OPTIMIZATION OF CURING TEMPERATURE PATTERNS FOR FRP PLATE WITH UNEVEN DEGREE-OF-CURE DISTRIBUTION

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ABSTRACT

Recently, low-cost manufacturing methods for thermosetting FRP, the OoA (Out-of-Autoclave) molding methods such as VaRTM and low-cost heating methods without a large furnace have been focused on by many researchers and manufacturers. Especially, regarding to low-cost heating for large-scale structure, temperature distribution with large unevenness may occur overall FRP. Avoiding quality reduction resulted from uneven progress of cure reaction, it is very important to optimize temperature condition for decreasing unevenness of degree-of-cure distribution. In this study, optimization of temperature patterns for cure process of single-side-heated glass/epoxy plates after VaRTM process was conducted. Temperature was loaded on the one-side by a sheet heater and another side was set on a steel die. First, the coupled analysis method to obtain distribution of temperature and degree-of-cure was constructed. The optical fiber sensors were used to measure degree-of-cure distribution of the FRP plate experimentally. It was shown that degree-of-cure distribution calculated by our analytical model agreed very well with the measured distribution by optical fiber sensors. Second, multi-objective optimization of a polyline pattern of molding temperature was conducted. It was found that the two nodes design was applicable to obtain good optimum solution for minimizing the cure completion time, unevenness of degree-of-cure distribution and energy cost of manufacturing.

1 INTRODUCTION

Recent advances of composites molding technologies allow us to manufacture large and complex-shaped composite parts such as wings and fuselages of aircraft. However, high manufacturing cost of FRP using an expensive autoclave is a disadvantage. In order to reduce manufacturing cost of thermosetting FRP, the OoA (Out-of-Autoclave) molding methods such as VaRTM and low-cost heating methods without a large furnace have been developed by many researchers and manufacturers. Especially, regarding to low-cost heating for large-scale structure, temperature distribution with large unevenness may occur overall FRP. Avoiding quality reduction resulted from uneven progress of cure reaction, it is very important to optimize temperature condition for decreasing unevenness of degree-of-cure distribution. However, optimization of molding condition of temperature and pressure has been carried out by human trail-and-error method, which costs high, traditionally. Therefore, more efficient and low cost method for obtaining the optimum molding condition has been desired commercially.

In situ sensing method using embeddable sensors is an attractive solution for efficient exploration of optimum molding condition of FRP. Among embeddable sensors, optical fiber sensors are very promising due to the easy embedability, durability and accuracy. It has been shown that Fresnel-based optical fiber sensors can be used for measuring degree-of-cure of thermosetting resin [1-4]. In addition, this technique provides us an easy method to create a precise model to predict distribution of temperature and degree-of-cure. Therefore, the optimization of molding condition using the analytical...
model created from measured data by the optical fiber sensors will be able to become an effective method to manufacture high-quality FRP products.

In this study, optimization of temperature patterns for single-side-heated GFRP plates after VaRTM process was conducted. Temperature was loaded on the one-side by a sheet heater and another side was set on a steel die. First, a one-dimensional analytical model to predict distribution of temperature and degree-of-cure was developed. The simulated results were confirmed by comparing experimental results obtained multi-points measurement using Fresnel-based optical fiber sensors. Second, multi-objective optimizations of temperature pattern were conducted using three objective functions which are cure completion time, unevenness of degree-of-cure and total heat flow.

2 MONITORING AND SIMULATION OF THICKNESS-WISE DISTRIBUTION OF DEGREE-OF-CURE USING EMBEDDED OPTICAL FIBER SENSOR

2.1 Cure monitoring system

The cure monitoring system using a Fresnel-based optical fiber sensor has been developed in our laboratory[1]. The figure 1 illustrates the monitoring system. The system has a 1310nm SLD light source, an optical receiver, an optical fiber sensor with distal-end and an optical circulator. The light intensity of Fresnel’s reflection at the interface between glass optical fiber and resin is measured by an optical receiver. The reflection rate is governed by refractive index of resin principally during cure process. Then, refractive index of resin can be obtained from the measured light intensity. The refractive index of resin depends on temperature and degree-of-cure. Therefore, degree-of-cure can be calculated from a diagram of refractive-index and temperature.

Figure 1: Fresnel-based optical fiber sensor and cure monitoring system.

![Figure 1](image1.png)

Molded plate
Optical fiber
FRP
Thermocouple
Silicone rubber heater
Room temperature 24 °C
Gas flushing film
Grass cloth 60 pieces
Molding plate

Figure 2: Experimental set-up for measuring distribution of degree-of-cure of textile GFRP.

