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**ON PROCESSING DEVELOPMENT FOR FABRICATION OF
FIBER REINFORCED COMPOSITE - II**

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FOREWORD

This work was sponsored by and performed at the Polymeric Materials Branch (PMB), NASA Langley Research Center (LaRC). Mr. Robert M. Baucom (PMB) was the Technical Monitor for this investigation.

High performance fiber reinforced composite materials have found increasing usages in the aerospace and automobile industries in recent years. Typically, these composite materials are available as Sheet Molding Compound (SMC) or in prepreg forms, and can be processed by press or Autoclave molding. For successful processing of high quality composite laminates, an effective design of the cure cycle is the foremost important factor. The design of the cure cycle is often influenced by material properties such as: resin flow, chemorheology, volatile escaping mechanism, heat transfer between the process environment and the specimen, fiber deformation, and the kinetics of the chemical reaction of resin, etc. This report presents an effective cure cycle design by means of the optimization technique. The design is implemented by Computer Aided Design (CAD) methodology.

This work has been accepted for publication in the *International Polymer Processing*, Volume IV, 1990. One previous publication which deals with the same subject matter is: *Autoclave Processing For Composite Laminate : An Analysis Of Resin Flows And Fiber Compactions For Thin Laminates*, NASA Contractor Report CR-178011, November, 1985.

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ON PROCESSING DEVELOPMENT FOR FABRICATION OF FIBER REINFORCED COMPOSITE - II*

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ABSTRACT

Fiber-reinforced composite laminates are used in many aerospace and automobile applications. The magnitudes and durations of the cure temperature and the cure pressure applied during the curing process have significant consequences for the performance of the finished product. The objective of this study is to exploit the potential of applying the optimization technique to the cure cycle design. Using the compression molding of a filled polyester Sheet Molding Compound (SMC) as an example, a unified Computer Aided Design (CAD) methodology, consisting of three uncoupled modules, (i.e., Optimization, Analysis and Sensitivity Calculations), is developed to systematically generate optimal cure cycle designs. Various optimization formulations for the cure cycle design are investigated. The uniformities in the distributions of the temperature and the degree of cure within the specimen cured under the optimal cycles are compared with those resulting from conventional isothermal processing conditions with pre-warmed platens. Recommendations with regards to further research in the computerization of the cure cycle design are also addressed.

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INTRODUCTION

High performance polymeric composites are used increasingly in the aerospace and automobile industries. Such materials are typically composed of long or chopped fibers embedded in a thermosetting resin matrix. The composite parts and structures are often manufactured by curing the prepreg or unmolded material. The cure process is accomplished by exposing the material to elevated (cure) temperatures and (cure) pressures for a predetermined length of time. The cure temperature provides the heat required for initiating and maintaining the chemical reactions in the resin during the curing process. Once the reactive matrix resin is heated, polymerization reactions start to occur. The heat comes from two sources, i.e., from the exotherm generated from the reactions and by conduction and convection from the surrounding environment. At the certain stage during the processing, the cure pressure is applied to squeeze out the excess resin inside the laminate. Consequently, the individual plies are consolidated and the vapor bubbles are compressed.

The magnitudes and durations of the cure temperature and the cure pressure applied during the curing process have significant consequences on the performance of the finished product. Thus, it is not a trivial task to properly design a cure cycle (temperature and pressure profiles). The material should be cured uniformly and completely with the lowest void content; the temperature inside the laminate must not exceed some maximum value; and the cure process should be completed within the shortest amount of time. In the past, most cure cycle designs for newly developed composite systems have been based on the technique of laboratory trial and error. Such an approach has long been recognized as costly and inefficient. Several simulation models have been developed recently for curing various thermoplastic [1] and thermosetting [2 - 4] matrix composites. These developments represent a significant advancement in computerizing the cure cycle design. The next logical step is then to develop a methodology which could be used to search for the optimal cure cycle for a given fiber/resin composite system. This paper reports and discusses the initial results from such an investigation which uses a unified Computer-Aided

Design (CAD) method for the cure cycle design, that incorporates an optimal design technique with an analytical model for a composite curing process.

