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Satoshi Kobayashi [a] , Atsushi Tanaka [a] & Tetsuya Morimoto [b]

[a] Faculty of Mechanical Engineering, Graduate School of Science and Engineering, Tokyo Metropolitan University, 1-1 Minami-Osawa, Hachioji, Tokyo, 192-0397, Japan

[b] Japan Aerospace Exploration Agency, 6-13-1, Ohsawa, Mitaka, Tokyo 181-0015, Japan

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Satoshi Kobayashi[a]*, Atsushi Tanaka[a] and Tetsuya Morimoto[b]

[a]*Faculty of Mechanical Engineering, Graduate School of Science and Engineering, Tokyo Metropolitan University, 1-1 Minami-Osawa, Hachioji, Tokyo 192-0397, Japan;* [b]*Japan Aerospace Exploration Agency, 6-13-1, Ohsawa, Mitaka, Tokyo 181-0015, Japan*

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In order to fabricate composite components more rapidly, a micro-braiding technique has been proposed as fabrication of intermediate material for continuous fiber reinforced thermoplastic composites. In the present study, a simple model considering pressure fluctuation during compression molding is proposed based on the Darcy's law and the continuity equation to predict resin impregnation process using micro-braided yarns. In order to measure pressure fluctuation, a mold die including a channel from the cavity to a pressure gauge was fabricated. To confirm validity of the model, carbon fiber reinforced polypropylene composites were molded under various molding conditions. Once molding pressure was applied, pressure on the molding pieces rapidly increased and gradually decreased to equilibrium with time. Analytical results well-predict resin impregnation during molding and the effectiveness of the model proposed was confirmed.

Keywords: impregnation; continuous fiber reinforced thermoplastics; Darcy's law; micro-braiding technique

1. Introduction

Recently, continuous fiber reinforced thermoplastic composites have attracted much attention because of their superior mechanical properties and lower cycle time for molding. However, it is difficult to impregnate fiber yarns with thermoplastics, because of the extremely high viscosity of molten thermoplastics. Thus, in order to improve impregnation, usage of intermediate materials, such as powder impregnation yarns [1], commingled yarns [2] and micro-braided yarns (MBYs) [3,4] have been developed. During processing of these kinds of intermediate materials, molding and impregnation proceed simultaneously. Therefore, it is important to understand the impregnation mechanism to optimize the molding condition. Gutowski et al. [5] conducted impregnation tests on constant viscosity oils and aligned graphite fibers to measure the deformation behavior of the fibers using a special apparatus constructed to measure the transverse permeability of graphite fibers. They also proposed a model to predict transverse permeability. Bernet et al. [6] proposed an impregnation model for thermoplastic composites from commingled yarns based on Darcy's law. Steggall-Murphy et al. [7] also proposed a model for thermoplastic melt impregnation of fiber bundles in a way similar to Bernet et al. [6]. Bourban et al. [8] discussed the processing parameters based on material

*Corresponding author. Email: koba@tmu.ac.jp

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phenomena during processing of powder impregnated yarns and commingled yarns. These studies treated the impregnation of plastics to reinforcing fiber yarns under constant molding pressure. In the actual molding, molding pressure fluctuates due to various factors, such as vibration of molding machine, flow of molten plastics, etc. An estimation of impregnation under fluctuation of molding pressure is necessary since molding condition is not always the same. In addition, analytical method on impregnation for MBY was limited. In the present study, we propose a simple impregnation model considering a fluctuation of molding pressure based on our previous study [9]. In order to measure pressure fluctuation during molding, a new mold die was fabricated. Impregnation experiments were conducted on MBYs consisting of carbon and polypropylene (PP) fiber yarns using the new mold die. The effectiveness of the model was evaluated by comparing the experimental results to the analytical prediction.

2. Prediction of impregnation

A prediction method for length of time required to impregnate a fiber yarn with resin is necessary to optimize a process condition and a material design. Therefore, we derive impregnation time based on Darcy's law for an incompressible fluid through a porous medium considering a pressure fluctuation. Here, a coordinate system as shown in Figure 1 is assumed for resin impregnation of a MBY.

