HIGH ACCURACY PREDICTION OF TEMPERATURE PROFILES IN COMPOSITE PARTS DURING AUTOCLAVE CURING

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Abstract
This paper addresses high accuracy prediction of temperature profiles in composite parts during autoclave curing by using finite element process modeling software, COMPRO. To improve prediction accuracy of thick composite part made of thermoset matrix composites, the appropriate measuring method for thermo-physical properties (heat of reaction, cure kinetics, specific heat capacity and thermal conductivity) was proposed. As a result, the prediction values of temperature profiles corresponded well with the experimental values. In particular, the difference of exothermic peak temperature between prediction and experimental values was within 5 C.

1. Introduction

Fiber-reinforced plastic composites are increasingly being used for various applications due to their superior properties such as high strength, light weight and high corrosion resistance. In general, thermoset matrix composites are manufactured through thermal processing using autoclave. Since the mechanical properties vary depending on the thermal history, the composite parts must be cured with an appropriate cure cycle.

However, the curing of thick composite parts easily induces the internal heat called exotherm during the cure of resin, and the parts temperature occasionally deviates from the required temperature range. Current trial-and-error approaches for development of the cure process are not well-suited and it is a critical issue to reduce the development cost.

For the purpose of the development cost reduction, the temperature profiles of the composite parts during an autoclave curing have been predicted by using a finite element process modeling software, COMPRO developed by Convergent Manufacturing Technologies, Inc. [1-3]. There is a thermo-chemical module that solves the thermal problems including heat transfer and the exothermic response of the matrix resin in cure process and predicts the internal temperature in the composite parts.

The primary focus of this study is to improve prediction accuracy of thick composite part made of thermoset matrix by applying the appropriate measuring method for thermo-physical properties such as heat of reaction, cure kinetics, specific heat capacity and thermal conductivity. As the measuring method, temperature-modulated DSC (Differential Scanning Calorimetry) was chosen for determining heat of reaction, cure kinetics and specific heat capacity. Thermal conductivity was measured by periodic heating and infrared radiation thermometer method that was capable of considering the thermal anisotropy of composites.

Firstly, this paper describes the measuring of thermo-physical properties of epoxy resin infused carbon fiber fabric prepreg, and then describes the thick laminate fabrication with thermocouples to measure the temperature profiles of the composite part. Finally, the part temperatures measured in the experimental tests are compared with those of prediction values.

2. Measuring of thermo-physical properties

2.1. Temperature-modulated DSC

Temperature-modulated DSC (MDSC) tests were performed on a TA Instruments Q1000 to determine the heat of reaction, cure kinetics and specific heat capacity. As shown in Table 1, epoxy resin infused carbon fiber fabric prepreg was evaluated. The samples were heated at a constant ramp rate of 1.0, 1.7, 3.5 and 6.0 C/min, including material manufacture's recommended ramp rate of 1.7 C/min.

Table 1. Technical data sheet of prepreg.

Prepreg Properties	
Material Type	Epoxy resin infused carbon fiber fabric
Fabric Architecture	5 harness satin weave (5HS)
Resin Content (RC)	37 %
Thickness	0.28 mm/ply
Curing Condition	1.7 C/min, 2 hours at 185 C, 0.65 MPa pressure

In the case of MDSC, the data not only provides the information obtainable from conventional DSCs, but also captures the response of the material to the temperature modulation. The total heat flow data (equivalent to the heat flow obtained from conventional DSCs) is obtained by averaging the modulated heat flow. The non-reversing heat flow component was used to determine the heat of reaction and cure kinetics. The reversing heat flow was used to obtain the specific heat capacity.

To determine the heat of reaction (HR) and cure kinetics from the non-reversing heat flow, a linear baseline is typically extended from the start point to the end point of the reaction. In the case of material evaluated in this study, the heat flow starts to increase at the end of reaction. This is associated with material degradation and hence a bilinear baseline is chosen to consider the material degradation at higher temperatures as shown in Figure 1. Once the baselines were established, the HR was calculated by integrating the difference between the non-reversing heat flow ($\dot{q}$) and the baseline ($\dot{q}_{Baseline}$). The input parameter for prediction was obtained by averaging the HR of four ramp rate.

