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EXPERIMENTAL ESTIMATION OF PROCESS PARAMETERS IN RESIN TRANSFER MOLDING

Technical Report

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ABSTRACT

A numerical method is presented for the estimation of experimental resin transfer molding (RTM) process parameters from in-mold temperature data. The approach is built around the simulation of either radial or axial flow of reactive resins through a fiber preform inside a mold cavity. The simulation considers a non-isothermal environment, with one-dimensional flow and one-dimensional heat transfer, and allows for either pressure or flow rate controlled inlet conditions. Based on the observed temperature history at a selected point in the mold, the best fit values of the unknown process parameters are obtained by minimizing the difference between the simulated and experimental temperature histories. This minimization is accomplished by integrating the simulation model with a MINPACK nonlinear least-squares routine (1). As an example, the chemical kinetic parameters for a step-wise polyurethane reaction are estimated based on temperature data reported by González (2). Simulated temperature histories using the parameter values obtained agree very well with experimental results.

KEY WORDS: Resin Transfer Molding, RTM, Estimation, Model, Fitting

1 INTRODUCTION

Resin transfer molding (RTM) and its higher speed relative, structural reaction injection molding (SRIM), are being widely recognized as having great potential for increased use in the production of composite parts. RTM is typically used in lower-volume, higher performance applications, such as in the aerospace industry. SRIM has received attention especially in the ground vehicle industry, where there is considerable interest in obtaining the benefits of polymer composite materials with low cost and high volume production rates. Benefits of RTM/SRIM processing include the ability to produce reduced weight replacements in existing applications and to reduce manufacturing costs (3). RTM and SRIM have some important advantages over thermoplastic injection molding, including low processing temperatures and reduced clamping pressure requirements (4, 5, 6, 7).

Currently, RTM/SRIM manufacturing processes are developed largely by trial and error, incurring significant expense during the design process and yielding potentially less-than-optimal process designs. Much progress has been made in recent years in developing accurate models of the process, which could be used to anticipate process difficulties and identify appropriate process parameters. Unfortunately, many of the physical parameters required to perform accurate simulations can be very difficult to obtain. Examples include heat transfer coefficients, thermal conductivities, edge and corner permeabilities, and chemical kinetic parameters.

The objective of this work has been to develop an efficient numerical method for obtaining physical parameter values under real molding conditions from in-mold temperature data. This is accomplished by minimizing the difference between the simulated and experimental temperature history using a non-linear least-squares optimization algorithm (1). The simulation model, which must be very fast to perform hundreds executions for a single fitting, uses a simplified one dimensional flow and one dimensional heat transfer. In order to verify the parameter estimation method, chemical kinetic parameters are estimated for a styrene-dimethacrylate copolymer material. The experimental temperature data are from González (2), representing constant volumetric flow rate injection in a radial mold.

2 SIMULATION MODEL

A Lagrangian framework is employed, in which the coordinate system is fixed on a particle as it moves radially outward. Since the field variables are symmetric about the midplane of the mold, the solution is sought between the midplane and the top of the mold. A finite-difference scheme is used to discretize both time and space.

This mathematical work is based on the conservation of volume and energy. Emphasis is placed on making appropriate assumptions to simplify the calculations while representing the major features of the process. The equations are expressed in terms of temperature.

2.1 Basic Assumptions In both RTM and SRIM, the thickness of mold cavity is usually small. This combined with the resistant pressure of fiber is assumed to result in a flow front with a flat profile (8). The range of the temperature variation is assumed to be small enough (21°C to 250°C in this study) that the thermal conductivity k, the heat capacity C, and the density ρ do not depend on temperature. The fluid is assumed to be incompressible and body forces are neglected. The heat transfer in the radial direction is considered negligible in comparison with the heat transfer in the transverse direction because the mold cavity is thin and the corresponding temperature gradients are much greater in the transverse direction.

