EFFECT OF WETTABILITY ON IMPREGNATION PROCESS OF VISCOUS FLUID TO WOVEN FIBER BUNDLES

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ABSTRACT

In order to realize high-quality resin transfer molding processes, it is important to understand the impregnation behavior of viscous fluid inside and between fiber bundles where the flow paths are complicated. In this research, a series of numerical simulations of liquid impregnation into single layer of woven fibers are conducted. We mainly focus on the microvoid formation due to complex convective field between the fiber bundles. Quantitative discussion on the temporal variation of the microvoid rate is also addressed. It is found that the impregnation process drastically changed by varying the kinematic viscosity as well as the contact angle between the viscous liquid and fibers. It is also revealed that the wettability has a greater impact on the impregnation process and microvoid formation than the viscosity in terms of the capillary and modified capillary numbers.

1 INTRODUCTION

There are various manufacturing methods of fiber reinforced plastics (FRP). Resin transfer molding (RTM) method and vacuum assisted resin transfer molding (VARTM) method are known as typical ones among them to produce FRP at relatively low cost. Generally, in these methods, dry fiber bundles are impregnated with the liquid-base material. The quality of FRP depends on the degree of impregnation; insufficient impregnation produces voids such as air bubbles in the product. The breakage of the material would be caused by the stress concentration at these voids. In order to increase the strength of the materials, it is of great importance to suppress the void formation.

Many researches on RTM and VARTM have been conducted with unidirectional and woven fibers as test fibrous media. The effects of impregnation rate on the void formation process have been studied in RTM with unidirectional [1,2], bidirectional, and continuous random [3] glass fiber mats. Characteristic velocity of the impregnation is generally expressed by capillary number (Ca) by the following equation,

$$Ca = \frac{\mu U}{\sigma}.$$  \hspace{1cm} (1)

where $\mu$ is the dynamic viscosity of the liquid, $U$ is the characteristic velocity of the impregnation, and $\sigma$ is the surface tension. When Ca is small, the free surface of the liquid tends to penetrate into the fiber bundles because the capillary force is dominant for the liquid. When Ca is large, on the other hand, the free surface of the liquid tends to impregnate along spaces among the fiber bundles because the viscous force is dominant for the liquid. Previous studies have shown that Ca of the order of $10^{-3}$ is optimal for minimizing voids formed in the fiber bundles [1, 3, 4]. Rohatgi et al. [1] introduced the modified capillary number (Ca') by considering the contact angle $\theta$ between the resin and fibers, and showed the correlation between the void formation and the resin impregnation. Here, Ca' is represented by the following equation,

$$Ca' = \frac{\mu U}{\sigma \cos \theta}.$$  \hspace{1cm} (2)

The voids generated in the process of impregnating liquid into fibers are roughly classified into two patterns depending on their sizes [3, 5]; the one is macrovoid, which is a relatively large void trapped outside the fiber bundles, and the other is microvoid, which is very small formed among the fibers inside the fiber bundles.
Impregnation processes have been investigated via numerical analysis as well in order to comprehensively understand and predict the complex flow fields. The Darcy’s law has been widely used to predict the impregnation process in VARTM and RTM [6, 8]. In this model, the flux is described to be proportional to the pressure gradient in a uniform porous body. This model has been applied to simulations such as RTM with a single-layer glass woven fiber bundles [7]. In the real system for the impregnation, one cannot avoid uneven boundary conditions, for instance, near the substrate surface in the mold and near the stitching yarn, where it is hard to apply the Darcy’s law. In order to overcome such problems, complex models of permeabilities have been prepared.

One can find a few numerical simulations without employing the Darcy’s law in impregnation processes; Okabe et al. [9] performed a two-dimensional numerical simulation of liquid-gas system by the moving particle semi-implicit (MPS) method for the process of impregnating viscous liquid into fiber bundles aligned perpendicular to the net flow direction. This method is based on the Navier-Stokes equation and can handle complex boundary conditions [10,11]. They also described the effect of wettability and fiber placement on void formation [9].

As above, there have been studies on the void formations and the behaviors of flow front in the impregnation processes, but one has to accumulate knowledge on the fluid behaviors around fibers and the formation of microvoids in spatially-inhomogeneous three-dimensional fields. In this study, we conduct a series of numerical simulations on RTM method in the single layer woven fiber bundles as the target geometry via MPS method. In particular, the effects of wettability on the three-dimensional behaviors of the flow front of the viscous liquid and on the formation processes of microvoids in fiber bundles are investigated by considering designated values of $\theta$.

