Continuous Manufacturing Technology of Stampable Thick Organo-sheet Using Fixed Rollers Double Belt Press

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ABSTRACT

Fixed rollers double belt press is an effective solution for the mass production of stampable thick organo-sheet. This system enables the continuous impregnation of multi-layers of carbon fiber fabrics with molten thermoplastic under the multiple rollers. For example, we can produce 6-mm thick organo-sheet continuously using 28 layers of carbon fiber fabrics and polyamide 6 resin films. The key point of this system is the optimization of process parameters such as belt speed, heating temperature and steel belt gaps. However, the impregnation mechanism under rollers is complicated compared to a hydraulic press system. Both impregnation of fabric and in-plane resin flow occurs with the change of pressure under rollers. In this study, this mechanism was investigated theoretically and experimentally under different process conditions and the prediction model was developed. The results will be applied to guide the optimization of the manufacturing process.

1. INTRODUCTION

Thermoplastic composite sheet, so-called organo-sheet, is a promising material for the aeronautic, industrial and automobile parts, because it will enable the short cycle production by stamp forming and welding [1, 2]. The key issues for manufacturing organo-sheet are the impregnation quality and the low cost process. However, it is difficult to achieve these properties at the same time because the impregnation of fabric with thermoplastic resin is difficult due to its high viscosity in molten state. Therefore, impregnation process needs high temperature, long time and much pressure, which makes the production cost high.

A double belt press has been used for the production of pre-preg sheet and the consolidation of the laminates of pre-pregs. The machine consists of the heat and pressure zone for impregnation and the subsequent cooling zone for consolidation. The continuous process is possible using this system without the long time to heat up and cool down the mold. In order to achieve the efficient process with double belt press, the process parameters should be optimized based on the understanding of the impregnation mechanism. Several investigations have been reported about double belt press process [3, 4, 5]. Several authors discussed the influence of the process parameters such as belt speeds, pressure and heating temperature on the impregnation quality and the mechanical property.

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We used a double belt press with a fixed rollers system in this study. This machine has several fixed rollers in the heating zone where the minimum gaps can be controlled between the vertical rollers. This enables the production of thick composite sheet made of multi-layers of dry fabrics and thermoplastic matrix resin, because the sequential belt gaps make the desired change of the material thickness during impregnation process. This is an advantage compared to a hydraulic press system. However, the impregnation mechanism under rollers is much complicated. Under rollers, both of through-thickness impregnation and in-plane resin flow occur and the resulting pressure is determined by the balance of these flows.

In this paper, we studied the impregnation mechanism of fixed-rollers double belt press using carbon fiber woven fabrics and polyamide 6 resin films. The theoretical impregnation model was developed to guide the optimization of double belt press process. Finally, we demonstrated the production of 6mm thick organo-sheet with the optimized process parameters.

2. EXPERIMENTATION

2.1 Materials

Carbon fiber plain woven fabrics (T700SC-12K, 3.27 yarns / 25 mm, 200 g/m^2, Toray co., ltd.) and polyamide 6 resin films (CM1006, Toray co., ltd.) were used in this study. The melted resin viscosity was measured with a parallel-plate rheometer in the range from 225 ºC to 290 ºC.

2.2 Double belt press machine

The schematic of double belt press (PROCESS SYSTEMS K. K.) is shown in Figure 1. The machine has two heat zones with eight pressure rollers per zone and subsequent one cooling zone with hydraulic pressure. The steel belt width is 600 mm. However, the effective width of good quality area of the product sheet is 490 mm due to the available width of cooling pressure. The heat is provided by the hot air blowers. The maximum temperature of hot air is 380 ºC. The heat transfers to the materials through the steel belt during the process. The vertical positions of rollers can be fixed at the inlet and the outlet of each zone. The loads are applied on the roller boxes. The set-up process parameters are the hot air temperatures, the loads applied on the rollers, the cooling fluid pressure, the steel belt gaps and the moving belt speed. The important issue is that these process parameters do not correspond to the parameters of the normal hot press process. Therefore, one needs to evaluate the effect of each parameter on the production process.