The curing process of interest is the compression molding of a polyester sheet molding compound (SMC). The finite element method is employed to convert the initial-boundary value problem into a set of first order differential equations which are solved simultaneously by the DE program [5]. The equations for thermal design sensitivities are then derived by using the direct differentiation method and are solved by the DE program. Finally, a recursive quadratic programming algorithm with an active set strategy, called a linearization method [6], is used to design the optimal cure cycle which is subjected to the given design performance requirements. The difficulties of casting the cure cycle design process into a proper mathematical form are recognized, and various optimal design problems are formulated to address these aspects. The optimal solutions of these formulations are compared and discussed, and the processing and material parameters which play major roles in the cure cycle design for a given composite system are identified. Recommendations with regard to the computerizing of the cure cycle design are also addressed.

THERMAL ANALYSIS

Problem Statement

The formulation of a complete analytical model for the curing process in the manufacture of fiber/resin composite laminates is rather complicated [3]. Such an analytical model should be able to address:

- a) the heat transmission between the surrounding environment and the specimen,
- b) the heat generated by the chemical reaction of the resin,
- c) the temperature distribution within the laminate,
- d) the phase changes of the resin during cure as commonly characterized by the chemoviscosity and the degree of cure, and
- e) the resin flow problems associated with the consolidation process among the prepreg plies and volatile removal from the specimen.

The major parameters which play important roles in modeling the cure process may be identified as the temperature, the degree of cure of the resin, the resin viscosity, the resin flow properties, the thickness of the specimen and the material properties, such as density, heat conductivity, etc. Most parameters are varied continuously with respect to the time and spatial location.

To focus on the optimization aspects of the present study, several assumptions which simplify the formulation of the curing process are introduced as follows:

- 1) the temperature and the degree of cure of the specimen are uniformly distributed in a plane parallel to the tool plates. As a result, the parameters of the problem are functions of the time and the position along the thickness of the specimen only.

- 2) the resin content in the material is assumed to be low so that there is only a small amount of excess resin. Consequently, the resin flow model as well as the pressure cycle are not considered.
- 3) the material properties which depend on the fiber-resin ratios are assumed to remain constant during the entire cure cycle,
- 4) the volatiles escaping mechanism is not considered, and
- 5) there is no deviation between the environment (tool) temperature and the temperature on the surfaces of the material adjacent to the tool plates. The temperature distribution and the degree of cure are the only two state variables which are governed by diffusion-reaction system equations.

After these simplifications, a CAD scheme for the cure cycle design is developed in the following sections for the curing process of the compression molding for a filled polyester SMC [2]. Similar procedures are also applicable to the curing of continuous fiber reinforced composite lamination process.

Diffusion-Reaction System Equations

The SMC curing process is modeled by a diffusion-reaction mechanism. The diffusion-reaction system equations consist of a one-dimensional unsteady heat-conduction equation with its boundary and initial conditions, and a kinetics model of the curing reaction for the reactive resin system. A computational algorithm, based on the Finite Element Method and a numerical integrator, is developed to analyze the thermo-chemical reaction.

The temperature distribution $T(z,t)$ and the degree of cure $\alpha(z,t)$ of the resin inside the composite laminate depend on the rate at which heat is transmitted from the environment. The temperature distribution inside the composite laminate can be calculated by using the law of conservation of energy together with an appropriate expression for the cure kinetics. By neglecting energy transferred by convection, the energy equation may be expressed as

$$\rho c \dot{T} = kT'' + \rho H_r \dot{\alpha} \quad (1)$$

with the boundary conditions,

$$T'(0,t) = 0, \quad 0 \leq t \leq \tau, \quad (2.a)$$

$$T(h,t) = T_c(t) \quad 0 \leq t \leq \tau, \quad (2.b)$$

and the initial condition,

$$T(z,0) = T_0(z), \quad 0 \leq z \leq h \quad (3)$$

where ρ and c are the density and the specific heat of the composite material, respectively, k is the thermal conductivity in the direction perpendicular to the plane of composite material, $2h$ is the total thickness, $T_c(t)$ is the cure cycle in $^{\circ}\text{K}$, τ is the time in seconds needed for the completion of one cure cycle, and H_r is the total or ultimate heat of reaction. Moreover, the dot (\cdot) on the top of a symbol indicates the time, t , derivative and the superscripts, ($'$) and ($''$), denote the first and second spatial, z , derivative respectively. According to the assumptions discussed previously, all coefficients in Eqs. (1) - (3) are treated as constants. Note that the cure temperature $T_c(t)$ appears in Eq. (2) as the boundary conditions; and the last term in Eq. (1), $\rho H_r \dot{\alpha}$, is the rate of heat generated by the chemical reaction as characterized by the degree of cure α .

