). After that the cross-section of specimens was investigated impregnation and void content by microscope, and it is analysed by imageJ software. Minitab software was used to analyse a relation between molding temperature, pulling speed and filling ratio.

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\text{Filling ratio} = \frac{\text{Cross-section of materials}}{\text{Cross-section of die}} \times 100 \quad (1)
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Figure 2. Schematic of thermoplastic pultrusion process for LFTP production.
Table 1. Volume fraction of long fiber thermoplastic pellets.

| Filling ratio (%) | VF of matrix (%) | VF of GF (%) |
|-------------------|------------------|--------------|
| 100.74            | 87.87            | 12.87        |
| 104.73            | 92.35            | 12.38        |

2.3. Design of experiments (DOE)
The optimization study of LFPT from pultrusion process was designed and analyzed by using statistical program Minitab 18 software (Licensee : Rajamangala University of Technology Thanyaburi and License ID is 90b4-43F5-A70b-443d-1d51-97d). The 3 factors consisted of filling ratio, molding speed and pulling speed were selected for optimization analysis of un-impregnation (Y). The general full factorial experiment, which generated from statistical program. The design consists of 2 level of filling ratio, 4 level of molding speed and 5 level of pulling speed as shown in table 2. There were 40 distinct experiments with 5 replicates. Therefore, the total of experiments were 200 runs. In addition, the results were used to analyze the effect of those factors upon un-impregnation for both main effect and interaction. The hypothesis testing was used for investigation between 3 factors and un-impregnation, which H0 : factors have affected to un-impregnation, H1 : factors have not affected to un-impregnation. Significance level (α) of this experiment was determined at 0.05.

Table 2. Factorial design of LFTP production.

| Factor            | Set up values           |
|-------------------|-------------------------|
| Filling ratio     | 100.74, 104.73 %        |
| Molding temperature| 200, 210, 220, 230 °C    |
| Pulling speed     | 10, 20, 30, 40, 50 cm/min|

2.4. Microstructure analysis
The microstructures were conducted on an Olympus microscope. They were investigated at 12.5 times magnification. The microstructure was carried out on void and un-impregnation analysis. The void contents can be calculated by void contents and cross-section area, and un-impregnation can be calculated by space area in the fiber bundle and fiber bundle area as shown in figure 3. The impregnation quality of LFTP was classified as shown in figure 4.
Figure 3. (a) Cross-section of specimen with void contents (b) Analysis of void contents by imageJ, (c) Cross-section of specimen with un-impregnation and (d) Analysis of un-impregnation by imageJ.

Figure 4. Good impregnation quality of LFTP cross-section.
3. Results and discussions
In this experiment, the long fiber thermoplastic pellets were conducted with varied molding temperature, pulling speed and filling ratio by pultrusion process. Molding temperature, pulling speed and filling ratio have been affected with an impregnation quality. They are possible to be affected with mechanical properties, when they are molded by other process. In the design of experiments, 3 factors of filling ratio, molding and pulling were designed to 40 conditions, run with 5 replicates.

The relation between un-impregnation, void content and molding temperature with a pulling speed of 30 cm/min are shown in figure 5 (a). It was found that un-impregnation of 100.74% filling ratio had a tendency to decrease with an increase of molding temperature. Un-impregnation values of 100.74% filling ratio of 200, 210, 220 and 230 °C were 31.70, 28.58, 27.11 and 25.27 %, respectively. Un-impregnation of 104.73% filling ratio had a tendency to decrease with an increase of molding temperature, which un-impregnation values of 200, 210, 220 and 230 °C are 12.72, 11.17, 10.85 and 10.80 %, respectively. The obtained results are in accordance with literatures [7, 9]. High molding temperature has made a lower viscosity of thermoplastic, which resin can be easily impregnated. In figure 6 confirmed the melt flow index of PP with 180 to 260 °C, which increased with an increase of molding temperature. The high melt flow index means that low viscosity [10, 11]. Relation between un-impregnation and filling ratio, it can be obviously seen un-impregnation of 104.73 % filling ratio lower than 100.74 % filling ratio. Filling ratio had highly affected to un-impregnation due to the increase of PP volume fraction, it can impregnate more than lower filling ratio. There are the directions in accordance with the literature [7]. The molding temperature had a little effect to void content. The void content at 100.74 % filling ratio of 200, 210, 220 and 230 °C were 0.11, 0.10, 0.09 and 0.10 %. The void content at 104.73 % filling ratio of 200, 210, 220 and 230 °C were 0.03, 0.01, 0.02 and 0.01 %. They may occur from the air inside glass fiber that can not be removed while polypropylene was impregnating to the glass fiber.