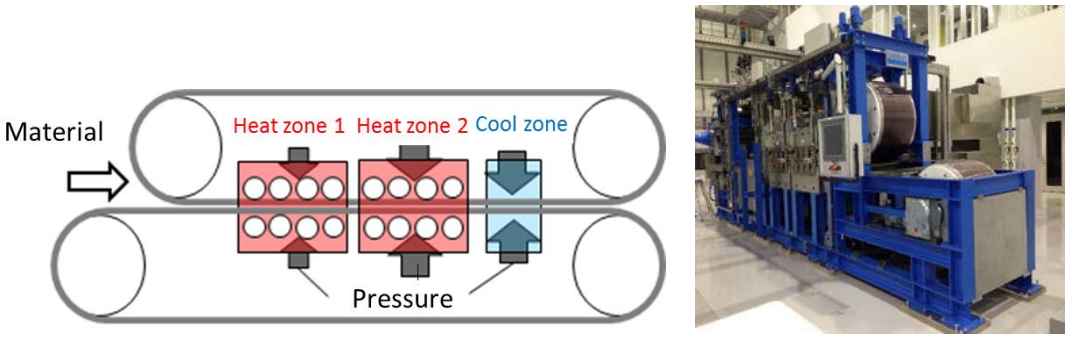


Figure 1. Schematic of fixed-rollers double belt press.

2.3 Measurement of pressure under rollers

A force sensor (9133B, Kistler) was mounted in the hole of aluminum plate (300 mm × 500 mm × 5 mm). The sensor was connected to a data logger with a cable. 5 layers of carbon fiber fabrics and 6 layers of polyethylene films (thickness: 100 μm) were stacked alternately and placed over the sensor. Polyethylene films were used in order to obtain the molten state of resin in the limited operating temperature range of the force sensor. It was processed with the double belt press at varied heating temperatures such as room temperature, 120 ºC and 140 ºC. The belt speed was 0.2 m/min. All belt gaps were fixed at 6.4 mm. The measured force was recorded with the logger at a time step of 0.1 s during the process.

2.4 Compression molding experiments

Compression molding tests were conducted using a small hydraulic press machine. 8 layers of carbon fiber woven fabrics and 9 layers of polyamide 6 films (thickness: 100μm) were stacked alternately. The material size was 150 mm × 150 mm. It was placed on the mold and heated up to the target temperature. The temperature was monitored with a thermocouple inserted in the middle layer of the material. The material was processed at a temperature of 245 ºC and at a pressure of 0.78 MPa. The holding time was varied from 30 s to 480 s. The material was finally cooled down to room temperature. A typical molding diagram is shown in Figure 2.

The impregnation quality of obtained composites was evaluated. The cross-section was polished and observed with microscopy. The area of impregnated region in a fiber bundle was measured with microscopy. The averaged impregnation length was defined as the value of the area divided by the width. 10 of fiber bundles were observed and the averaged impregnation length was calculated.

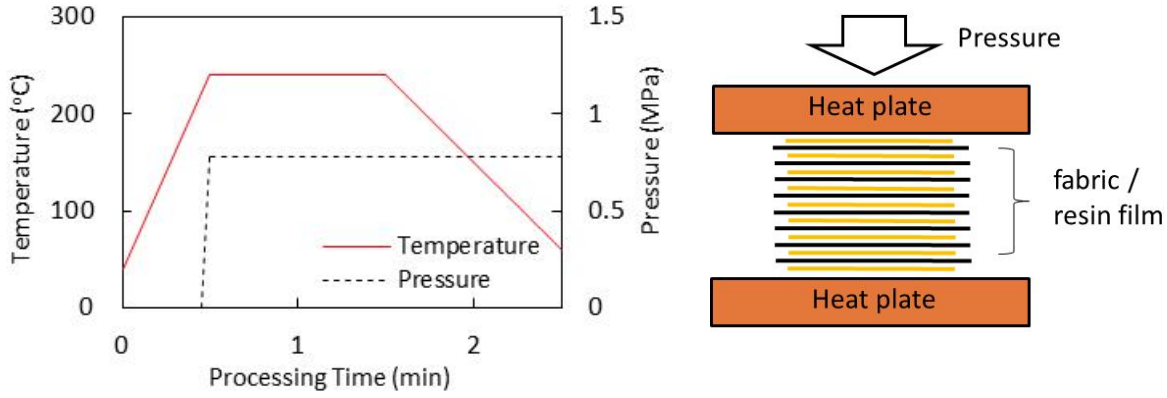


Figure 2. Typical press molding diagram.