The degree of cure, α , is defined as the fraction of heat, $H(t)$, released up to time, t , for the resin system under cure:

$$\alpha = H(t)/H_r \quad (4)$$

Both $H(t)$ and H_r in Eq. (4) can be measured experimentally by Differential Scanning Calorimetry (DSC). For an uncured material, α approaches zero, and for a completely cured material, α approaches one. The reaction rate, $\dot{\alpha}$, depends strongly on the curing temperature. As an example, the cure rate equation of a stepwise isothermal curing process is used for a polyester SMC [2] as follows:

$$\begin{aligned}\dot{\alpha} &= f(\alpha, T) \\ &= (K_1 + K_2 \alpha^m)(1 - \alpha)^n \\ &= (a_1 e^{-d_1/RT} + a_2 e^{-d_2/RT} \alpha^m)(1 - \alpha)^n\end{aligned}\quad (5)$$

where a_1 , a_2 , d_1 , d_2 , m and n are constants, R is the universal gas constant, and K_1 and K_2 are exponential functions of the temperature. Some observations of interest are noted: (1) The state equations of the cure process are coupled with two state variables, namely, the temperature distribution $T(z,t)$ and the degree of cure $\alpha(z,t)$, and (2) The non-homogeneous boundary value, $T_c(t)$, is to be considered as the design variable in the design optimization problem. Using the following replacement for the temperature $T(z,t)$:

$$T(z,t) = \bar{T}(z,t) + T_c(t), \quad (6)$$

the heat-conduction problem stated in Eqs. (1) - (3) can be simplified as an equation of $\bar{T}(z,t)$

$$\rho c \bar{T} = k \bar{T}'' - \rho c \dot{T}_c + \rho H_r f(\alpha, \bar{T}, T_c), \quad (7.a)$$

with the homogeneous boundary conditions,

$$\bar{T}'(0,t) = 0, \quad 0 \leq t \leq \tau, \quad (7.b)$$

$$\bar{T}(h,t) = 0, \quad 0 \leq t \leq \tau, \quad (7.c)$$

and the initial condition,

$$\bar{T}(z,0) = T_o(z) - T_c(0), \quad 0 \leq z \leq h \quad (7.d)$$

where the function f is defined in Eq. (5). From here on, \bar{T} is abbreviated as T for further simplification. Because the initial temperature $T_o(z)$ is the same as the initial cure temperature for most applications, Eqs. (7) may have a homogeneous initial condition as well.

The finite element discretization is introduced herein to convert the initial-boundary value problem, Eqs. (5) and (7), into a set of first order matrix equations. Quadratic and linear polynomials are used to interpolate the states of temperature and the degree of cure, respectively. Notation T_{2i-1} , T_{2i} and α_i denote the temperature, the temperature gradient and the degree of cure, respectively, at node i :

$$\rho c[C] \{\dot{T}\} + k[K] \{T\} + \rho c\{P\}\dot{T}_c = \{Q(\alpha, T, T_c)\}, \quad (8.a)$$

$$[M] \{\dot{\alpha}\} = \{R(\alpha, T, T_c)\}, \quad (8.b)$$

where the unknowns are the vector of temperature $\{T\} = \{T_1, T_2, \dots, T_{2n}\}^T$ and the vector of the degree of cure, $\{\alpha\} = \{\alpha_1, \alpha_2, \dots, \alpha_n\}^T$ for $n = NE + 1$ as the total number of nodes. The matrices $[C]$, $[K]$ and $[M]$ result from the finite element discretization of the terms \bar{T} and \bar{T}'' in Eq. (7.a) and the term $\dot{\alpha}$ in Eq. (5). Moreover, the vector $\{P\}$ is associated with the term \dot{T}_c in Eq. (7.a) and the vectors $\{Q\}$ and $\{R\}$ appearing in Eq. (8) are the discretized forms of the terms pertaining to $f(\alpha, T)$ in Eqs. (5) and (7.a). Note that $T_1(t)$ and $T_{2n}(t)$ are always zeros in the analysis according to the boundary conditions of $T(h,t) = 0$ and $T'(0,t) = 0$, respectively. The initial conditions for each of the components in $\{T\}$ and $\{\alpha\}$ are