For the phenomenon of an incompressible fluid flow through a porous medium in z direction, the following equation is obtained according to Darcy's law.

$$u = -\frac{k}{\mu} \cdot \frac{\partial P}{\partial z}, \tag{1}$$

where u is the Darcy's velocity of fluid in z direction, μ is the viscosity of the fluid, $\partial P/\partial z$ is the pressure gradient and k is the permeability. From the equation of continuity,

$$\nabla \cdot \mathbf{u} = 0. \tag{2}$$

Equation (2) is expressed in a cylindrical coordinate system as

$$\frac{\partial u_x}{\partial x} + \frac{1}{r}\frac{\partial}{\partial r}(ru_r) + \frac{1}{r}\frac{\partial u_\theta}{\partial \theta} = 0. \tag{3}$$

In order to predict length of time for impregnation, the phenomenon of the uniform impregnation in the r direction in a fiber yarn with a radius R_0 is considered. Assuming the

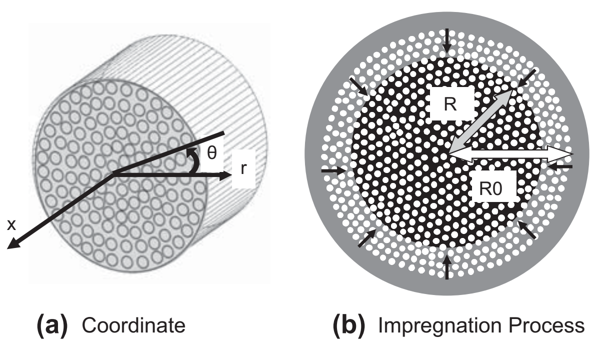

Figure 1. Impregnation model.

velocity in the x (axial) direction is 0 and considering an axisymmetric condition, Equation (3) is expressed as

$$\frac{1}{r}\frac{\partial}{\partial r}(ru_r) = 0. \tag{4}$$

From Equations (4) and (1),

$$\therefore \frac{\partial P}{\partial r} = -\frac{\mu}{k}\cdot\frac{C_1}{r}. \tag{5}$$

Integrating Equation (5) with respect to r,

$$P = -\frac{\mu}{k}\cdot C_1 \ln r + C_2, \tag{6}$$

is obtained, where C_1 and C_2 are constants.

As the boundary conditions: (1) $P = P_m$ at $r = R_0$ (P_m: resin pressure) and (2) $P = P_0$ at $r = R$ (R: distance from the center of the yarn to flow front of the resin, P_0: pressure atmosphere) are used to determine C_1 and C_2. Assuming $P_0 \approx 0$ because $P_m >> P_0$, then Equation (6) is expressed as

$$P = \frac{P_m}{\ln(R_0/R)}\ln r - \frac{P_m}{\ln(R_0/R)}\ln R = \frac{P_m}{\ln(R_0/R)}\ln\left(\frac{r}{R}\right). \tag{7}$$

From Equation (1),

$$u_r = -\frac{k}{\mu}\cdot\frac{\partial P}{\partial r} = -\frac{k}{\mu}\cdot\frac{\partial}{\partial r}\left\{\frac{P_m}{\ln(R_0/R)}\ln\left(\frac{r}{R}\right)\right\} = -\frac{k}{\mu}\cdot\frac{P_m}{\ln(R_0/R)}\cdot\frac{1}{r}, \tag{8}$$

is obtained. Assuming that Darcy's velocity $u_r = V$ at the flow front,

$$V = (1 - V_f)\frac{dR}{d\tau} = -\frac{k}{\mu}\cdot\frac{P_m}{\ln(R_0/R)}\cdot\frac{1}{R}, \tag{9}$$

is obtained, where V_f is fiber volume fraction and τ is impregnation time. Rearranging Equation (9)

$$\therefore R\ln\left(\frac{R_0}{R}\right)dR = -\frac{k}{\mu(1 - V_f)}P_m\, d\tau. \tag{10}$$

Integrating both members of Equation (10),

$$\frac{R^2}{2}\ln\left(\frac{R_0}{R}\right) + \frac{R^2}{4} = -\frac{k}{\mu(1 - V_f)}\int P_m\, d\tau + C_3, \tag{11}$$

where C_3 is constant. In experiments, $R \neq R_0$ at $\tau = 0$, because of the weight of the die and the capillary action during cooling [9]. So assuming the boundary condition $\int P_m dt = 0$ and $R = R'_0$ at $\tau = 0$,

$$C_3 = \frac{R_0'^2}{2} \ln\left(\frac{R_0}{R_0'}\right) + \frac{R_0'^2}{4}, \tag{12}$$

is obtained. Impregnation ratio, I, is defined as the ratio of the impregnation area to cross-sectional area of the yarn, as

$$I = \frac{\pi R_0^2 - \pi R^2}{\pi R_0^2} = 1 - \left(\frac{R}{R_0}\right)^2. \tag{13}$$