2 NUMERICAL SIMULATION

In this research, we use Particleworks (ver. 6.1) (Prometech Software, Inc., Japan), based on MPS method. The woven fiber bundles are considered as the target geometry as shown in Fig. 1 (a). Since it is difficult to reproduce the precise geometry of the fiber bundles used in the experiments in the real spatial scale, we prepare the fiber bundles in a rather larger spatial scale but with the same fiber volume fraction. The continuous fibers are simulated by inserting the woven fiber bundles between parallel plates and giving periodic boundary conditions to the both sides. It is assumed that all fibers and plates are rigid. In this research, the gap or null area between fiber bundles is called a channel as called in the previous study [4]. Figure 1 (b) illustrates an example of the calculation results; green and blue particles indicate the gas and the liquid, respectively, incorporated into the target model as shown in Fig. 1 (a). The both substrates are omitted for the sake of visibility. As the initial condition, the whole region except the fibers are filled with the gas particles. At $t = 0$, the liquid particles are injected from the inlet, that is, the upstream side of the mold, at a constant designated velocity in $x$ direction.

![Fig. 1 Numerical model utilized in this study. (a) Bird-eye view without liquid and gas particles, and (b) that with liquid (blue) and gas (green) particles. Fiber bundles are inserted between rigid substrates. The both substrates are omitted for the sake of visibility.](image-url)
Although we employ the fibers of a larger spatial scale than the real fibers used in the experiments, we set \( \text{Ca} \) comparable to the experimental study [1]; \( 0.02 \leq \text{Ca} \leq 0.1 \). The contact angle between the viscous fluid and the wall, \( \theta_w \), is fixed at 0°, and the one against the fiber, \( \theta_f \), is varied from 0° to 60°. Physical parameters of liquid and gas particles are shown in Table 1.

<table>
<thead>
<tr>
<th></th>
<th>Liquid (blue)</th>
<th>Gas (green)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (kg/m³)</td>
<td>1100</td>
<td>1.2</td>
</tr>
<tr>
<td>Kinematic viscosity (mm²/s)</td>
<td>36.36 - 90.91</td>
<td>15</td>
</tr>
<tr>
<td>Surface tension (mN/m)</td>
<td>21</td>
<td>0</td>
</tr>
<tr>
<td>Initial velocity (mm/s)</td>
<td>10</td>
<td>0</td>
</tr>
<tr>
<td>Particle diameter (mm)</td>
<td>0.57</td>
<td>0.57</td>
</tr>
<tr>
<td>Contact angle (fiber) (°)</td>
<td>0 - 60</td>
<td>-</td>
</tr>
<tr>
<td>Contact angle (substrate) (°)</td>
<td>0</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 1. Physical properties.

RESULTS & DISCUSSION

Figure 2 shows an example of impregnation under \((\text{Ca}, \text{Ca'}, \theta_f) = (3.0 \times 10^{-2}, 3.0 \times 10^{-2}, 0)\). In Fig. 2, in order to track the impregnation process of the liquid particles easier, the gas particles originally filled in the mold are not visualized intentionally. Under this condition, the flow front precedes the channel region preferably. Then, it impregnates between the fiber bundles, and passes through the channel to reach the outlet. At \( t = 1.2 \) s, since the liquid particles precedes between the fiber bundles, the flow front exhibits a turn to \( y \) direction to flow back into the fiber bundles in the region as indicated as ‘A’ in the frame. This qualitatively reproduces the phenomenon as seen in the experiment [1].

Figure 3 illustrates (a) the pressure distribution in the liquid, and the velocity fields in (b) \( y \)- and (c)\( z \)-directions. In Fig. 3 (a), the pressure is high near the inlet, and is low between and in the fiber bundles. The vectors in Fig. 3 (b) and (c) represent the thinning rate as 0.85, and the length of the vector is fixed.

Fig. 2 Temporal variations of impregnation captured by numerical calculation with \( \text{Ca}, \text{Ca'} = 3.0 \times 10^{-2}, \) and \( \theta_f = 0^\circ \). Note that only fiber bundles and liquid are visualized.