$$T_{2i-1}(0) = T_o - T_c(0),$$

and $T_{2i}(0) = \alpha_i(0) = 0$ where i is the nodal number. Moreover, T_o represents the temperature of the composite at the onset of the cure process and this is usually uniformly distributed. The detailed derivation of the matrix equations, Eqs. (8), is given in Ref. [7]. The above equations, coupled with each other through the non-linear terms on the right side of Eqs. (8), can be solved simultaneously by using the computer program DE [5].

The DE program is one of predictor-corrector integration algorithms using the Adams family of formulas. The truncation error is controlled by varying both the step size and the order of the polynomial approximation. The DE program is quite easy to use and has the capability of managing moderately stiff differential equations which happen frequently in the chemical kinetics problems. To maintain a unified accuracy in the analysis, the computation of two state variables, namely, the temperature and the degree of cure, are subjected to the same error tolerance in this study. Numerical solutions of several examples designed to verify Eqs. (8) with existing analytical solutions can be found in Ref. [7].

THERMAL DESIGN SENSITIVITY ANALYSIS

The derivatives of functionals of responses with respect to the design variables are often referred to as design sensitivity derivatives. Most general optimization algorithms require such derivatives which can be used to approximate constraints and to choose a search direction to obtain a set of improved design variables. It is then necessary to have a reliable means to calculate the design sensitivity derivatives.

To focus on the study of thermal design sensitivity analysis in this section, the cure uniformity is simply represented by the least-square integral of the deviation between the pointwise temperature and the averaged temperature as

$$\phi_o = \int_0^{\tau} \left[\int_0^h T^2 dz - \left(\int_0^h T dz \right)^2 / h \right] dt. \quad (9)$$

The above functional defines a global sense of temperature uniformity across the thickness of the composite during the cure process. It is expected that the temperature uniformity within the curing specimen will translate into the uniformity of the curing reaction. Other mathematical expressions of the cure uniformity will be discussed in the following section of optimal cure cycle design. On the other hand, the following pointwise inequality may be used to force the cure reaction to reach certain predetermined value, α_f , at the end of cure cycle:

$$\alpha(z, \tau) \leq \alpha_f, \quad 0 \leq z \leq h \quad (10)$$

where τ is the total time required to complete one cure cycle. The numerical techniques to calculate the design derivatives of the functional ϕ_o and the state variable α , $\frac{d\phi_o}{db}$ and $\frac{d\alpha}{db}$, will be addressed hereafter. Note that the design variable b is a parameter associated with the cure cycle (temperature profile).

In general, there are four ways to calculate the thermal design derivatives, namely, the finite difference method, the Green's function approach, the direct differentiation method (the behavior space approach), and the adjoint variable method (the dummy load method). The last two methods are often mentioned in the literature [8 - 10]. Both methods lead to a set of linear equations that have a similar structure to the original system. However, based on the numerical study [11], it has been concluded that the direct differentiation method is superior to the adjoint variable technique in terms of accuracy and physical interpretation of results. Thus, the thermal design sensitivity is calculated by using the direct differentiation method in this study.