Heat transfer between the resin and the fiber preform is assumed to occur on a much shorter time scale than between the resin and mold due to the large ratio of surface area to volume of the fibers. Following the example of González (2) and Lin (9), the temperature of the fiber and the polymer are assumed to reach equilibrium within one time step after the resin reaches the fiber. Based on the conservation of energy, this equilibrium temperature can be obtained by:

$$T = \frac{m_r C_r T_r + m_f C_f T_f}{m_r C_r + m_f C_f} \tag{1}$$

Table 1: Thermal Properties
Data from González-Romero (2)

parameter	value
ρ_p (g/cm^3)	1.1
ρ_f (g/cm^3)	2.54
C_p (cal/g °C)	0.45
C_f (cal/g °C)	0.2
k_p (cal/sec cm^3)	0.0004
k_f (cal/sec cm^3)	0.0016
k_m (cal/sec cm^3)	0.56
λ (cal/°K cm^3)	1.0468×10^{-5}

where m_r is the mass of resin, m_f is the mass of fiber, C_r is the heat capacity of resin, C_f is the heat capacity of fiber, T_r is the temperature of resin, T_f is the temperature of fiber, and T is the equilibrium temperature of the resin/fiber. The properties of the resin/fiber are obtained using a equivalent heat capacity and density, and a harmonic average (10):

$$C = \frac{m_r C_r + m_f C_f}{m_r + m_f} \tag{2}$$

$$k = \frac{k_r k_f}{(1-\Phi)k_r + \Phi k_f} \tag{3}$$

$$\rho = \Phi \rho_r + (1-\Phi)\rho_f \tag{4}$$

where C is the heat capacity of the resin/fiber, k, k_r, and k_f are the thermal conductivities of the resin/fiber composite, resin, and fiber respectively, ρ, ρ_r, and ρ_f are the densities of the resin/fiber composite, resin, and fiber respectively, and Φ is the porosity which is the volume fraction of resin. The values of these thermal properties are listed in Table 1.

2.2 Governing Equations The conservation of energy leads to the energy equation for a given material particle:

$$\rho C \frac{\partial T}{\partial t} = k \frac{\partial^2 T}{\partial y^2} + \dot{q} \tag{5}$$

where t is time and y is the dimension in the transverse direction, and $\dot{q}$ is the heat generation rate. The initial conditions can be obtained from Equation 1 by substituting $T_f = T_m$ and $T_r(t=0) = T_o$. T_m is the mold temperature and T_o is the initial temperature of resin. The boundary conditions are

$$k \left. \frac{\partial^2 T}{\partial y^2} \right|_{y=\frac{H}{2}} = \begin{cases} \frac{h}{\Delta y}[T_m - T] & \text{during filling} \\ \frac{k_b}{\Delta y^2}[T_m - T] & \text{during curing} \end{cases} \tag{6}$$

$$\left. \frac{\partial T}{\partial y} \right|_{y=0} = 0 \tag{7}$$

where h is the heat transfer coefficient and k_b is the boundary thermal conductivity at the wall between mold and polymer. The value of h is based on the radial velocity of resin, v_r, as described by González (2):

$$h = \lambda v_r \tag{8}$$

where λ is the heat transfer coefficient constant with the value given in Table 1. The boundary thermal conductivity k_b is given by:

$$k_b = \frac{2kk_m}{k + k_m} \tag{9}$$

Based on conservation of volume, the continuity equation is

$$v_r = \frac{Q}{2\pi \Phi r H} \tag{10}$$

where Q is the flow rate, r is the radial position, H is the mold thickness, and v_r is the velocity in radial direction.

2.3 Numerical Scheme During the filling stage, each resin segment moves out like a circular ring as the mold is filled. For constant flow rate mold filling, the one dimensional flow assumptions mean that the location of the flow front does not need to be found numerically but can be calculated directly from Equation 10 at any point in time. As described above, the heat transfer within the Lagrangian framework is assumed to be 1-dimensional in the transverse direction (negligible radial conduction), and the symmetry with respect to the midplane of the mold reduces the calculation domain to one half of the mold thickness.

Each circular ring of the resin/fiber composite is divided into several segments in the y–direction; ten segments were used for the results reported in Section 5. Unconditional stability was achieved by implementing a fully implicit numerical scheme, with backward differences in time and central differences in space used to discretize the governing equation (Equation 5). In this approach, the calculations for heat conduction are based on the unknown current temperatures, while the temperature of the material at the end of the previous time step is used in the solution of the kinetic equations to calculate the heat generation term.

