Predictions of Optimum Conditions for Pultrusion Process of Composite Materials by Numerical Approach

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ABSTRACT

The pultrusion process is one of the most typical manufacturing technique for fiber reinforced composites. This technique is to realize to manufacture the excellent composite materials which have high longitudinal strength and constant cross section.

In this study, the curing behavior of material during the pultrusion process was calculated by proposed numerical model which can consider the exothermic heat and the kinematics of resin. And the optimization procedure which can estimate simultaneously various optimum conditions of pultrusion process was proposed and evaluated by various conditions.

As a result, so that the validity of the proposed numerical model is confirmed in simulating the curing behavior of material in die during pultrusion process and the numerical result are good agreement with the experimental result. Furthermore, the proposed combined optimization procedure for the complex problem which have many design parameters is useful. This optimization procedure is that the number of design parameter can be reduced by deciding few design parameter as optimum condition in the first optimization operation and the rest of design parameter is calculated in the second optimization operation.

1. Introduction

The pultrusion process is one of the most typical manufacturing technique for fiber reinforced composites. This technique is to realize to manufacture the excellent composite materials which have high longitudinal strength and constant cross section.

In decent yeas, various studies about extrusion process are performed by experimental and numerical approach.[1][2] These studies mainly focused on behavior of materials in die and reported the viscosity, temperature, pressure and degree of cure of resin in die. To simulate these parameter in pultrusion process, the curing behavior of thermoset resin in die must be clear. So the numerical calculation of thermal transfer and curing behavior of resin with exothermic heat is performed by Aylward[3]. And many papers reported about numerical model for curing behavior in pultrusion process. However these studies are limited within influence of molding condition on pressure and pultrusion force in extrusion process, and there is few study which treat the optimum problem of molding condition in extrusion process[4][5]. Wu got optimum condition of temperature distribution of die and pultrusion speed by numerical analysis and experimental data. But this case used no mathematical optimum procedure but simple try and error method.

In this study, the numerical procedure which can estimate curing ratio of materials in pultrusion process is proposed due to simulate the behavior of materials in die. Furthermore the optimization method of molding condition for pultrusion process is also proposed by using curing estimation method.

2. Pultrusion model

Fig. 1 shows schematic diagram of pultrusion process[6]. This process is divided into five steps. These steps are supplying part of reinforcing fiber, resin impregnation part, curing part of resin, pull out part and cut, trimming part. The material behavior in curing part decides quality and performance of product. The models of curing behavior in curing part had been developed by experimental and theoretical procedure. Fig. 2 shows curing model which is to be summarized these curing models[1]. This model defined that the material in the die can be divided into core part of reinforcement with resin and surface part of resin. And this model is divided into three different area due to curing behavior of material in die. The first zone is heating stage in which temperature of resin is increased by heating from die. In this zone, pultrusion force is dominated by friction between die and fluid resin. The second zone is curing stage. Temperature is increased rapidly by reaction heat of resin and in this zone, pultrusion force is controlled by adhesive force of resin with die and friction between die and solid resin. The last zone is solid stage in which pultrusion force is friction between die and solid resin. In all stage, the curing and flow behavior of material can be assumed to be steady state. This study adopt the above curing model to optimization analysis.

3. Analysis method

In this study, the steady state model mentioned in previous chapter is used.

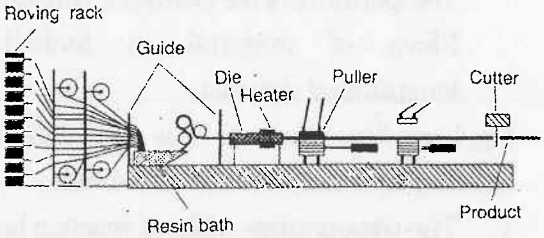


Fig1 Schematic diagram of pultrusion process.

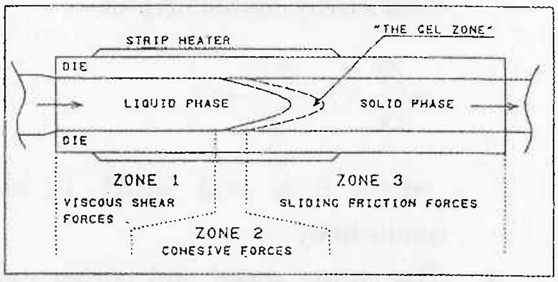
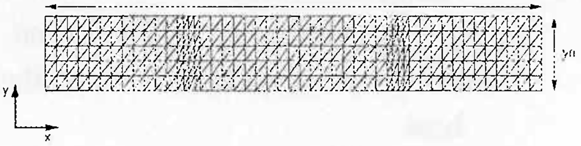