![Figure 2](image2.png)
2.2 Experimental set-up

For manufacturing FRP, epoxy resin (ARALDITE LY5052, hardening: ARADUR 5052 CH) was employed as matrix resin and glass cloths of thickness of 0.1mm were used as reinforcement. The experimental configuration for measuring thickness-wise distribution of degree-of-cure of a GFRP plate is shown in Fig. 2. The optical fiber sensors and thin thermocouples were embedded in bottom, central and top layers of the 60 glass cloths’ preform. The three positions are called the lower, middle and upper in this study. Thickness of the FRP was 5.3 mm. After VaRTM process, a sheet heater was put on the surface of vacuuming bag of the material. The heater setting was a single-step heating process composed of the 2K/minutes heating stage from room temperature (25 °C) to 80 °C and the temperature-maintaining stage for three hours at 80 °C. Large temperature gradient will be generated due to the large thickness and low thermal conductivity of GFRP.

2.3 Coupling simulation of temperature distribution and degree-of-cure

In the present study, one-dimensional heat equation and a kinetic model of cure reaction were used to simulate temperature distribution and degree-of-cure. The heat equation was written as

\[
\rho c_p \frac{\partial T}{\partial t} = \lambda \frac{\partial^2 T}{\partial x^2} + Q.
\]

(1)

Where, \( T \) is absolute temperature, \( x \) is thickness-wise coordinate, \( \rho \) is density, \( c_p \) is specific heat and \( Q \) is volumetric heat flux. The heat flux is defined by exothermic cure reaction as below.

\[
Q = \rho H \frac{d\alpha}{dt}
\]

(2)

Here, \( H \) is total heat of cure reaction per weight and \( \alpha \) is degree-of-cure. The degree-of-cure can be calculated by the following Kamal’s kinetic model[5].

\[
\frac{d\alpha}{dt} = (k_1 + k_2 \alpha^n)(1 - \alpha)^n.
\]

(3)

Where, \( m \) and \( n \) are degree of reaction, and \( k_1 \) and \( k_2 \) are constants which are defined by Arrhenius formula.

\[
k_1 = A_1 \exp\left(-\frac{E_1}{RT}\right), k_2 = A_2 \exp\left(-\frac{E_2}{RT}\right).
\]

(4)

Where, \( A_1 \) and \( A_2 \) are constants, \( E_1 \) and \( E_2 \) are activation energies, and \( R \) is the gas constant. The parameters of Kamal model were obtained experimentally using a DSC (Differential Scanning Calorimeter) and optical fiber sensors.

Figure 3 illustrates the analytical model used in this study. The simulation was carried out using Mathematica. Thickness \( L \) of FRP was 5.3 mm, the initial condition of temperature was 25 °C and the initial condition of degree-of-cure was zero.

Figure 3: Simulation model of single-side-heated FRP plate.
The boundary conditions are described as bellow.

\[
T(t, 0) = T_h(t), \quad q(t, L) = -\lambda \frac{\partial T}{\partial x}(t, L) = h(T(t, L) - 25),
\]

Here, \( T_h \) is temperature of a heater, \( h \) is heat transfer coefficient between steel and the air.

2.4 Experimental and simulated results

Temperature at the three positions were measured by the multiple thermocouples and simulated by our model. Figure 4 plots measured and simulated distribution of temperature as functions of experimental results. Additionally, it was found that large temperature gradient occurred during cure process. Final temperatures at the upper, middle and lower positions were 78°C, 65°C and 53°C, respectively. The final temperature gradient was 4.7 K/mm.

![Figure 4: Measured and simulated temperature as functions of curing time.](image)

![Figure 5: Measured and simulated distribution of degree-of-cure as functions of curing time.](image)
Degree-of-cure at the three positions were measured by the multiple optical fiber sensors and simulated by our model. The measured and simulated distribution of degree-of-cure are plotted in Fig.5. From the figure, it was found that the estimated degree-of-cure curves agreed very well with the experimental curves. Therefore, it appeared that the simulation model made using data measured by optical fiber sensors had good accuracy to predict degree-of-cure.

From the degree-of-cure curves, it was seen that cure process delayed due to the large temperature gradient. The completion times of curing reaction of the upper, middle and lower parts were 2, 3 and 4.5 hours. In the present paper, curing temperature condition will be optimized to minimize the cure completion time at the lower position, difference of degree-of-cure between at the upper and lower positions and also consumption energy heating the material through heater.