2.5 Double belt press experiments

The double belt experiments were conducted to evaluate the influence of the belt speed on the impregnation quality of the composite sheets. 2 layers of carbon fiber fabrics and 3 layers of polyamide 6 films (thickness: 50 μm) were laid-up alternately and processed with double belt press. 50μm thickness films were used in order to prevent the in-plane flow of resin layers. From our experience of double belt press experiments, using thick resin films such as 100 μm often cause the in-plane resin flow and the resulting fiber movement, which would make it difficult to

evaluate the impregnation process accurately. The material size was 500 mm × 500 mm. The process parameters are shown in Table. 1. The belt speed was varied from 0.2 m/min to 0.6 m/min.

Table 1. Process parameters of double belt press experiments

Belt speed (m/min)	**Heat zone 1**			**Heat zone 2**			**Cool zone**
0.4, 0.6, 0.8	**Hot air (°C)**	**Roller (kN)**	**Gap (mm)**	**Hot air (°C)**	**Roller (kN)**	**Gap (mm)**	**Pressure (MPa)**
	320	6	0.55 – 0.45	320	24	0.4 – 0.35	1

2.6 Thick composite sheet production with double belt press

The composite sheet with a thickness of 6 mm was produced with double belt press. 28 layers of carbon fiber fabrics and 29 layers of polyamide 6 films (thickness: 100 μm) were used as the material. The material size was 550 mm × 2000 mm. The process parameters are shown in Table. 2.

Table 2. Process parameters of 6mm thickness composite sheet production

Belt speed (m/min)	**Heat zone 1**		**Heat zone 2**		**Cool zone**
0.2	**Hot air (°C)**	**Roller (kN)**	**Hot air (°C)**	**Roller (kN)**	**Pressure (MPa)**
	380	9	320	18	2

2.7 Evaluation of composites

Specimens were cut from the center area of the composite sheets. The cross-sections were polished and observed with microscopy in order to evaluate the degree of impregnation. Flexural properties of 6 mm thickness composite sheet were evaluated with three-point bending tests. The specimens parallel to the length direction and the width direction were cut from the center area of the composite sheet. The specimen size was 15 mm × 260 mm and the cross-head speed was 16 mm/min. 5 specimens were tested for each direction.

3. RESULTS

3.1 Results of pressure under rollers

Figure 3 shows the pressure profiles measured in double belt press at different temperatures. X-axis indicates the belt positions and each pressure peak corresponds to the position under the roller. It is thought that the deflection under a roller made the bell-shape profile of pressure. The peak value of the pressure profile decreased along with the process direction at 120 ºC and 140 ºC. It's because that the material thickness would decrease due to the progress of resin impregnation into fabrics. It should be noted that this result can't be applied directly to other matrix resin such as polyamide due to the difference in the viscoelasticity. At least, this result indicates the importance of the appropriate setting of belt gaps in order to keep enough pressure to achieve the good impregnation.

The impregnation process model under a roller is illustrated in Figure 4. As the belt gap decrease, a part of resin will flow into the fabric layer in the through-thickness direction. And other part of resin will flow in the in-plane direction. The resulting pressure is balanced by these flows behavior. Another important issue is the change of the permeability during the process. The permeability depends on the local fiber volume fraction, which could be affected by the change of pressure. Now we try to study this process using the simulation software, however, it's not in the scope of this study.

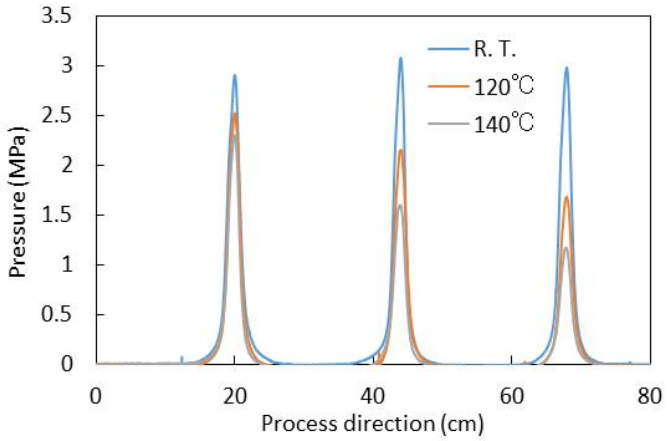


Figure 3. Pressure profiles under rollers in double belt press at various temperatures.