The direct differentiation method is an approach that takes derivatives of differential equations with respect to a single design variable directly. For a given functional, Eq. (9), and the governing equations of the diffusion-reaction system, Eqs. (5) and (7), the direct differentiation results in the following equations in terms of design derivatives $\frac{dT}{db}$ and $\frac{d\alpha}{db}$ as:

$$\phi_{\alpha,b} = \int_0^{\tau} \left[\int_0^h 2T \frac{dT}{db} dz - \frac{2}{h} \left(\int_0^h T dz \right) \left(\int_0^h \frac{dT}{db} dz \right) \right] dt, \quad (11)$$

$$\begin{aligned} \rho c \frac{dT}{db} = & k \frac{dT''}{db} - \rho c \frac{dT_c}{db} + \rho H_r \frac{\partial f}{\partial \alpha} \frac{d\alpha}{db} + \rho H_r \frac{\partial f}{\partial T} \frac{dT}{db} \\ & + \rho H_r \frac{\partial f}{\partial T_c} \frac{dT_c}{db}, \end{aligned} \quad (12)$$

and,

$$\frac{d\alpha}{db} = \frac{\partial f}{\partial \alpha} \frac{d\alpha}{db} + \frac{\partial f}{\partial T} \frac{dT}{db} + \frac{\partial f}{\partial T_c} \frac{dT_c}{db}. \quad (13)$$

It is assumed that $T(z,b,t)$ and $\alpha(z,b,t)$ have enough regularity in the time-spatial domain and in the design space. Thus, the order of the differentiation is interchangeable.

Based on the same finite element discretization as used in solving the original diffusion-reaction system discussed previously, Eqs. (11) - (13) can be converted into a set of matrix equations

$$\phi_{\alpha,b} = \int_0^{\tau} \left(2\{T\}^T [C] \{T_b\} - \frac{2}{h} \{T\}^T \{P\} \{P\}^T \{T_b\} \right) dt, \quad (14)$$

$$\rho c [C] \left\{ \dot{T}_b \right\} = -k [K] \{T_b\} - \rho c \{P\} \frac{dT_c}{db} + \{Q\}, \quad (15.a)$$

and

$$[M] \left\{ \dot{a}_b \right\} = \{R\}. \quad (15.b)$$

The construction of the above equations is discussed in Ref. [5].

Note that the coefficient matrices of $\left\{ \dot{T}_b \right\}$ and $\left\{ \dot{\alpha}_b \right\}$ in Eqs. (15) are identical to those of $\{T\}$ and $\{\alpha\}$ defined in Eqs. (8). Therefore, the same numerical scheme and numerical tolerance can be applied to solve both Eqs. (8) and (15) simultaneously for state variables ($\{T\}$ and $\{\alpha\}$) and design derivatives ($\{T_b\}$ and $\{\alpha_b\}$). In this way, the state variables and the design derivatives achieve the same numerical accuracy, though an additional set of equations such as Eqs. (15) is needed in this approach for each design variable b .

Once $\{T\}$ and $\{T_b\}$ are available, the design derivative given in Eq. (14) can be easily obtained by the numerical integration. Another suggestion is to rewrite the integral form of Eq. (14) as a differential equation of ϕ given as

$$\dot{\phi}_{\alpha,b} = 2\{T\}^T [C] \{T_b\} - \frac{2}{h} \{T\}^T \{P\} \{P\}^T \{T_b\} \quad (16)$$

The above equation of $\phi_{\alpha,b}$ can then be solved simultaneously with equations of ($\{T\}$, $\{\alpha\}$) and ($\{T_b\}$ and $\{\alpha_b\}$). In this way, one extra design derivative $\phi_{\alpha,b}$ for each design variable is

introduced in the design sensitivity analysis. However, the accuracy of $\phi_{o,b}$ is secured. Equation (16) is used in the next section to compute $\phi_{o,b}$.

OPTIMAL CURE CYCLE DESIGN

The optimal design of cure cycles is studied here to find the optimal cure cycle such that the specimen is cured as uniformly as possible, and that a certain preset degree of cure is achieved at the end of the curing process.