3 OPTIMIZATION SETTING OF TEMPERATURE CONDITION

3.1 Design parameters

In this study, the temperature pattern was defined as a polyline in which the initial and maximum temperatures were 25 and 115 °C as shown in Fig.6, respectively. The temperature becomes maximum at 60 minutes and the temperature is maintained for 2 hours. Design parameters of the temperature pattern are defined as mid-point nodes of the temperature polyline. One node has two design parameters of time and temperature. Ranges of the node’s parameters are from 2 to 58 minutes in time and from 25 to 115 °C in temperature. Three types of parameters’ combination are shown in Table.1. The single node design has two design parameters and a five level full fractional design (25 samplings). The double node design has four design parameters and a five level central composite design (CCD). These two designs were used to obtain response functions and minimizations of objective function are conducted. The multiple nodes design has nine nodes and eighteen design parameters and were used to obtain response by Monte-Carlo simulation. The Pareto solution obtained by response function methods using single node and double nodes designs will be compared with results of the Monte-Carlo simulation using the nine nodes design.

<table>
<thead>
<tr>
<th>Number of nodes</th>
<th>Number of parameters</th>
<th>Design method</th>
<th>Calculation method</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2</td>
<td>5 level Full</td>
<td>Response function</td>
</tr>
<tr>
<td>2</td>
<td>4</td>
<td>5 level CCD</td>
<td>Response function</td>
</tr>
<tr>
<td>9</td>
<td>18</td>
<td>Random</td>
<td>Monte-Carlo</td>
</tr>
</tbody>
</table>

Table 1: Design parameters and calculation methods.
3.2 Response functions

Three response functions, the cure completion time, unevenness of degree-of-cure and energy cost were paid attention to in this optimization. The cure completion time $t_{0.95}$ is defined as the time when the lowest degree-of-cure exceeds 0.95. The unevenness of degree-of-cure $\Delta \alpha_{UE}$ is the maximum difference of degree-of-cure in thickness-wise direction during cure process. The energy cost $q_{0.95}$ is defined as total heat flow from the heater into the material until curing time becomes $t_{0.95}$. Optimizing temperature patterns using the single node and double nodes designs, the second-orders response functions as described below were fitted from calculation results using the sampling data by the least squared method.

$$f(x_i) = \beta_0 + \beta_i x_i + \beta_{ij} x_i x_j$$

\[i, j = 1, 2, ..., n \quad \beta_i = \beta_{ij} \]

3.3 Objective functions

Two objective functions $F_1$ and $F_2$ composed of linear combination of two of three response functions were used for multi-objective optimization. These functions are described as

$$F_1 = a_{t_{0.95}} + (1 - a) \Delta \alpha_{AE}$$

$$F_2 = a_{q_{0.95}} + (1 - a) \Delta \alpha_{AE}$$

Here an overbar of variables indicates that the value is normalized by mapping $[\text{max}, \text{min}]$ to $[1, 0]$ and $a$ is a weight coefficient of the linear combination. Conducting global minimization with changing the weight from 0 to 1, a group of Pareto solutions is obtained.

4 RESULTS AND DISCUSSIONS

4.1 Single node design

The figure 7 shows Pareto solution of the objective function $F_1$ composed of the cure completion time and unevenness of degree-of-cure. The temperature patterns designs for Pareto solutions are plotted in the same figure. The figure showed that the unevenness of degree-of-cure was maximum when the cure completion time was minimum. It was also found that the unevenness tended to decrease with increase of the cure completion time. From the results, it seemed that the curve of Pareto solution had a discontinuous section when the cure completion time was in a range from 6450 to 6800 seconds. This reason is because the unevenness of solution curve is not nonatomic and has the local maximum in that range. The multi-objective function $F_1$ which is a linear combination type is not stable to obtain such Pareto solutions.

As for the optimum designs of temperature pattern, the Pareto solution has two types. One is a design group of patterns indicated by (d), (e) and (f) in Fig.7. In this design group, temperature increases with high heating rate at the first section and the design is suitable to decrease the cure completion time. Another design group are shown with (a), (b) and (c) in the figure and has almost uniform heating rate. This design group can minimize the unevenness of degree-of-cure.