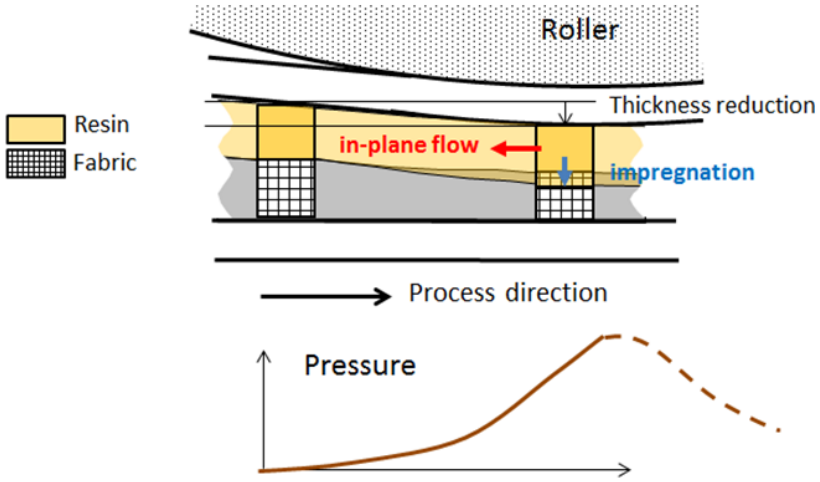
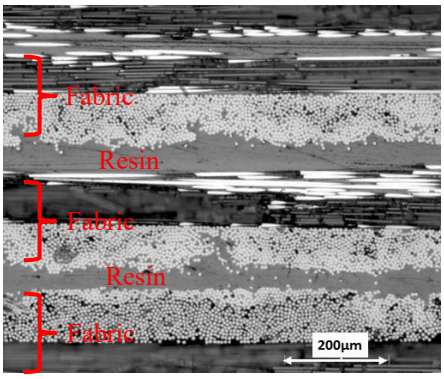


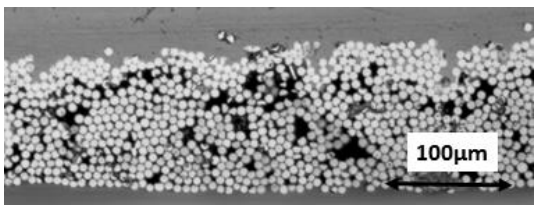
Figure 4. Impregnation and in-plane flow behavior under a roller.

3.2 Results of compression molding experiments

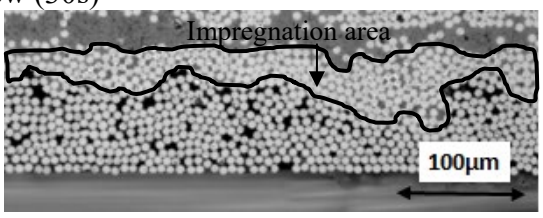
The cross-sectional photographs of the composites at different holding times are shown in Figure 5. The typical overview showed the laminated structure of fabric layers and resin film layers. The woven fabric layer is between the resin film layers. Therefore, the impregnation starts from the both surfaces of fabrics. Now we focus the impregnation in a fiber bundle. In this case, the impregnation in a fiber bundle seemed to start from the border with the resin layer as shown in Figure 5. As the holding time increased, the flow-front moved forward in the through-thickness direction. The fiber bundles were almost filled up at a holding time of 480 s.



Overview (30s)