Using the superscript, i to represent time and the subscript, j to represent the position in the y-direction, the energy equation can be expressed as:

$$T_j^i - T_j^{i-1} = \frac{k\Delta t}{\rho C \Delta y^2}[T_{j+1}^i + T_{j-1}^i - 2T_j^i] + \frac{\Delta t}{\rho C}\dot{q}(T_j^{i-1}) \tag{11}$$

where Δt is the time step and Δy is the thickness in y–direction of each segment. The subscript j ranges from 1 at the mold wall to N at the midplane, while i varies from 1 for the first material entering the mold to M for the last. To simplify the equations, a dimensionless heat conduction parameter β is defined as follows:

$$\beta \equiv \frac{\Delta t k}{\rho C \Delta y^2} \tag{12}$$

Equation 11 then becomes:

$$T_j^i - T_j^{i-1} = \beta[T_{j+1}^i + T_{j-1}^i - 2T_j^i] + \frac{\Delta t}{\rho C}\dot{q}(T_j^{i-1}), \quad i = 1, M, \quad j = 2, N-1 \tag{13}$$

At the midplane and at the mold wall, applying the boundary conditions leads to slight modifications of Equation 13. Considering symmetry about the mold midplane, the governing equation at the center of the cavity can be written as:

$$T_N^i - T_N^{i-1} = 2\beta[T_{N-1}^i - T_N^i] + \frac{\Delta t}{\rho C}\dot{q}(T_N^{i-1}) \tag{14}$$

Table 2: Kinetic Parameters
Data from González-Romero (2)

parameter	value
$t_Z(\mathrm{s}^{-1})$	$B_1 \exp(B_2/T)$
$k_X(\mathrm{s}^{-2})$	$A_X \exp(-E_X/T)$
k_Z/k_P	25
$A_X(\mathrm{s}^{-2})$	1.15×10^6
$E_X(^\circ\mathrm{K})$	-9.0×10^3
$B_1(\mathrm{s}^{-1})$	2.05×10^{-20}
$B_2(^\circ\mathrm{K})$	1.91×10^4

At the mold wall, the governing equation can be written:

$$T_1^i - T_1^{i-1} = \beta_m[T_m - T_1^i] - \beta[T_1^i - T_2^i] + \frac{\Delta t}{\rho C}\dot{q}(T_1^{i-1}) \tag{15}$$

where β_m is

$$\beta_m = \begin{cases} \dfrac{\Delta t h}{\rho C \Delta y} & \text{during filling} \\ \dfrac{\Delta t k_b}{\rho C \Delta y^2} & \text{during curing} \end{cases} \tag{16}$$

This numerical scheme converges with a relatively large time step. An adaptive time step was selected for subsequent calculations. A further numerical simplification employed based on the stated assumptions allows the tracking of only the material point or points of interest once filling is complete. Since filling is usually a small fraction of the total simulation time, this allows tremendous additional time savings when only one or a few locations contain information of interest.

3 POLYMERIZATION KINETICS

As an example, the critical kinetics parameters for a styrene-dimethacrylate (SDM) copolymer material model based on González' work are estimated in the results presented below. This polymerization system contained equal amounts, by mass, of ethoxylated bisphenol A dimethacrylate and styrene. The resin also contained 1% by weight of paratertiary butyl peroxide used as a free radical initiator, and an undetermined amount of hydroquinone which is contained as an inhibitor in the commercial resin. The inhibitor was used to delay reaction during the filling stage, and to reduce the risk of excessively high temperature during curing which can degrade the material. The material reacts via free radical co-polymerization, setting its shape by crosslinking.