And impregnation ratio at $\tau = 0$ is obtained as

$$I_0 = 1 - \left(\frac{R_0'}{R_0}\right)^2. \tag{14}$$

Finally, the relation between impregnation ratio and the integral of resin pressure with respect to time is obtained as

$$\int P_m \, dt = -\frac{\mu R_0^2 (1 - V_f)}{4k} [(1 - I)\{1 - \ln(1 - I)\} - (1 - I_0)\{1 - \ln(1 - I_0)\}]. \tag{15}$$

If a resin pressure history and I_0 are known from an experiment, the relation between the impregnation ratio and time is calculated numerically. In the present study, τ is defined as the pressurizing duration from the beginning of pressurizing to the beginning of the cooling (pressure is released as 0). In order to fit the analytical values to the experimental values at $\tau = 0$, I_0 is obtained from experimental values at $\tau = 0$.

3. Experiments [9]

Materials used were carbon fiber yarn (TR50S-6L, 6000 filaments, 400 tex, Mitsubishi Rayon Co.) and PP yarn (760T120-20S, 760 tex, MRC Pylene Co.) as reinforcement and matrix, respectively. In the present study, MBY were fabricated as an intermediate material with a medium-class braider (Kokubun Limited Co.). A reinforcing fiber yarn was located at the center of the braider and 4 matrix fiber yarns were braided around the reinforcing fiber yarn.

MBY was hot-pressed into rectangular specimens. A mold die used in this study is illustrated in Figure 2. The mold die included a channel from cavities to one lateral side, where a pressure gauge (PHC-B-20MP, Kyowa Co.) was located. Before molding, the channel was filled up with PP fibers. Pressure fluctuation during molding was measured with the gauge.

MBY was wound on a metallic frame 15 times per 10 mm. MBY wound on the frame was placed on the preheated mold die with a demold treatment and was hot-pressed. Hot press system used was IMC-1837 (Imoto Co.). In the present study, molding was conducted at constant load and molding pressure was defined as nominal pressure at the cavity, which is different from ‘real’ pressure on the molding pieces. Molding temperature and time were also defined as the temperature of the heating platens and the duration where pressure and temperature were kept, respectively. Molding condition conducted in this study is shown in Table 1. After keeping the molding pressure and the temperature for a given molding time, pressure was released and the mold die was cooled by water flow in the cooling pipes through the heating platens to less than 50 °C. For molding time of zero min, it is noted that the mold die was pressed to the molding pressure and pressure was released immediately once reaching the molding pressure. Two specimens with 10 mm width and 200 mm length were obtained in one molding.

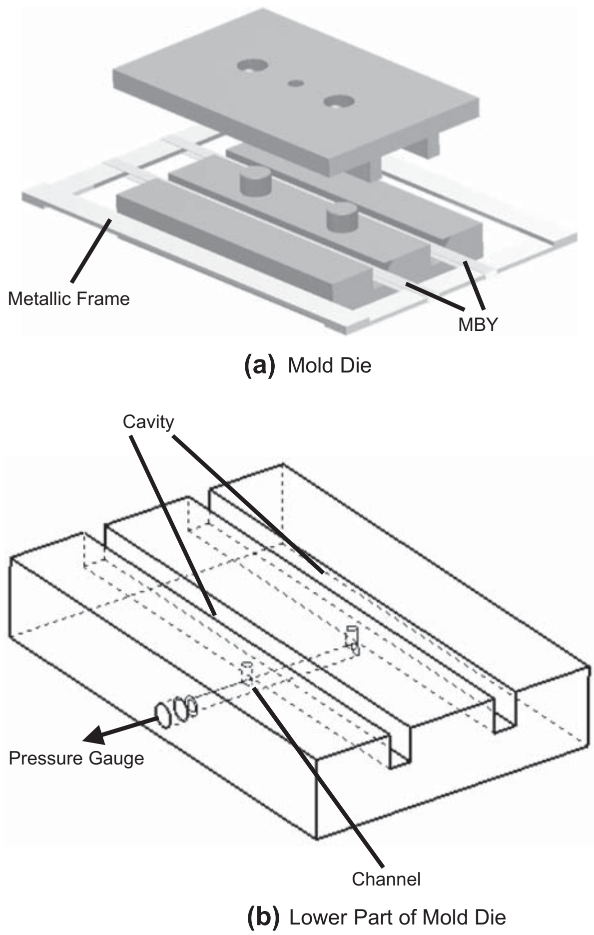

Figure 2. Mold die developed to measure pressure fluctuation.