5s



120s

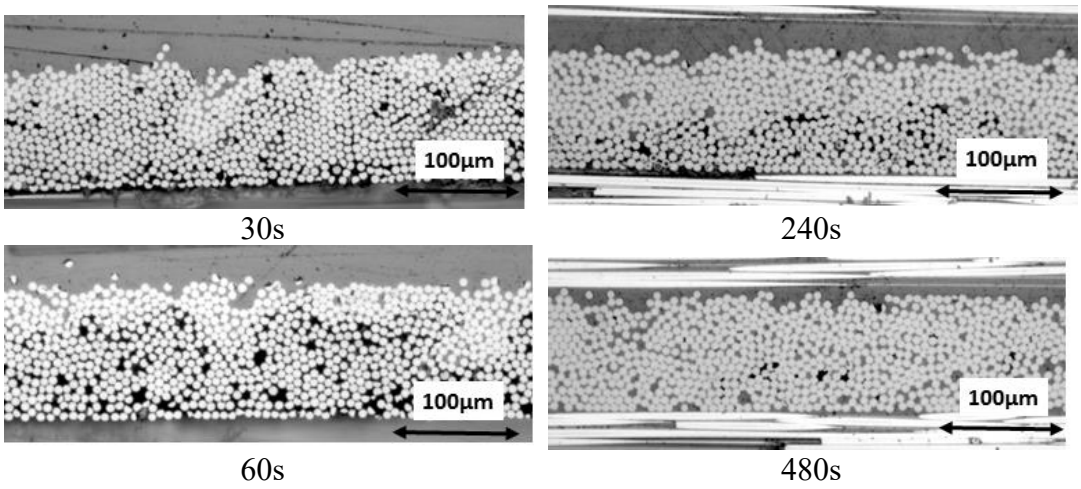


Figure 5. Cross-sectional photographs of composites at different holding times.

3.3 Analysis of permeability

Many kinds of investigations have been reported about the model of thermoplastic impregnation using Darcy's law [6, 7, 8]. The models have been modified depending on their process and conditions. As discussed in the previous section, the impregnation in a fiber bundle could be assumed as one directional flow in the through-thickness direction. For the simplicity, we assumed that the fiber volume fraction (v_f) was constant and homogeneous in the impregnated region during the process. Thus, Darcy's law can be formulated as the following simple equation.

$$\frac{dl}{dt} = \frac{K}{\mu}\frac{P}{l} \tag{1}$$

Here l is the impregnation length in a fiber bundle, K is the permeability, μ is the resin viscosity and P is the applied pressure. By integrating the equation over the process time t, we can obtain the following equation.

$$l^2 = 2KP\int \frac{1}{\mu}dt \tag{2}$$

The relationship between the temperature and the melt viscosity of polyamide 6 is shown in Figure 6. The experimental curve can be approximated with Arrhenius equation. Thus, the melt viscosity can be calculated for any temperature with the equation.

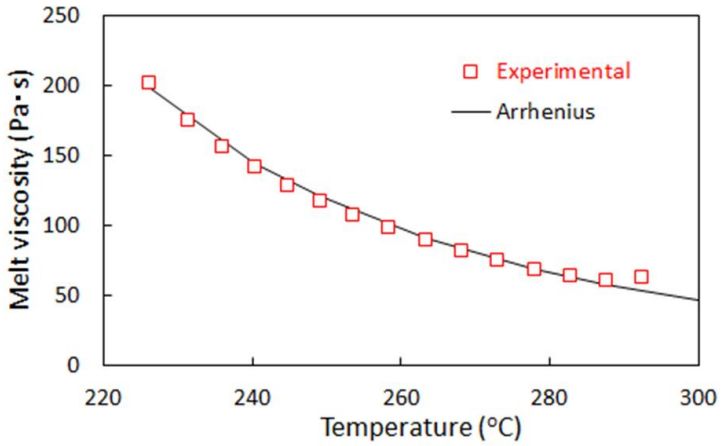


Figure 6. Relationship between temperature and melt viscosity of polyamide 6.

Figure 7 shows the impregnation length measured from the cross-sectional photographs of composites at different holding times. The calculated curve using equation (2) was fitted into this experimental curve and the permeability K was obtained. The value was 6.54×10^{-16} m^2 from this experiment.

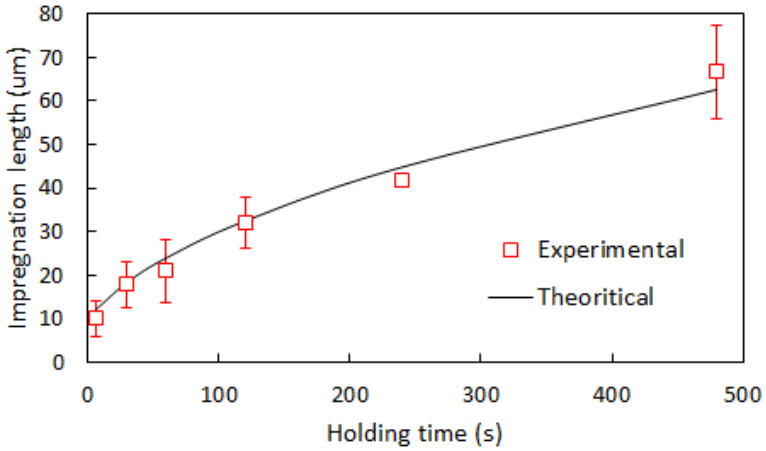


Figure 7. Impregnation length as a function of holding time.