Figure 3 illustrates (a) the pressure distribution in the liquid, and the velocity fields in (b) \( y \)- and (c)\( z \)-directions. In Fig. 3 (a), the pressure is high near the inlet, and is low between and in the fiber bundles. The vectors in Fig. 3 (b) and (c) represent the thinning rate as 0.85, and the length of the vector is fixed.
As shown in Fig. 3 (b), the liquid particles from the inlet travel from the channel to the fiber bundles, then pass through the channel near the outlet, and reach the outlet. The liquid particles with positive velocity in the $y$ direction and those with negative velocity intersect between the fiber bundles. This indicates that the liquid particles flowing through each channel are intermingled among the fiber bundles. In other words, in the woven fiber bundles, there are several channels in the model due to the bent warp and weft, which causes a complicated flow path inside the system. As shown in Fig. 3 (c), the liquid particles flowing in the channel near the bottom plate flow in the $+z$ direction between fiber bundles, and reach the outlet near the bottom plate. From Fig. 3 (b) and (c), it can be seen that not all liquid particles passing through a channel follow the same trajectory. This is a typical example that the woven fiber bundles cause a three-dimentional complex impregnation within the mold.

Fig. 3 Cross sectional view of the simulation results visualizing (a) pressure distribution and velocity distributions in (b) $y$-, and (c) $z$- directions. A cross section is taken at the center portion of the fiber model in the height direction ($z$ direction in Fig. 1). The calculation conditions are the same as those in Fig. 2.

Next, the influence of kinematic viscosity in $\text{Ca}$ on the formation of microvoids is discussed. Figure 4 (a) and (b) show the temporal variations of the microvoid rate and the void rate of the entire system, respectively, from the start of the liquid impregnation under $\theta_f = 0^\circ$. We count the number of air particles existing between fiber bundles as $N_1$ and those in the channel $N_2$. Then the number of the microvoids $N$ are defined as $N = N_{\text{total}} - (N_1 + N_2)$, where $N_{\text{total}}$ is the total number of the air particles initially filled in the geometry. The initial microvoid rate is of 67.7%. As the impregnation proceeds, the microvoid rate and the total void rate decrease against time, and they gradually converge to approximately constant values. Since the values of the microvoid rate and the total void rate almost the same, one can see that almost no macrovoids are formed under this condition. Note that the variations are not apparent until $t \sim 6$ s under various kinematic viscosity. After $t \sim 6$ s, different tendencies emerge in the temporal variations under different kinematic viscosities. This instant roughly coincides with the time when the liquid particles complete impregnation between fiber bundles and across the
channel. That is, when the liquid particles are impregnated in the fiber bundles, the influence of the viscous force is remarkably observed. The slopes of the microvoid rate and the void rate become smaller as increasing the kinematic viscosity (or in large Ca'). Generally, there are two methods for measuring the void rate; a method to measure the void rate after the cure of the impregnated resin, and another to measure the void rate immediately after the completion of impregnation. In this study, the time when the impregnation is completed is defined as the instant when the time change rate of the microvoid rate reaches 0.05 (%/s), and the rate at that time is defined as the final microvoid rate.

Figure. 5 (a) and (b) show the results of the final microvoid rate by Ca' and Ca. This figure shows the influence of $\theta_f$ and $\nu$ on the final microvoid rate. As shown in Fig. 5 (a), the final microvoid rate is suppressed by varying $\nu$ more significantly than the case varying $\theta_f$. The effects of $\theta_f$ and $\nu$ are considered to be equivalent when expressed as dimensionless numbers, but the definition of Ca' needs to be modified since it is shown that those contributions are not equivalent as shown in Fig. 5 (a). In Fig. 5 (b), the microvoid rate is plotted against normal Ca. The variation of $\theta_f$ is not apparently reflected to the microvoid rate against Ca. The correlation between the final microvoid rate and $\theta_f$ is shown in Fig. 6. The final microvoid rate increases as $\theta_f$ and $\nu$ increase. It is also found that the effect of the viscosity is smaller under larger $\theta_f$ than 0°, i.e., the influence of $\theta_f$ is greater than $\nu$. Under the present calculation conditions, the formation of microvoids can be suppressed when the viscosity is small and the wettability is good.
CONCLUSIONS

We analyze the liquid impregnation process induced by the pressure difference for a single-layer woven fiber mat within a pair of the flat parallel plates via numerical approaches. It is indicated a correlation between the microvoid formation and the impregnation process in terms of Ca and Ca’. We obtain the same tendency that the microvoids are formed by preceding of the liquid between the fiber bundles as in the previous studies. We realize the process of viscous fluid impregnation into woven fiber bundles initially filled with the gas by three-dimensional numerical calculation with moving particles semi-implicit (MPS) method. Modified capillary number, Ca’, considering the effect of the contact angle between the liquid and the fibers [1] is changed by varying the kinematic viscosity of the liquid to investigate their effect on the formation of the microvoids. It is indicated that the influence of the contact angle is underestimated in the definition of Ca’.

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REFERENCES