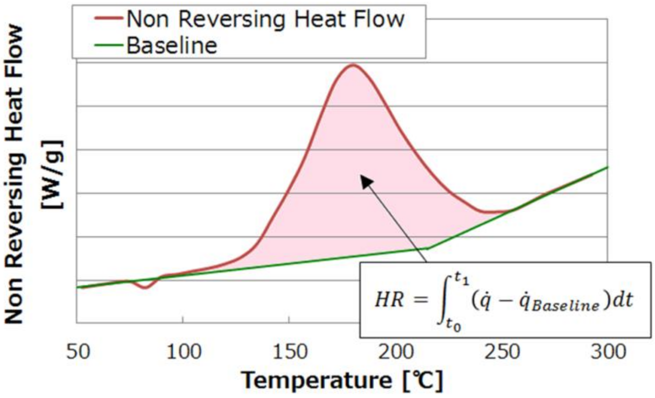


Figure 1. Heat of reaction calculation (1.7 C/min).

To determine the cure kinetics, the experimental data was fit using cure kinetics model. The mathematical forms of cure kinetics model are defined by Eq. 1 [4]. Figure 2 shows a comparison of the cure rate from experimental data and calculated by the model.

$$\frac{d\alpha}{dt} = (K_1 + K_2\alpha^m)(1-\alpha)^n \qquad (1)$$
$$K_i = A_i e^{-\Delta E_i/RT}$$

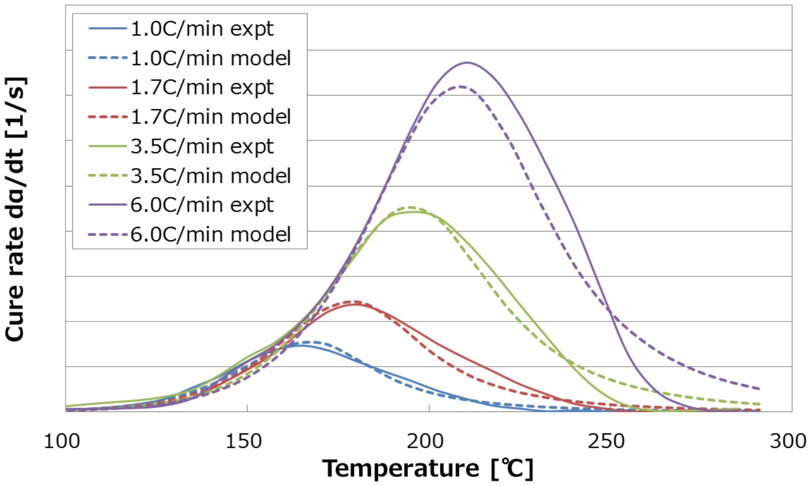


Figure 2. Comparison of cure rate between experiment and model.

The specific heat capacity was obtained from the reversing heat flow in the MDSC tests. As shown in Figure 3, specific heat capacity of the prepreg depends linearly on the temperature. The specific heat capacity at ramp rate of 1.7 C/min, suggested by material manufacture, showed the nearly average value of four ramp rates. The heat capacity model for COMPRO was defined by Eq. 2 [5], where each parameter was obtained by linearly approximating the specific heat capacity at ramp rate of 1.7 C/min.

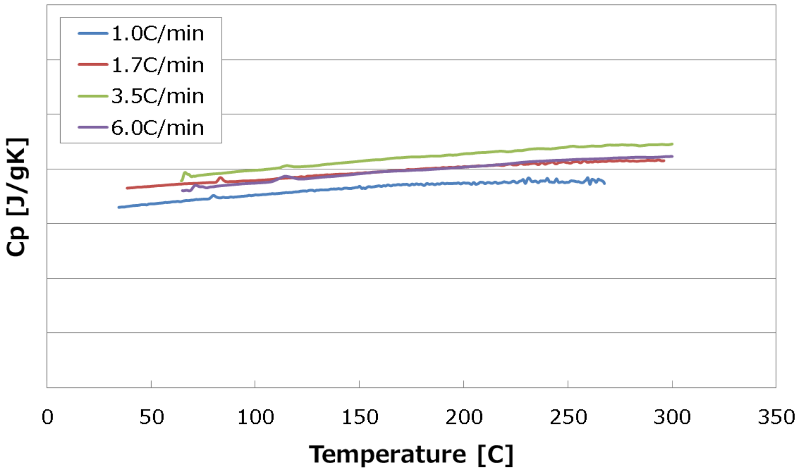


Figure 3. Raw data of specific heat capacity.

$$C_p = C_{p0} + T_f(T - T_0) \tag{2}$$

2.2. Periodical heating radiation-temperature measurement

Although, the laser flash method is well known as a measurement method for thermal conductivity, the measurable direction is limited to the thickness direction (Z-direction) of a sample, and a thin sample such as prepreg cannot be measured. In this study, periodical heating radiation temperature measurement method was applied to measure the thermal conductivity. The tests were performed on Thermowave Analyzer. It is capable to measure not only in the Z-direction but also in the in-plane direction (X, Y-direction) by periodically heating the samples at pinpoint by beam with a diameter of 100 to 150 μm. In addition, it can also be applied to thin samples.