Numerical results presented in this section have been obtained by a recursive quadratic programming algorithm with an active set strategy, called a linearization method [6]. The linearization method, with its active set strategy, minimizes the number of constraints which must be considered in each design iteration. The detailed derivation of this algorithm can be found in Refs. [6,12]. The concept of this algorithm is that, rather than directly solving the optimal criteria, a small perturbation for each design variable is determined in each iteration to reduce the objective function and correct the violation. Note that, in this approach, the reduction of the objective function and the correction of the constraint violations are approximated by the design gradients. In the linearization method it has been proved [13] that a local optimal solution is found when the l^2 norm of the perturbation of design variables approaches zero. The complete flow chart of optimal cure cycle design is listed in Fig. 1. As shown in the figure, the proposed CAD method consists of three uncoupled modules; namely, Optimization, Analysis, and Sensitivity Calculation. Once the analysis capability of any cure process is established, the Optimization and Sensitivity Calculation modules can be added onto it to constitute a unified CAD methodology which can generate optimal cure cycles under various constraints systematically.

Problem Statement

As mentioned, optimal design techniques have been successfully applied to various transient problems. In general, an optimal design problem consists of the design variables, the objective functions, the constraint functions, and the state equations which describe the physical model of interest.

In our study, the process of interest is the compression molding of a filled polyester resin reinforced by chopped glass fibers SMC [2]. The state equations are limited to a heat conduction equation, Eq. (1), and an empirical equation which addresses the chemical-kinetics reaction of resin, Eq. (5). Since this study excludes the resin flow and the effects of the pressure cycle, the selection of the cure cycle is then limited to the following considerations:

- a) The maximum temperature inside the composite laminate at any instant during the cure cycle can not exceed 500 °K,
- b) At the end of the curing cycle, the degree of cure of the reactive resin system is required to reach at least 0.85, and
- c) The specimen should be cured uniformly at any instant during the entire cycle.

The first two requirements may be formulated as constraints

$$\phi_1 : T(t) \leq 500 \text{ °K}, \quad 0 \leq t \leq \tau, \quad (17)$$

$$\phi_2 : \alpha(\tau) \geq 0.85, \quad (18)$$

Here the curing period, τ , is predetermined by the user, and is not considered as a design variable in the current design formulation. However, it is not difficult to include τ as an additional design variable in the proposed formulation. The last requirement can be met by formulating it as the objective function of the optimal design. Since the degree of cure is a function of temperature, a uniform distribution of temperature across the thickness will tend to insure the uniformity of the cure within the specimen. The objective of the optimal design may then be set to achieve uniformity of the temperature across the section of the material during the cure process. The objective function ϕ_0 is thus defined as the greatest value of the standard derivation of temperature distribution which happens during the cure process

$$\phi_0(T_c, t) = \text{Maximum}_{0 \leq t \leq \tau} \left[\int_0^h T^2 dz - \left(\int_0^h T dz \right)^2 / h \right]. \quad (19)$$

It follows that the minimum of the objective function, $\phi_0(T_c, t) = 0$, corresponds to a uniform temperature distribution inside the composite at any time during the cure process. To sum up, the optimal cure cycle design is defined as follows:

The optimal cure cycle (temperature profile) is designed, subject to the limitations regarding the maximum temperature and the state of cure, so as to achieve a uniform temperature distribution along the cross section of the material at any time during the entire cure cycle.

The mathematical formulation of the stated optimal design problem is a minimax problem given as

$$\text{Minimum}_{T_c(t)} \text{Maximum}_{0 \leq t \leq \tau} \left[\int_0^h T^2 dz - \left(\int_0^h T dz \right)^2 / h \right]$$

subject to the constraints stated in Eqs. (17) - (18) where the temperature $T(t)$ and the degree of cure $\alpha(t)$ are the solutions of state equations.

It is known that the objective function of a minimax problem is discontinuous in the design space [14]. To avoid computational difficulty, one may modify the optimal design problem to a standard form by introducing an extra design variable b_0 and an additional constraint ϕ_3 as follows:

$$\text{Minimize } \phi_0 = b_0 \\ T_c(t), b_0$$

subject to

$$\phi_1 : T(t) \leq 500 \text{ }^\circ\text{K}, \quad 0 \leq t \leq \tau, \quad (20)$$

$$\phi_2 : \alpha(\tau) \geq 0.85, \quad (21)$$

$$\phi_3 : \left[\int_0^h T^2 dz - \left(\int_0^h T dz \right)^2 / h \right] \leq b_0, \quad 0 \leq t \leq \tau. \quad (22)$$