The relation between un-impregnation, void content and pulling speed of molding temperature at 210 °C are shown in figure 5 (b). The un-impregnation of 100.74 filling ratio had tendency to increase with an increase of pulling speed, which un-impregnation values are 23.80, 24.41, 28.58, 29.43 and 30.89 % at pulling speed of 10, 20, 30, 40 and 50 cm/min. The un-impregnation of 104.73 filling ratio had tendency to increase with an increase of pulling speed, which un-impregnation values are 8.03, 8.85, 11.17, 12.27 and 13.92 %. The decrease of pulling speed had affected to the decrease of un-impregnation due to the intermediate materials molded for a long time in the die which helps resin to impregnate for a longer time. There are the directions in accordance with the literature [7] of un-impregnation results. The filling ratio had highly affected to un-impregnation as same as previous results. In addition, the void content had a little effect to un-impregnation as same as previous results. The void contents at 100.74 % filling ratio were 0.09, 0.09, 0.05, 0.07 and 0.05 % at pulling speed of 10, 20, 30, 40 and 50 cm/min, respectively. The void contents at 104.73 % filling ratio were 0.02, 0.02, 0.01, 0.02 and 0.02 %.
Figure 5. Relation between (a) Un-impregnation, void content with molding temperature and (b) Un-impregnation, void content and pulling speed.

Figure 6. Melt flow index results with varied molding temperatures.

The un-impregnation results were used to analyze an interaction term of 3 factors. In addition, the main effect plot and optimize conditions were analyzed by Minitab 18. Plot of residuals versus predicted and normality plot for un-impregnation are shown in figure 7 (a) and (b), and it was found that the residuals follow normal distribution. Variances are constant as according with the literatures [12, 13]. Significance level (α) was determined at 0.05. All independent and interaction terms were influenced to un-impregnation. The analysis of variance results is shown in table 3. The hypothesis testing, P-value of 2-way interaction and 3-way interaction were over 0.05 of Significance level (α) that mean P-value is not significant with H₀ or all factors are independent [12]. Main effects of 3 factors for un-impregnation are shown in figure 8. Un-impregnation decreased with an increase of filling ratio. Un-impregnation decreased with an increase of molding temperature. Finally, the un-impregnation increased with an increase of pulling speed. The optimization of un-impregnation from Minitab was filling ratio of 104.73 %, molding temperature of 230 °C and pulling speed of 10 cm/min, which the lowest un-impregnation is 5.85 %. From the result, we can predict the un-impregnation with factor of filling ratio, molding speed and pulling speed as presented in equation (2). The coefficient of factors is shown in table 4.
Figure 7. (a) Normal probability plot and (b) Versus fits plot of un-impregnation for LFTP.

Table 3. Analysis of variance for LFTP.

| Source                        | DF | Seq SS   | Adj SS   | Adj MS   | F-value | P-value |
|-------------------------------|----|----------|----------|----------|---------|---------|
| Filling ratio                 | 1  | 13684.71 | 13684.71 | 13684.71 | 401.84  | 0.000   |
| Molding temperature           | 3  | 416.79   | 416.79   | 138.93   | 4.08    | 0.008   |
| Pulling speed                 | 4  | 1198.17  | 1198.17  | 299.54   | 8.80    | 0.000   |
| Filling ratio*Molding temperature | 3  | 76.21    | 76.21    | 25.40    | 0.75    | 0.526   |
| Filling ratio*Pulling speed   | 4  | 16.92    | 16.92    | 4.23     | 0.12    | 0.974   |
| Molding temperature*Pulling speed | 12 | 22.40    | 22.40    | 1.87     | 0.05    | 1.000   |
| Filling ratio*Molding temperature*Pulling speed | 12 | 45.71    | 45.71    | 3.81     | 0.11    | 1.000   |
| Error                         | 160| 5448.78  | 5448.78  | 34.05    |         |         |
| Total                         | 199|          |          |          |         |         |