The heat generation rate, $\dot{q}$, is given by the following equations:

$$\dot{q} = \Phi H_r \frac{\partial X}{\partial t} \tag{17}$$

$$H_r = \rho_r C_r T_{ad} \tag{18}$$

where Φ is the porosity, H_r is the chemical heat of reaction, X is the conversion of monomer, ρ_r is the density of resin, C_r is the heat capacity, and T_{ad} is the adiabatic temperature rise, which for this system was reported to be 162°K. There are three species in this chemical system: the monomer, the radicals, and the inhibitor. The equations of species balance are given by the following set of differential equations, where R^* is the conversion of radicals, and Z^* is the conversion of inhibitor:

$$\frac{\partial X}{\partial t} = k_X R^*[1 - X] \tag{19}$$

$$\frac{\partial R^*}{\partial t} = 1 - k_X t_Z \frac{k_Z}{k_P}[1 - Z^*]R^* \tag{20}$$

$$\frac{\partial Z^*}{\partial t} = k_X \frac{k_Z}{k_P}[1 - Z^*]R^* \tag{21}$$

where k_X represents the activation energy and is given by

$$k_X = A_X \exp(-E_X/T) \tag{22}$$

and t_Z is the inhibition time, given by

$$t_Z = B_1 \exp(B_2/T) \tag{23}$$

The ratio k_Z/k_P is assumed to be constant, and A_X, E_X, B_1, and B_2 are treated here as the unknown parameters to be estimated.

The initial conditions for all of these three species are zero. The polymer gels when the conversion of the monomer reaches 0.64% (0.0064). The parameters reported by González (2) were obtained using isothermal DSC experiments, and are reproduced in Table 2.

4 LEAST-SQUARES REGRESSION

The regression analysis deals with an investigation of the relationship between two or more variables related in a nondeterministic fashion (11). For this problem, the temperature is a function of the unknown process parameters (e.g. kinetic parameter values), and the relationship involves a complex nonlinear simulation model. The nonlinear regression process requires first making a guess for initial values of the unknown parameters. The simulated temperature history is then compared with the experimental temperature history data. Based on the total integrated temperature difference and the derivatives of this difference with respect to the unknown parameters, a direction to adjust the values of the parameters is identified. The least squares method, which can be traced back to the German mathematician Gauss (1777-1855), is used to obtain this best fit to the observed data points (12).

In this example, we assume temperature is a function of the unknown kinetic parameters, $\vec{p}$, $\vec{p} = (A_x, B_1, B_2, E_x)$. We can write the simulated temperature as $T_s(A_x, B_1, B_2, E_x)$ or $T_s(\vec{p})$. By the least squares method, we can write

$$F = \begin{bmatrix} T_{s1} - T_{e1} \\ \vdots \\ T_{sN} - T_{eN} \end{bmatrix} \tag{24}$$

where E is the residual vector, T_e is the experimental temperature history data at one sensor location during the molding time, and the subscripts $1, \cdots, N$ are the number of experimental data.

Table 3: Molding Conditions
Data from González-Romero (2)

parameter	Exp.1	Exp.2	Exp.3	Exp.4	Exp.5
R_m(cm)	10	10	10	10	10
H(cm)	0.59	0.58	0.63	0.59	0.52
Φ	0.764	0.764	0.764	0.764	0.764
Q(cm^3/sec)	24	22	25	24	27
T_o(°K)	303	297	294	303	294
T_m(°K)	395	411	399	395	407
r(cm)*	3.2	3.2	3.2	3.2	2.3
ζ*	0.1	0.1	0.1	0.1	0.1

* sensor location

Table 4: Kinetic parameter estimation for A_x, E_x, B_1, and B_2

Exp.#	A_x	E_x	B_1	B_2
1	0.10120E+07	-8453.0	0.92278E-19	19093.
2	0.12986E+07	-8836.1	0.18134E-19	19210.
3	0.20197E+07	-8580.0	0.26283E-19	19446.
4	0.17327E+07	-8620.7	0.83270E-19	19197.
Ave.	0.1516E+07	-8622.5	0.5499E-19	19236.5

For the nonlinear least squares minimization, the Levenberg-Marquardt method (13) is efficient, and has become a standard for nonlinear least squares curve fitting. It is based on calculating the first partial derivatives of the residual F with respect to the unknown parameters. The Jacobian matrix is (14)

$$J(\vec{p}) = \frac{\partial F}{\partial \vec{p}} \tag{25}$$

The step direction and length are calculated from the Jacobian matrix. This work used the MINPACK public domain optimization library implementation which uses a trust region approach to the Levenberg-Marquardt method (1).