Table 1. Molding conditions.

Temperature (°C)	Pressure (MPa)	Holding time (min)
200, 220	8	0–10 (every 1 min)
	12	0–5 (every 1 min)

In order to measure the impregnation ratio, cross-sectional observations were conducted at the center of the specimens molded at each condition. After the molded pieces were embedded in epoxy resin and the resin was cured, the cross-section was polished using #180∼2000 emery papers and was finished by buffing with alumina slurry (0.3 μm, Maruto Co.). Then, polished surface was observed using a video microscope and digital images obtained were converted to bitmap files. Impregnation ratio was calculated as the ratio of the number of pixels in impregnation region to that of yarn region, including cross-sectional area of fibers.

4. Results and discussion

Figure 3 shows pressure fluctuation during molding. Once molding pressure was applied, pressure on the molding pieces rapidly increased and gradually decreased to equilibrium with

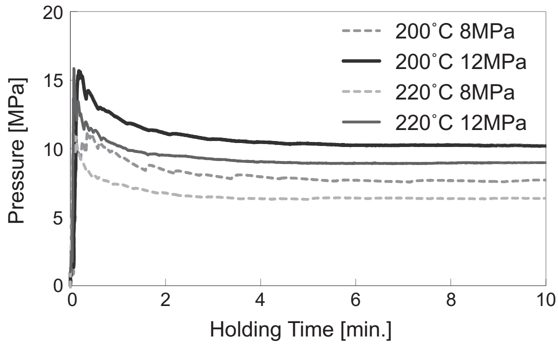

Figure 3. Pressure fluctuation during molding.

time. This decrease in pressure is caused by contact among fibers. During molding, fibers were getting stuck in one another and the increase in contact point allows the fibers to carry some part of the molding pressure. Another possible reason is overflow of molten plastics. Figure 4 shows the end of the mold die. Molten plastics were observed during molding. Pressure drop might be caused by flow of molten plastics according to Bernoulli's theorem.

Figure 5 shows an example of a cross-sectional observation. Impregnated and unimpregnated regions were easily distinguished. Figures 6 and 7 show impregnation ratio as a function of molding time at molding temperature of 200 and 220 °C, respectively. Figures 6 and 7 include the analytical predictions made by curve fitting an Equation (15) to the experimental results with varying the value of μ/k. The value of integral of P was calculated from Figure 1. The yarn radius was measured from the micrograph as $R_0 = 0.6$ mm. In order to fit an Equation (15) to experimental results, the values of μ/k were selected as 1.0×10^{17} kg/s m^3 for 200 °C and 7.2×10^{16} kg/s m^3 for 220 °C. As shown in Figures 6 and 7, fair agreements for impregnation between experiments and analyses were obtained, regardless of molding pressure. Thus, once μ/k value is determined for a given molding pressure, the prediction for arbitrary pressure is available at the same molding temperature. Experimental results did not reach the full-impregnation, whereas the analytical predictions did. This is mainly due to the packing effect which results in the lower permeability. In the

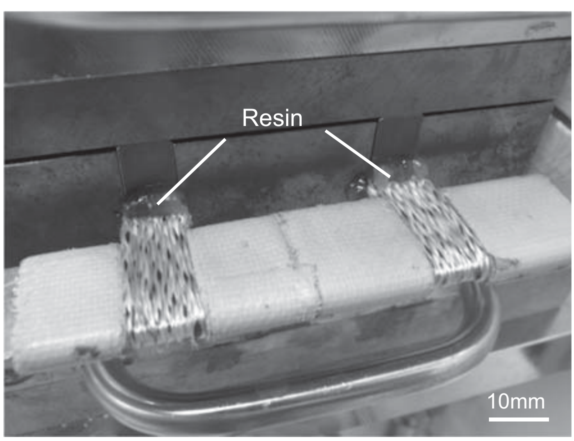

Figure 4. Portion of the opening at the end of the mold die during molding.