Fig.2 Three zone model of pultrusion process



$y=y_0$ and $0 \leqq x \leqq L$, $T=T(x)$ (die wall temperature profile)
$x=0$ and $0 \leqq y \leqq y_0$, $T=300K$ (Initial temperature profile)
$y=0$ and $0 \leqq x \leqq L$, $(\partial T/\partial x)=0$ (symmetry condition)
$L = 820$ mm, $y_0 = 2.08$ mm

Fig.3 Finite element division of calculation area

The calculation of heat transfer is performed by finite element method. The pultrusion die with 20.8*820mm rectangle cavity is adopted as analysis region. Fig.3 shows the finite element division of calculation region. This calculation area is limited in the half of the analysis region with considering of symmetry. In this calculation, following assumptions is adopted;

Curing and flow behavior of material can be assumed to be steady state.

Flow speed of material in die is uniform.

Heat transfer is limited in thickness direction.

Temperature of die surface is constant.

Move of material is limited in longitudinal direction.

Fig.4 is flow chat of this calculation for estimation of temperature distribution.

1. The temperature without reaction heat of material in die is calculation by following 2-dimensional steady state heat conductivity equation(Q=0).

$$u\frac{\partial T}{\partial x} = ky\frac{\partial^2 T}{\partial y^2} + Q \qquad (1)$$

where u is pull speed, k_y is heat conductivity.

2. The curing speed and curing ratio are calculated.
3. The exothermic heat is evaluated by curing kinematics.
4. The temperature distribution is calculated by equation 2 with exothermic heat.

In this calculation, curing kinematics is calculated by following equations

$$\frac{d\alpha}{dt} = k(1-\alpha)^n \qquad (2)$$

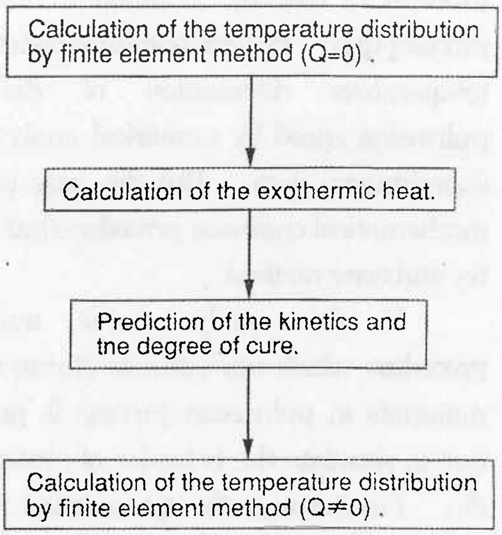


Fig.4 Flow chart of curing behavior simulation

$$k = A_0 \exp\left(-\frac{E}{RT}\right) \qquad (3)$$

where α, A₀, E, R and T are curing ratio, Aulenius constant, exothermic energy, gas constant and temperature respectively. A_0, E and n are obtained from DSC analysis. And equation 2 is integrated by time t. The curing ratio is obtained by following equations.

$$\begin{aligned} \alpha &= 1 - \exp(-kt) \quad (n=1) \\ \alpha &= 1 - \left[(n-1)kt + 1\right]^{\frac{1}{1-n}} \quad (n \neq 1) \end{aligned} \qquad (4)$$

The curing kinematics is obtained by substituting eq.4 to eq.3.

$$\begin{aligned} \frac{d\alpha}{dt} &= k\left[\exp(-kt)\right]^n \quad (n=1) \\ \frac{d\alpha}{dt} &= k\left[(n-1)kt + 1)\right]^{\frac{n}{1-n}} \quad (n \neq 1) \end{aligned} \qquad (5)$$

And exothermic heat is

$$Q = \Delta H\frac{d\alpha}{dt} \qquad (6)$$

where ΔH is total exothermic heat of resin.

The temperature distribution in die is obtained by using above equations. In

Table 1 Material properties of composites

	Density (g/cm)	Heat capacity (cal/g·K)	Heat transfer coefficient (cal/cm·s·K)
Fiberglass	2.55	0.197	2.48×10^{6}
Unsaturated polyester	1.11	0.45	4.05×10^{-7}

Table 2 Kinetic parameters of unsaturated polyester

Parameter	Symbol	Units
Pre-exponetial constant	A_0	1.59×10^{8}(1/s)
Activevation energy	ΔH	74.8 (cal/g)
Heat of reaction	E	14.94(kcal/g·mol·K)
Order of reaction	n	1.94

equation 4, heat transfer is assumed to be steady in order to by unsteady in actual pultrusion stage. So in the case of the estimation of curing ratio of resin, the curing ratio is assumed to be a function of temperature T and time t.