3.4 Impregnation process model in double belt press

Figure 8 shows the material temperature profiles during the double belt press process at different belt speeds. The temperature started to increase at the inlet of heat zone 1, and went over the melting point, finally reached the maximum value at the outlet of heat zone 2. Then, it decreased quickly in cool zone. The temperature profile was quite different for each belt speed. We can consider the impregnation region over the melting point of 225 ºC. The result showed that this region is strongly affected by the belt speed.

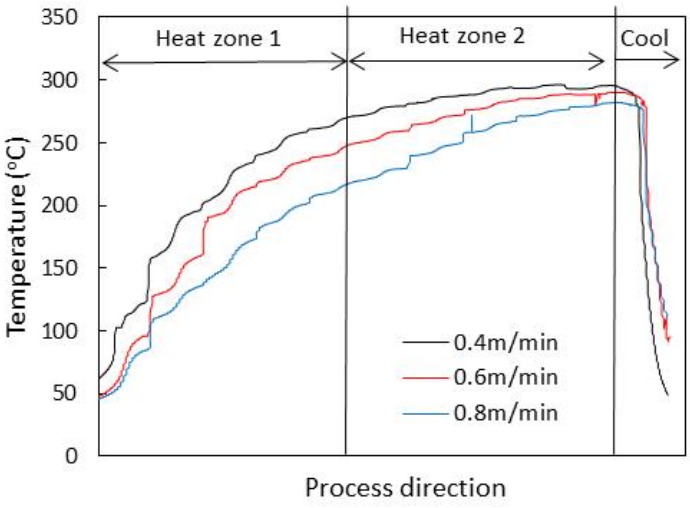


Figure 8. Material temperature profiles in double belt press at different belt speeds.

Figure 9 shows the material temperature profile and the corresponding pressure zones in the double belt press at a belt speed of 0.6 m/min. We can find five effective pressure zones under rollers for the impregnation. And also, the cooling fluid pressure can also be effective until it reaches the solidification temperature.

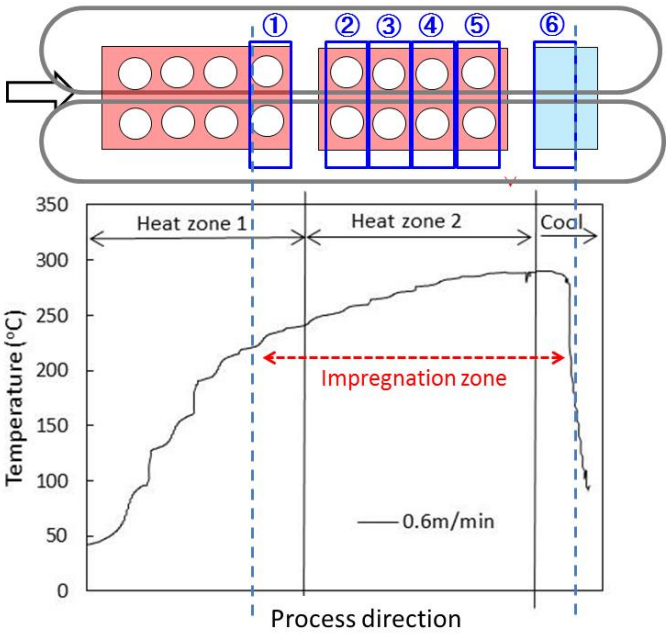


Figure 9. Material temperature profile and corresponding pressure zones in double belt press.