5 RESULTS AND DISCUSSION

Five experimental runs from the work of González (2) are presented here, with the molding conditions given in Table 3. A sixth run was not included, although the fit was comparable, because it contained some unexplained anomolies. The parameters are estimated individually using the approach just described for each of the first four runs. The averages of these fitted values were then used in a fully 2-D simulation to evaluate the predictive capability of the fitted parameters.

The parameter values obtained by fitting the four kinetic parameters are summarized in Table 4 together with their averages. The simulated temperature histories with these parameter values and the experimental data are shown in Figures 1 to 4, demonstrating the quality of the fit achieved. The main discrepancy apparent in all four cases is that

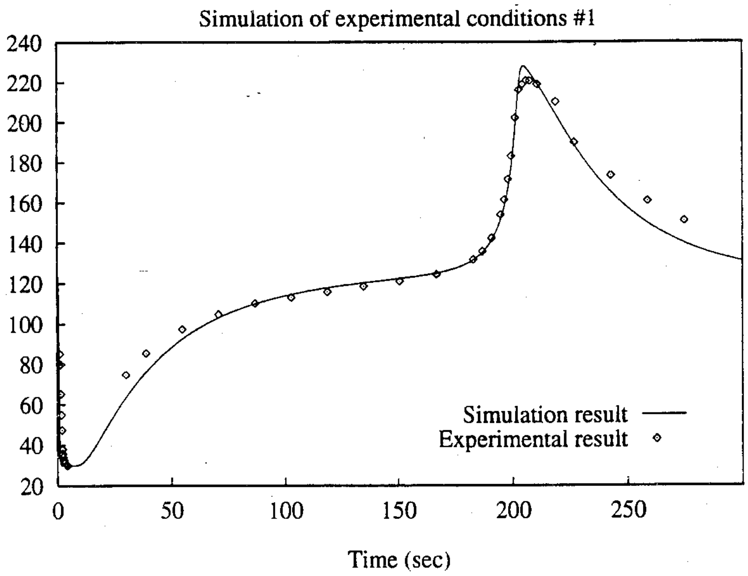

Figure 1: Experimental and fitted temperature vs. time for experimental conditions 1 in Table 3.

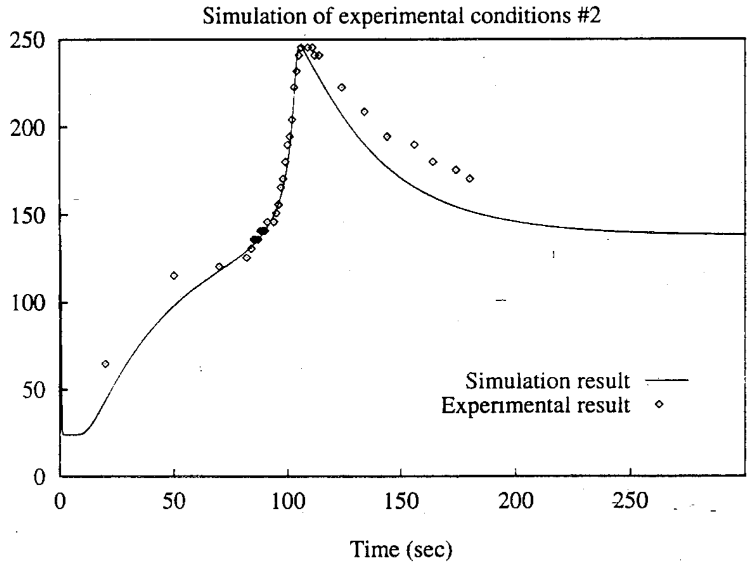

Figure 2: Experimental and fitted temperature vs. time for experimental conditions 2 in Table 3.