The results of Monte-Carlo simulation are plotted as a graph of relationship between the unevenness of degree-of-cure and the cure completion time in Fig.8. In the figure, red plots were the solutions of multi-objective optimization shown in Fig.7. From the figure, it appeared that the Pareto solution between 4000 and 6000 seconds showed the lower limit of the results of Monte-Carlo simulation. However, the results of Monte-Carlo simulation showed better solutions than the Pareto solutions, for example of the result a in Fig.8, when the cure completion time was larger than 6000 seconds. The temperature patterns of the result (a) was very similar to a so-called double-steps pattern.
Therefore, in order to minimize the unevenness of degree-of-cure, it appeared that number of the nodes should be equal to or larger than 2 to express a double-step temperature pattern.

![Figure 7: Pareto solutions of the objective function $F_1$ with temperature pattern designs.](image)

Pareto solution of the objective function $F_2$ composed of total heat flow (energy cost) and unevenness of degree-of-cure were plotted in Fig.9. From the figure, it was found that the Pareto solution curve had linearity without a range of total heat flow from 18 to 18.95. The discontinuous section of the Pareto solution was caused by the same reason as the Pareto solution of $F_1$. Paying attention to the optimized temperature pattern, the solution minimizing total heat flow was combination of very low heating rate in the first section and high heating rate in the second section. This shape of optimized temperature pattern was opposite to the temperature pattern which minimize the cure completion time as mentioned regarding to Fig 7.

Results of Monte-Carlo simulation are plotted in a graph of relationship between the total heat flow and the unevenness with temperature pattern designs in Fig.10. From the figure, it appeared that the Pareto solution was the lower limit except the discontinuous section. The results of Monte-Carlo simulation showed the lower limit of the solution group has a linear relationship. Therefore, it was found that the temperature polyline which has two or more nodes was necessary to obtain Pareto solution useful in the all range of the total heat flow.
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Figure 9: Pareto solutions of the objective function $F_2$ with temperature pattern designs.

![Figure 9: Pareto solutions of the objective function $F_2$ with temperature pattern designs.](image)

Figure 10: Results of Monte-Carlo simulation with temperature pattern designs and results of single node design in Fig.9.

4.2 Double nodes design

Figure 11 shows Pareto solutions of $F_1$ of the double nodes design. In this figure, the Pareto solutions of the double nodes design and results of Monte-Carlo simulation are also plotted. Comparing the solutions of the double nodes design with the single node design, it was found that the both optimized solutions were almost the same as each other when the cure completion time was less than 6000 seconds. In addition, it appeared that the Pareto solutions of the double nodes design did not have discontinuous section. However, the figure shows that the Pareto solutions of the double nodes are not lower limit of results of Monte-Carlo simulation. This reason is that error of the second-order response surface of the double nodes design is large when the cure completion time is larger than 6000 seconds. Therefore, it is necessary to narrow the range of design parameters to obtain the better solutions.

Pareto solutions of $F_1$ of the double nodes design obtained by the narrower range of design parameters are plotted in Fig.12. This figure shows that the optimized solutions could represent the lower limit of the results of Monte-Carlo simulation successfully. Therefore, it was found that the
double nodes design was applicable to obtain optimum solution to minimize the unevenness of degree-of-cure.

![Diagram](image1.png)

**Figure 11:** Pareto solutions of $F_1$ of the double nodes design (red) with results of Monte-Carlo simulation (blue) and the optimized solutions of single node design in Fig.7 (green).

![Diagram](image2.png)

**Figure 12:** Pareto solution of $F_1$ of the double nodes design by the narrower range of design parameters (red) with results of Monte-Carlo simulation (blue) and the optimized solution of single node design in Fig.7 (green).

5 CONCLUSIONS

In the present study, development of prediction model of thickness-wise distribution of temperature and degree-of-cure of FRP plate and optimization of temperature patterns were conducted. The knowledges obtained in this study are summarized as bellow.

1. The coupled analysis method to obtain distribution of temperature and degree-of-cure was constructed successfully. The degree-of-cure distribution calculated by our analytical model was confirmed by the measured distribution by optical fiber sensors.

2. Multi-objective optimization of a polyline pattern of molding temperature was conducted successfully. It was found that the two nodes design was applicable to obtain good optimum solution for minimizing the cure completion time, unevenness of degree-of-cure distribution and energy cost of manufacturing.
3. Minimizing solution of the cure completion was high heating rate in the first section. Minimizing solution of the unevenness of degree-of-cure was double-steps pattern. Minimizing solution of the energy cost was a combination of very low heating rate in the first section and high heating rate in the second section.

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