At first, we consider the impregnation process under a roller. In this case, the resin viscosity is assumed to be constant. However, the pressure changes significantly during the process. Darcy's equation (2) could be modified as the followings.

$$l^2 = \frac{2K}{\mu}\int Pdt \tag{3}$$

Here the pressure is integrated with time under a roller during the process. As discussed in the previous section, the actual pressure is determined by the balance of the impregnation flow and the in-plane resin flow. However, we simplified this problem in this model and assumed that the roller force was totally applied on resin without resin in-plane flow. Thus, equation (3) can be arranged as the following equation.

$$l^2 = \frac{2K}{\mu}\frac{1}{v}\int Pdx = \frac{2K}{\mu}\frac{F}{vw} \tag{4}$$

Here v is the belt speed, w is the material width and F is the roller force. We can apply equation (4) for the impregnation estimation under a roller. And also, we can apply equation (2) for the impregnation estimation under cooling fluid pressure. The total impregnation length L of the double belt press can be obtained by adding up each impregnation length l under the rollers and the cooling fluid pressure.

$$L^2 = \sum l^2 \tag{5}$$

3.5 Result of double belt press experiments and validation

Figure 10 shows the cross-sectional photographs of the composite sheets produced by double belt press at different belt speeds. Un-impregnated region was found in the center of a fiber bundle. This was a different phenomenon compared to the result of hydraulic hot press (Figure 5). The impregnation length was measured from the cross-sectional photographs. The experimental values were compared with the calculated values using equation (5) as shown in Figure 11. The theoretical values showed a good correlation with the experimental data. However, there is still some discrepancy. One of the possible reasons is that the theoretical model was simplified by neglecting the influence of resin in-plane flow and assuming that the acting pressure was just given by the roller force. Another point is that 50 μm thickness resin films were used for this experiment. This might cause some different impregnation phenomena from the compression molding results. Now in our research work, we combine the heat transfer theory and Darcy's model mentioned above, which leads to the simplified impregnation simulation for double belt press with the parameters of the process and the material information. It's not detailed in this paper.

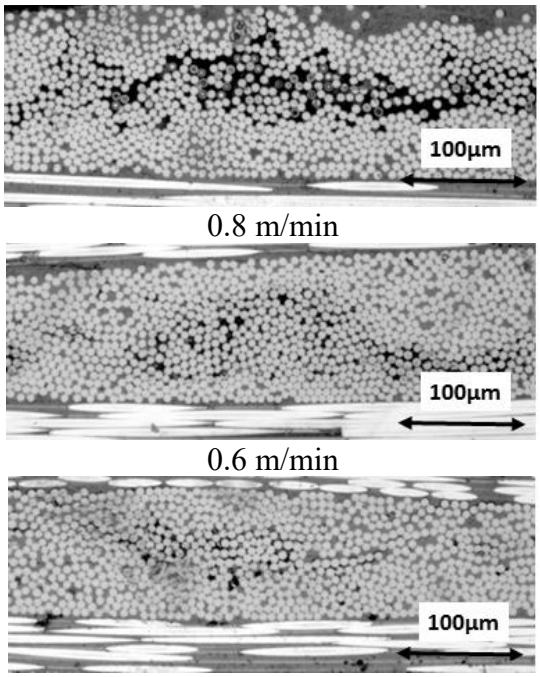


Figure 10. Cross-sectional photographs of composite sheets produced with double belt press at different belt speeds.

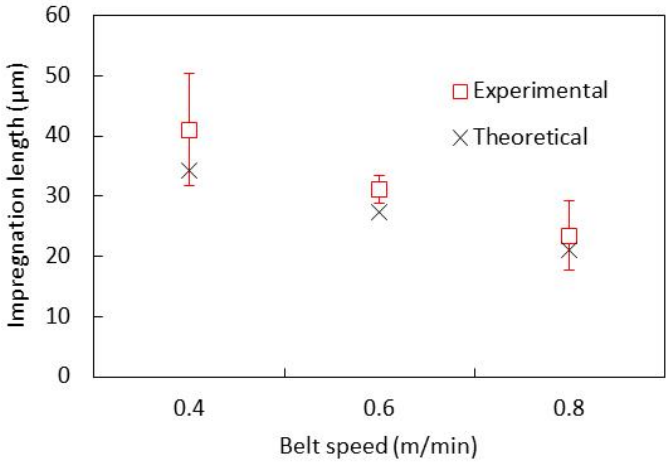


Fig. 11 Comparison of experimental and theoretical impregnation lengths at different belt speeds.