Table 2 shows the measurement conditions. After measuring the thermal diffusivity of uncured samples (prepreg) and cured samples, the thermal conductivity was calculated by multiplying the density and the specific heat capacity of the materials. To evaluate the temperature dependence on thermal conductivity, the cured samples were measured under minimum temperature (25 C) and maximum temperature (185 C) of the recommended cure cycle. The uncured samples were measured at 25 C and 50 C at which the cure reaction of the resin is not started. The measurement directions were fiber direction (X-direction) and thickness direction (Z-direction) that can be input to COMPRO.

As shown in Figure 4, thermal conductivity showed the temperature dependence and the values in the X-direction was higher than that in the Z-direction. Since the uncured and cured thermal conductivity showed close values, the cured thermal conductivity was applied to COMPRO. The COMPRO input parameter was defined by Eq. 3 [5] in the X-direction and Eq. 4 [5] in the Z-direction.

Table 2. Measurement conditions of thermal conductivity.

Specimen Type	Ply number	Temperature (C)	Measurement Direction
Uncured	1 ply	25, 50	X, Z
Cured	1 ply	25, 185	X, Z

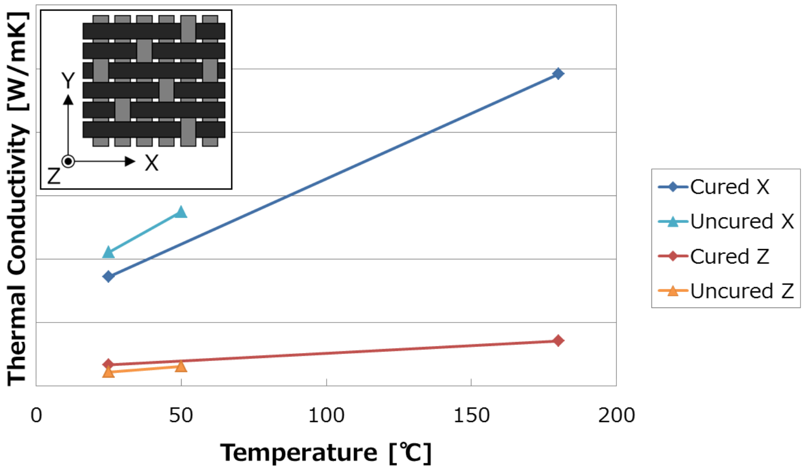


Figure 4. Measurement results of thermal conductivity.