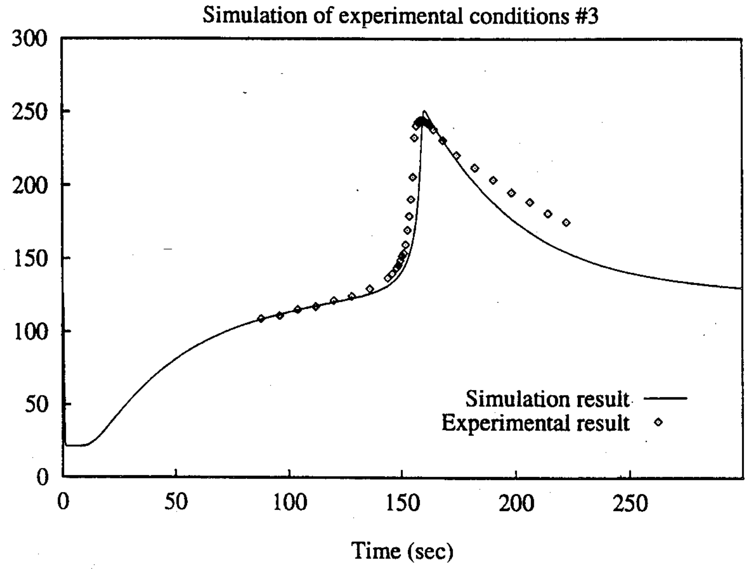

Figure 3: Experimental and fitted temperature vs. time for experimental conditions 3 in Table 3.

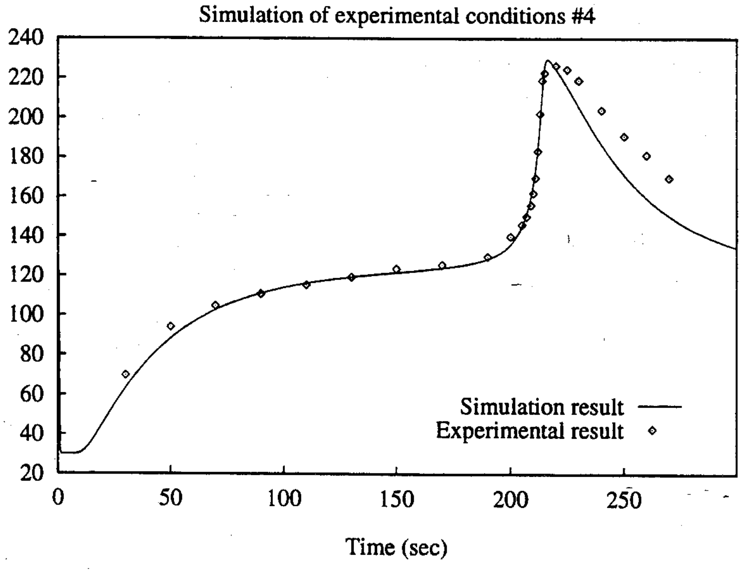

Figure 4: Experimental and fitted temperature vs. time for experimental conditions 4 in Table 3.

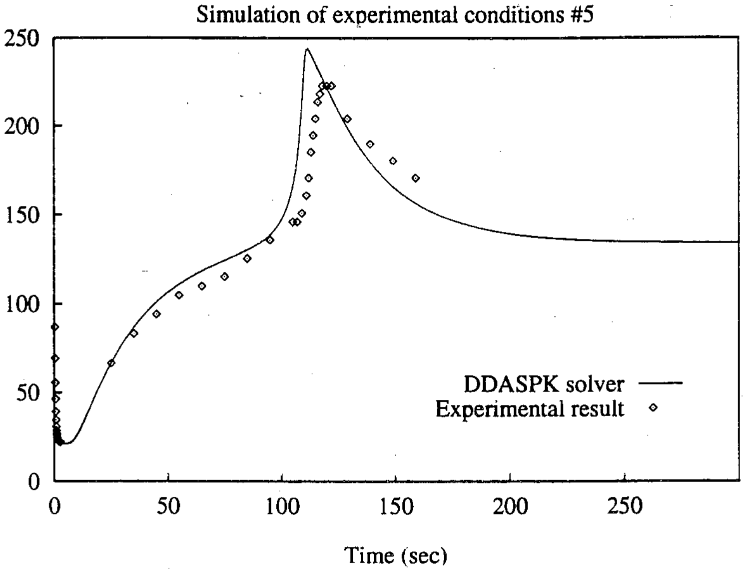

Figure 5: Comparison of predicted and experimental temperature vs. time using a fully two-dimensional model and the average estimated parameters for experimental conditions 5 in Table 3.

the rate of cooling observed following the peak temperature was consistently slower than that predicted by the simulation model. It is believed that this is due to an increase in actual mold temperature during the experiments which is not modeled numerically. Since this part of the fit is quite insensitive to the values of the kinetics parameters used, and since there are in no case a large number of data points in this region, this discrepancy does not appear to have introduced significant error into the parameter estimates.