S = 5.83565   R-Sq = 73.94%   R-Sq(adj) = 67.59%

Figure 8. Main effect plot of un-impregnation.
Y = b_0 + b_1X_1 + b_2X_2 + b_3X_3 \quad (2)

When Y is response variable (Un-impregnation), b_0 is constant, b_1 is filling ratio coefficient, b_2 is molding temperature coefficient, b_3 is pulling speed coefficient and X_1, X_2, X_3 are coded values of variables, filling ratio (X_1), molding temperature (X_2), pulling speed (X_3), respectively.

| Predictor             | Coef  | Se Coef | T     | P  |
|-----------------------|-------|--------|-------|----|
| Constant              | 465.46| 20.97  | 22.19 | 0.000 |
| Filling ratio         | -4.1463| 0.1911 | -21.70| 0.000 |
| Molding temperature   | -0.11777| 0.03409 | -3.45 | 0.001 |
| Pulling speed         | 0.17196 | 0.02695 | 6.38 | 0.000 |

### 4. Conclusion

In this study, the fabrication of PP reinforced with a long glass fiber by pultrusion process, filling ratio, molding temperature and pulling speed were studied by investigation of un-impregnation and void content from microscale. They are possible to effect with mechanical properties when using by other processes. From the studied results, the molding temperature had directly affected to un-impregnation quality. The un-impregnation deceased at 210 °C, and it increased after 210 °C of molding temperature. Relation between filling and un-impregnation, un-impregnation decreased with an increase of filling ratio. Relation between pulling speed and un-impregnation, un-impregnation increased with an increase of pulling speed. However, the void content had a little effect to molding temperature and pulling speed, or they have not almost effect. From Minitab results, the response optimization of DOE was pilling ratio of 104.73 %, molding temperature of 230 °C and pulling speed of 10 cm/min, which the lowest un-impregnation is 5.85 %. From the regression equation, the lowest un-impregnation is 5.85 = 465.46 + (-4.1463*104.73) + (-0.11777*230) + (0.17196*10).

However, this research was preliminary of relation filling ratio, molding temperature and pulling speed with un-impregnation and void content. Un-impregnation of 0% was targeted for future work. Mechanical properties were expected to increase with a decrease of un-impregnation. Lowest un-impregnation is just only 5.85 %. The optimization of condition in this paper can be used to produce a LFTP for injection molding or compression molding.

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### References

[1] Nijssen R P L 2015 Composite Materials: An Introduction (Amsterdam: A VKCN publication) chapter 1 pp 13–38

[2] Ibrahim I D, Jamiru T, Sadiku R E, Kupolati W K, Agwuncha S C and Ekundayo G 2015 J. Reinf. Plast. Comp. 34 1347–56

[3] Nirmal U, Hashim J and Ahmad M M H M 2015 Tribol. Int. 83 77–104

[4] Adrian P P and Gheorghe B M 2010 AUO-FMTE IX (XIX) 3.1–3.6

[5] Ning H, Lu N, Hassen A A, Chawla K, Selim M and Pillay S 2019 Int. Mater. Rev. 65 1–25

[6] Tan Y, Wang X and Wu D 2015 J. Reinf. Plast. Comp. 34 1804–20

[7] Memon A and Nakai A 2013 Energy Procedia 34 818–29

[8] Bindal A, Singh S, Batra N K and Khanna R 2013 Indian J. Eng. Mater. Sci. 2013 1–6

[9] Angelov I, Wiedmer S, Evstatiev M, Friedrich K and Mennig G 2007 Compos. Part A Appl. Sci. Manuf. 38 1431–8

[10] Lan H-Y and Tseng H-C 2003 J. Chin. Inst. Chem. Engrs. 34 405–15
[11] Mohamed M E R 2014 *Viscosity Measurement by Using Melt Flow Index for Thermoplastic Polymers* (Sudan: Sudan University of Science and Technology College) chapter 4 pp 41–51
[12] Mat Kandar M I and Akil H M 2016 *Procedia Chem.* **19** 433–40
[13] Suresh S and Senthil Kumar V S 2014 *Procedia Eng.* **97** 632–41