$$K_l = K_{l0} + T_l(T - T_0) \tag{3}$$

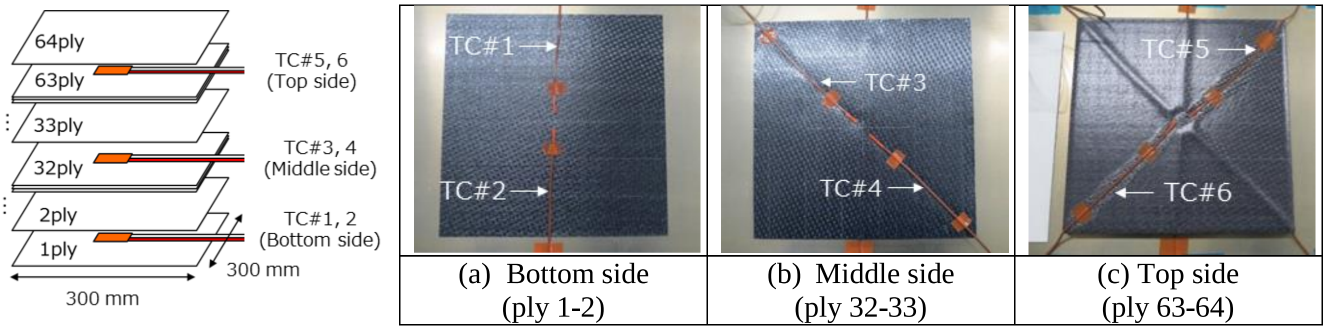
$$K_t = K_{t0} + T_t(T - T_0) \tag{4}$$

4. Verification test

4.1. Fabrication of thick composite laminate

To verify the prediction accuracy, thick composite laminate with thermocouples was fabricated to obtain detailed temperature profiles through the thickness of composite part during autoclave cure process. Composite laminate was made with 64 layers of carbon fiber fabric prepreg with a lay-up sequence of $[0/90]_{32}$. The dimension of the laminate was about 300 mm x 300 mm in plane and 18 mm in thickness.

As shown in Figure 5, thermocouples were placed in bottom side (between ply 1 and ply 2), middle side (between ply 32 and ply 33) and top side (between ply 63 and ply 64). Two thermocouples were installed at each measurement point.



(a) Bottom side (ply 1-2)
(b) Middle side (ply 32-33)
(c) Top side (ply 63-64)

Figure 5. Fabrication of thick composite laminate.

The composite part was placed on a 10 mm thick aluminum tool, and bagged as shown in Figure 6. A release film and a breather cloth were applied on the top of the composite part. The periphery of the composite part was insulated with the silicone rubber block. The autoclave cure cycle was set to manufacture's recommended condition shown in Table 3.

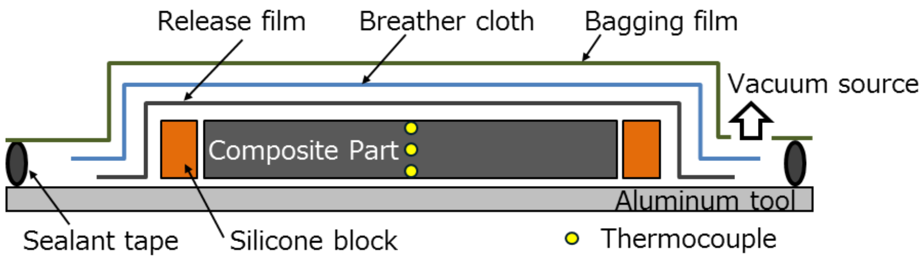


Figure 6. Schematic cross section of bagging configuration for laminate fabrication.

Table 3. Autoclave cure cycle for laminate fabrication.

Ramp rate (C/min)	Hold temperature (C)	Hold time (minutes)	Autoclave Pressure (MPa)
1.7	185	120	0.65

4.2. Cure process prediction

A two-dimensional finite element process modeling software, COMPRO was used to evaluate the experimental results. Figure 7 shows the COMPRO mesh model used for prediction. The convection boundary conditions were applied to the top surface of the composite part and the bottom surface of the tool. The convective heat transfer coefficient at the boundary between material and autoclave air was defined. The boundary conditions at the left and right side were set as adiabatic.

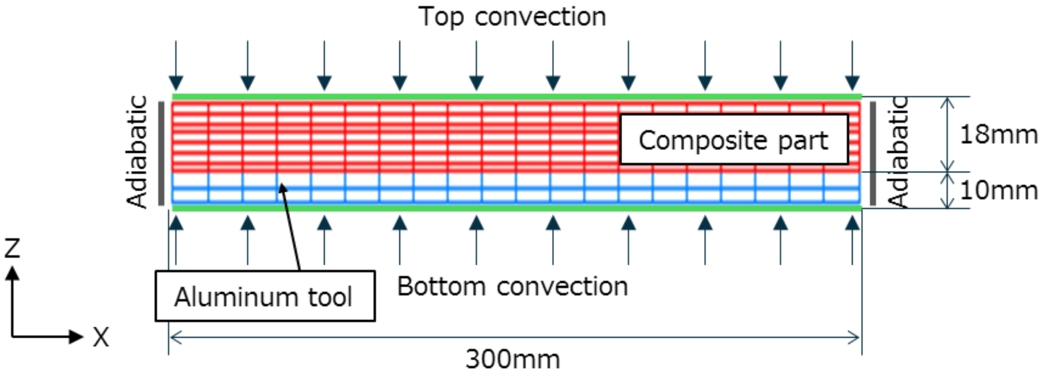
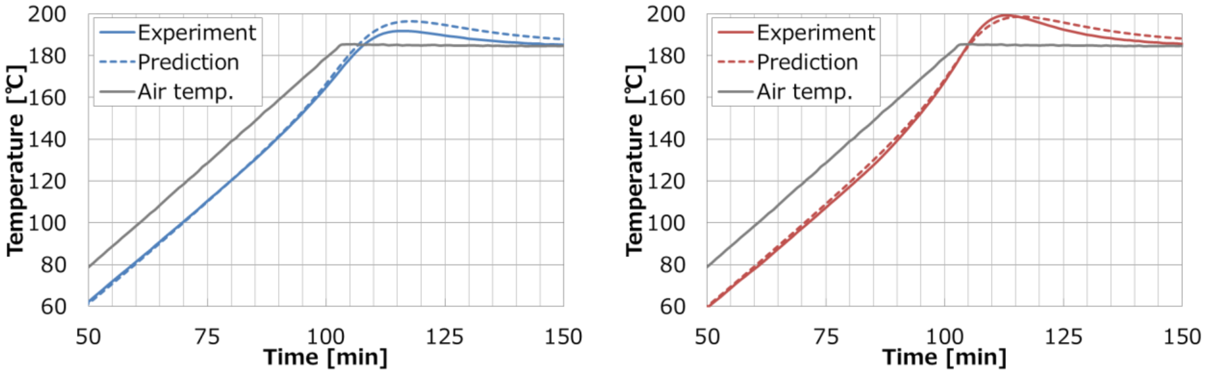