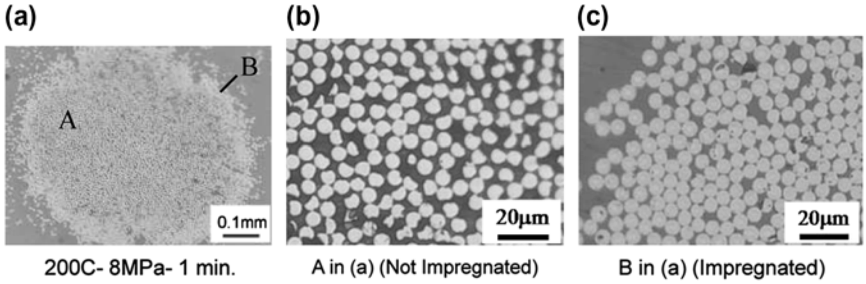

Figure 5. Examples of cross-sectional observation.

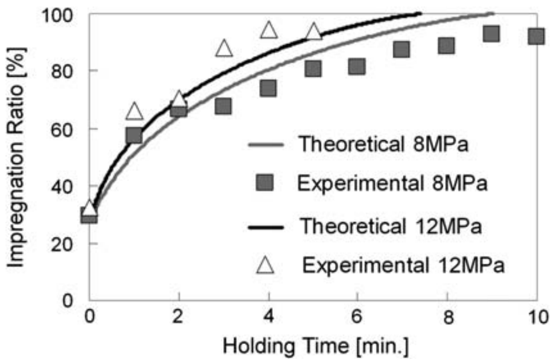

Figure 6. Impregnation ratio as a function of holding time (200 °C).

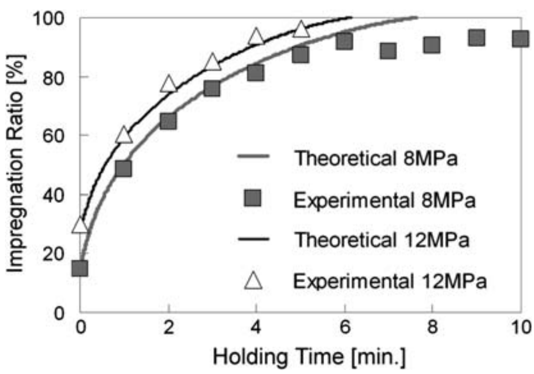

Figure 7. Impregnation ratio as a function of holding time (220 °C).

proposed method, packing effect is neglected. This kind of mechanism was investigated by Gutowski et al. [5]. We will modify our analysis to consider this effect in the near future.

The μ/k values decreased with molding temperature. This is attributed to the decrease in viscosity of PP. From Ref. [10], the viscosities of PP are 4260 Pas for 200 °C and 3100 Pas for 220 °C. With these values, k values are calculated as $4.26 \times 10^{-14}\,\mathrm{m}^2$ for 200 °C and $4.31 \times 10^{-14}\,\mathrm{m}^2$ for 220 °C. This result suggests that k is a constant for a combination of a reinforcing yarn and matrix, and is independent from molding condition. In other words, when viscosity is known as a function of temperature, once k is determined in a preliminary experiment, it is possible to predict the impregnation behavior at any molding condition. On the other hand, analytical prediction overestimates the impregnation for longer molding time at 220 °C. The possible reasons are pyrolysis of resin and/or fluctuation in k value due to fiber stuck in. k values depend on the distance between fibers, which also depends on the molding pressure and viscosity of resin and affects fiber volume fraction. In future work, these effects should be considered in the analysis.

5. Conclusion

In order to optimize the molding condition, a simple resin impregnation model for a MBY was proposed considering pressure fluctuation during molding. A new mold was designed to monitor pressure history during molding. MBYs were fabricated with a carbon fiber yarn and PP yarns. MBYs were molded under various molding conditions. Impregnation ratio was measured as functions of molding time and temperature. Pressure history measured was used to predict impregnation ratio, analytically. Analytical predictions were in good agreement with experimental results and the effectiveness of the proposed model was confirmed. In addition, it is suggested that the permeability is independent from molding temperature, thus, viscosity of molten plastics.

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