3.6 Result of 6 mm thickness composite sheet with double belt press

Figure 12 shows the photograph of 6 mm thickness composite sheet produced with double belt press. The product size is 550 mm × 2000 mm × 6 mm. The cross-sectional photograph of the composite sheet is also shown in Figure 12. The impregnation quality was good and few voids were found in there. The properties of composite are shown in Table 3. The averaged void fraction measured with the burn-off test was 1.4 %. The averaged bending strength was around 600 MPa and the bending modulus was around 60 GPa. The bending strength was almost 90

percent of the value achieved by the normal hot press process. In this case, the belt gap at the inlet of heat zone 1 was set to 8.5 mm of the lay-up thickness, and the belt gap at the outlet of heat zone 2 was set to 6mm of the product thickness. The hydraulic double belt press machine is not able to afford this thickness change, because that system generally uses the sealed fluid in order to generate the hydraulic pressure and the fluid would leak due to the radical thickness change. The applied belt speed was 0.2 m/min. It took a long time to heat up the thick-layer material compared to the thin-layer material for which we can apply the belt speed of 0.5 m/min. One of the other challenges is to develop the efficient pre-heating process for the thick material. It would help to achieve the affordable higher belt speed for the future mass-production process.

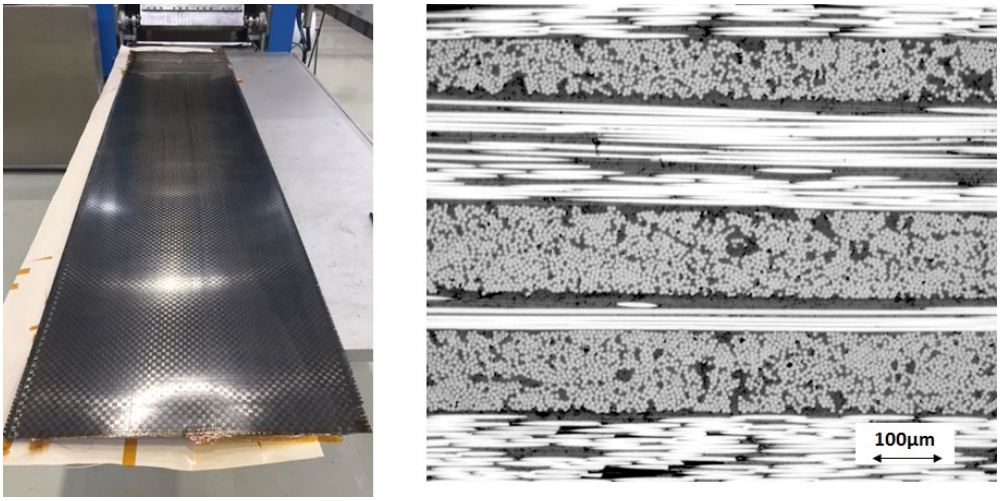


Figure 12. Photographs of 6 mm thickness composite sheet and its cross-sectional microscopy

Table 3. Properties of 6 mm thickness composite sheet

V_f	Void fraction	Bending strength	Bending modulus
55	1.4%	600MPa	60GPa

4. CONCLUSIONS

In this study, we showed the continuous manufacturing technology of thick organo-sheet with the fixed rollers double belt press. The used material was the multi-layers of carbon fiber woven fabrics and polyamide 6 resin films. At first, we investigated the impregnation process with hydraulic hot press and estimated the permeability based on Darcy's law. Then, we studied the mechanism of double belt press system and discussed the effect of the process parameters on the impregnation quality. The theoretical impregnation model of double belt press was developed based on the discussion. The model correlates well with the experimental results. Thus, this model is helpful to optimize the process parameters in double belt press. The remaining issue is the real pressure profile under rollers during the impregnation process. The resin pressure is

generated based on the balance between impregnation flow and in-plane flow. The study is currently in progress using a simulation software.

Finally, we demonstrated the production of 6 mm thickness composite sheet with the double belt press. The material consisted of 28 layers of carbon fiber fabrics and 29 layers of polyamide 6 films. The produced composite sheet showed good impregnation quality and good mechanical property. Now we develop the pre-heating system for double belt press, which will make the production speed higher for the future mass production process.

5. ACKNOWLEDGMENTS

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