Following the paramter estimation from the first four cases, we used the average of the values obtained to simulate the RTM process under the experimental condition #5. For this simulation, a fully two-dimensional represention of the problem was solved using the partial differential equation solver DASPK (15). In this simulation, The chemical reaction and heat transfer are solved simultaneously, and the previously neglected in-plane heat conduction is considered. The simulated temperature history is in good agreement with the experimental data, as shown in Figure 6. This suggests that the fitted parameters, while varying slightly from case to case, are useful in predicting behavior for conditions not necessarily identical to those in which they were fit. It is also seen that the parameters should be appropriate for use in more complex two- and three-dimensional codes.

As a final check on the validity of the assumptions employed in the model presented here, the same averaged parameter values were used to compare the simulated temperature histories from the presented model and the two-dimensional DASPK solution. The comparison is shown in Figure 5, where almost no difference can be discerned between the two results. It is reasonable to expect that this would not be true at every point in the mold, as neglecting in-plane conduction is known to introduce errors that can

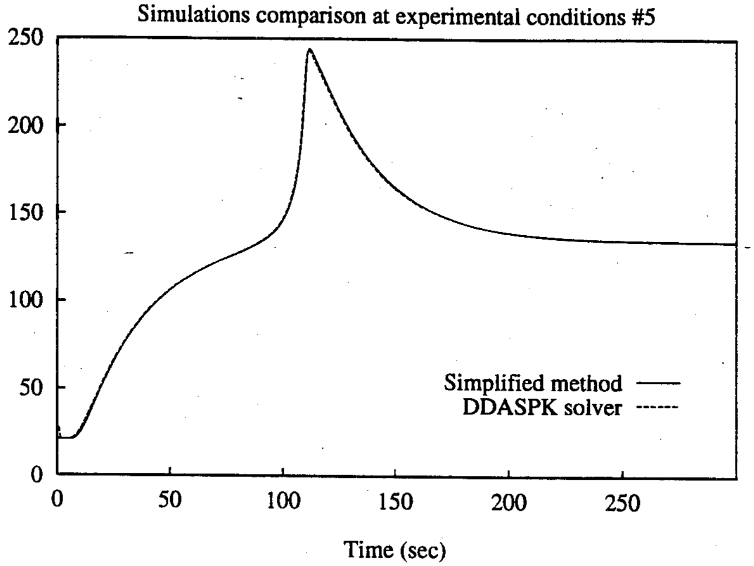

Figure 6: Comparison of simplified one-dimensional and fully two-dimensional model predictions using the same kinetic parameters for experimental conditions 5 in Table 3.

be significant near the flow front. However, for the purpose of parameter estimation, it is clear that the approximations used are appropriate for the sensor locations used in this set of experiments. Since the fully two-dimensional solution required roughly 1000 times more computing time than the model presented here (>1000 seconds vs. 1 second on an engineering workstation), this simplified model is much more appropriate for the purposes of parameter estimation. The importance of this can be emphasized by noting that the estimations presented here typically required several hundred simulations each.

6 CONCLUSION

A parameter estimation study for resin transfer molding has been developed based on a one-dimesional simulation model with coupled flow, chemical reaction and heat transfer for resin transfer molding, using nonlinear least squares regression. Excellent fits were obtained for each experimental condition studied. The fitted paramter values were seen to have useful predictive value for experimental conditions not included in the fitting process. Finally, excellent agreement between a fully two-dimensional simulation and the predction of the simplified model for the same experimental conditions and parameter values demonstrates the appropriateness of the assumptions employed.

In a further study, a more complex package in parameter estimation is envisioned, in which the user can select a kinetic model and find the best values of the parameters for that model. Since the model is capable of simulating either axial or radial flow, and either constant flow rate or constant pressure injection, a range of experimental conditions will be able to be considered for parameter estimation. It is also envisioned that the approach may be useful for estimating difficult to measure thermal or other properties.

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