Figure 7. COMPRO mesh model for process prediction.

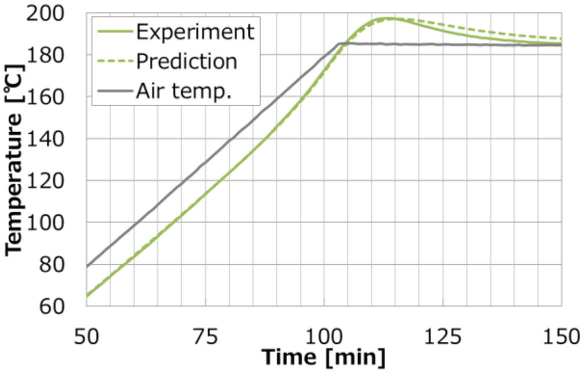
4.3. Comparison of experiment and prediction

Figure 8 shows the predicted temperature profiles at the bottom side, middle side and top side compared with experimental results. The comparison of the predicted results for the temperature profiles in the composite part showed good agreement with the experimental values. In the predicted temperature profiles, the temperature drop after the exotherm was slower than the experiment. This is attributed to the tool size effect and the three-dimensional effect of the composite part and tool. Although the composite part and tool were modeled with the same size in the X-direction (Figure. 7), the tool size was larger than the composite part in actual experiment and the heat conduction from the composite part to the tool after exotherm became faster.

The comparison results of exothermic peak temperature and peak arrival time are summarized in Table 4. The differences of exothermic peak temperature between experiment and prediction values were within 5 C of all measurement points. In addition, the arrival time to peak temperature also showed good agreement, and the differences were within 3 minutes. It is indicated that the thermo-physical properties determined in this study were appropriate.



(a) Bottom side (b) Middle side



(c) Top side

Figure 8. Comparison of temperature profiles between experiment and prediction.

Table 4. Comparison of peak temperature and peak arrival time.

Evaluation point	Peak temperature (C)			Peak arrival time (minutes)		
	Experiment	Prediction	Error	Experiment	Prediction	Error
Bottom	191.8	196.5	4.7	115	118	3
Middle	199.3	198.7	0.6	113	116	3
Top	197.5	197.2	0.3	113	115	2

5. Conclusions

In this study, the appropriate measuring method for thermo-physical properties was proposed to improve prediction accuracy of finite element process modeling software, COMPRO. As the measuring method, temperature-modulated DSC was chosen for determining heat of reaction, cure kinetics and specific heat capacity. The periodic heating and infrared radiation thermometer method was proposed for measuring the thermal conductivity.

In the verification test, the predicted temperature profiles in the composite laminate clearly showed good agreement with the experimental values. In particular, the difference of exothermic peak temperature between prediction and experimental values was within 5 C. In addition, the arrival time to peak temperature also showed good agreement, and the differences were within 3 minutes. It is indicated that the thermo-physical properties determined in this study were appropriate.

Through the application of preliminary prediction of temperature profiles in the composite parts, the number of tests for the development of cure process can be reduced, which will lead to cost reduction.

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