To clarify this calculation procedure, the calculation is performed in case of a constant temperature at die surface which had been reported by Ma[7] in the experimental approach. Fig.5 shows temperature profile along die surface and these are applied to the boundary conditions. Table 1, 2 are material properties and kinetic variables of resin respectively. In this case, the material is grass reinforced unsaturated polyester resin. From these conditions the curing ratio and temperature distribution of material in die are calculated. Fig. 6,7 show numerical result and experimental result which had been reported by ref.[7]. The numerical result shows good agreement with the experimental result. Therefore, it is ensured that the proposed numerical procedure is available to estimate the curing behavior of material during pultrusion process.

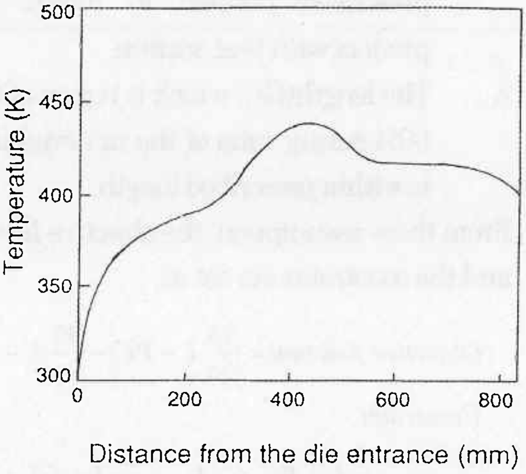


Fig.5 Die wall temperature distribution

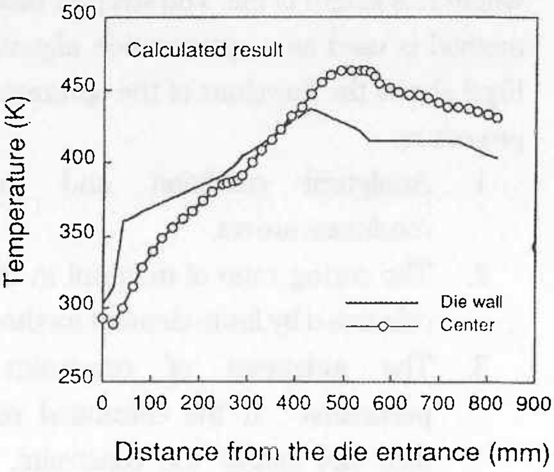


Fig.6 Temperature distribution (Calculated result)

4. Optimization procedure

In this optimization, the wall temperature of die and pultrusion speed are selected as design parameter, because these parameter can be changed easily in actual pultrusion process. The wall is divided into three zone as same as Fig.2 and wall temperatures at each zone are assumed to be control independently. To decide the optimum conditions of pultrusion process, the condition which realize the best surface of product and pultrusion force as follow is selected[5].

1. The position(PC) where the curing ratio of the material reach 90% is located on prescribed position to realize good product with best surface.
2. The length(GL) which is region of 60%-90% curing ratio of the material in die is within prescribed length.

From these assumption, the objective function and the constraint are set as

$$Objective\ function = \left|\frac{13}{24}L - PC\right| - \left|\frac{13}{48}L - GL\right|$$

$$Constraint$$

$$\frac{1}{3}L < PC < \frac{3}{4}L \quad , \quad \frac{5}{24}L < GL < \frac{1}{3}L$$

where L is length of die. The steepest descent method is used as a optimization algorithm. Fig.8 shows the flowchart of the optimization procedure.

1. Analytical condition and initial conditions are set.
2. The curing ratio of material in die is calculated by finite element method.
3. The judgment of constraint is performed. If the calculated result does not satisfy the constraint, the objective function is added to a penalty value.
4. The value of objective function is calculated.
5. The judgment of improvement of objective function is performed. If the objective function is improved, the design parameters are changed by steepest descent method.

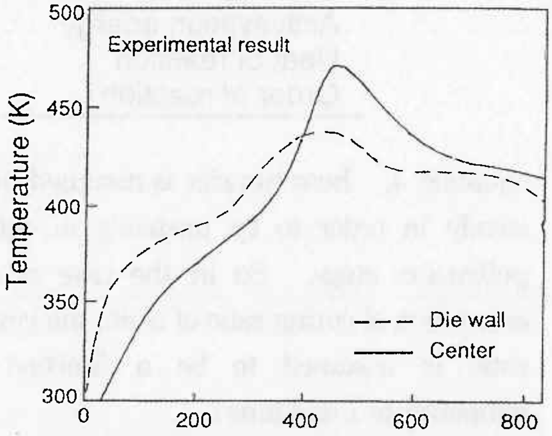


Fig.7 Temperature distribution (Experimental result)

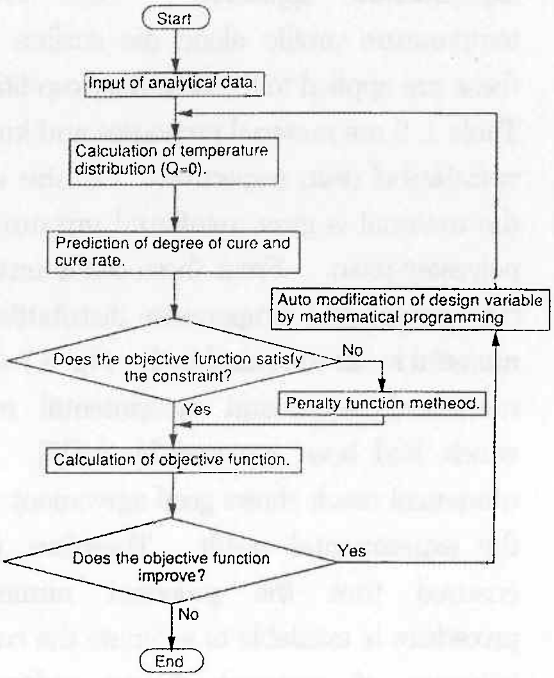


Fig.8 Flowchart of optimization procedure

Table 3 Optimization result

	Initial condition	Optimized condtion
Die wall temperature (K)	350-400-400	357-411-411
Pull speed (mm/s)	10.73	10.86

Table 4 Optimization results in the different initial conditions

	Initial condition	Optimized condtion
Die wall temperature (K)	342-390-390	357-411-411
Pull speed (mm/s)	10.46	10.86
Die wall temperature (K)	350-400-400	357-411-411
Pull speed (mm/s)	10.73	10.86
Die wall temperature (K)	360-410-400	361-411-411
Pull speed (mm/s)	11.00	11.00

And in other case, this optimization procedure is over.

The material properties and boundary conditions are same as those in previous chapter.

5. Results and discussions

Table 3 is numerical results obtained by the optimization procedure. To evaluate this numerical result as optimum condition, three calculations with different initial conditions are performed. Table 4 is these numerical results. In these result, each calculated results show different value. It is considered that this result shows that the solution field of the pultrusion process have several local optimum conditions. Therefore, the estimation of the optimum condition of the pultrusion process is difficult by the simple optimization procedure.

The combined optimum procedure is considered to get the optimum condition under above circumstances. This procedure include two optimization stage. In table 4, the wall temperature 2 and pull speed show constant value under various initial conditions. So these design parameters are assumed to be get as optimum value, and in next optimization operation, these parameters are omitted from design parameters and are fixed as first optimization result. The second optimization operation is performed under the limited conditions. In this case, the design parameter is only limited in wall temperature 1, linear search method is adopted as optimization operation.

The second optimizing calculations are performed under three initial conditions (wall 1 temperature 350, 360 and 370K). As a result, the same result which is wall 1 temperature 385K is obtained in all initial conditions. So in this pultrusion process, the wall temperature 382-411-411K and pull speed 10.86mm/s can be assumed to be optimum conditions.

Therefore, for the complex problem which have many design parameters such as pultrusion process, it is useful that the number of design parameter reduced by deciding few design parameter as optimum condition by the first optimization operation and the rest of design parameter is calculated by second optimization operation.

6. Conclusions

In this study, the curing behavior of material during the pultrusion process was calculated by proposed numerical model which can be considered the exothermic heat and the kinematics of resin. And the optimization procedure which can estimate simultaneously various optimum conditions of pultrusion process was proposed and evaluated by various conditions.

As a result, the proposed numerical model is useful to simulate the curing behavior of material in die during pultrusion process and the numerical result is good agreement with the experimental result. Furthermore, it is useful that proposed combined optimization procedure applied to the complex problem such as pultrusion process. These proposed optimization techniique is valuable for various polymer processing process because the optimization part exist independently form the numerical analysis method.

7. References

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