The design function $T_c(t)$ can now be parameterized by a linear combination of design parameters and given functions. In this presentation, the design function is simply represented by a combination of linear polynomials and sinusoids

$$T_c(t) = T_0 + b_1 t + b_2 \sin(\pi t/\tau) + b_3 \sin(2\pi t/\tau) + b_4 \sin(3\pi t/\tau), \quad (23)$$

where T_0 is the room temperature and $b_1, b_2, b_3,$ and b_4 are to be determined by the optimal design algorithm. As a result, the design space becomes finite dimensional, and the design sensitivities (or gradients) of temperature $T(t)$ and state of cure $\alpha(t)$ are taken as derivatives with respect to design parameters $b_1, b_2, b_3,$ and b_4 . For example, the design derivatives of constraint functions, Eqs. (21) and (22), can be derived as

$$\frac{\partial \phi_2}{\partial b_i} = \left(\frac{\partial \alpha(\tau)}{\partial b_i} \right) / 0.85,$$

and

$$\frac{\partial \phi_3}{\partial b_i} = \frac{2}{b_0} \left[\int_0^h T \frac{\partial T}{\partial b_i} dz - \left(\int_0^h T dz \right) \left(\int_0^h \frac{\partial T}{\partial b_i} dz \right) / h \right].$$

for $i=1-4$. It is obvious that the design sensitivities of $\frac{\partial T}{\partial b_i}$ and $\frac{\partial \alpha}{\partial b_i}$ are needed for the calculation of the above function gradients. Note that the pointwise constraints, ϕ_1 and ϕ_3 , are imposed at every time grid point in this study. With the information of thermal sensitivities, the optimal cure cycle $T_c(t)$ of the above problem can be found numerically by using a gradient-based mathematical

programming technique. As discussed in the last section, the direct differentiation method is employed hereafter to compute the required thermal design derivatives.

Numerical Examples

To show the applicability of the proposed CAD scheme for the optimal cure cycle design, four examples associated with various problem formulations are discussed and presented in this section.

Example 1.

The objective function ϕ_0 defined by Eq. (19) is minimized to find an optimal cure cycle design for processing a typical chopped glass fiber reinforced polyester SMC with a thickness of 10 mm by compression molding. The total processing time is limited to 100 seconds. The maximum temperature allowed inside the SMC is 500 °K; and the degree of cure, α , is required to reach at least 0.85 at the end of the cure cycle.

The physical and kinetics properties of the SMC material are given in Table 1 [2]. The optimization algorithm requires 22 iterations to obtain the optimal solution for this case. Numerical results for the optimal design are listed in Table 2. Fig. 2 shows the cure cycles computed at iterations 1, 11, and 22, respectively. It should be noted that the cure cycles designed in this example are always made to start from room temperature which is different from the common practice with pre-warmed press platens employed in the compression molding process. The initial cure cycle (iteration 1) has two peaks and a valley at around 20, 85 and 60 sec., respectively. During the iterations toward the optimal solution, the initial heating rates are seen to decrease gradually; and a single peak near 70 sec is obtained eventually for the iteration 22 solution. The values of the objective function are shown in Fig. 3. The optimal cure cycle indeed delivers a more uniformly distributed temperature. The optimal solution has reduced the maximal value of temperature deviation from 2,700 to 378 units.

Temperature distributions across the thickness of the SMC at selected instants of 20, 60 and 100 sec during the cure process are compared for iterations 1, 11 and 22, shown in Fig. 4(a), (b) and (c), respectively. As noted previously in Fig. 2, the lower initial heating rate helps the optimal cure cycle (iteration 22) achieve a more uniform temperature distribution at the early 20 sec. into the cure process. At the 60 sec., the cure cycle of the eleventh iteration yields a better distribution profile as seen in Fig. 4(b), which is due to the negative heating rates (cooling) between 30 to 50 sec. as seen in Fig. 2. On the other hand, the higher initial heating rate provided by the cure cycle of iteration 1 (Fig 2) has strongly triggered the kinetics reaction in the first 20 sec. into the cure process, and consequently results in a highly non-uniform temperature distribution between 30 to 50 sec., in spite of the provision of subsequent steep cooling rate within the same time interval in the cure cycle as shown in Fig. 2. In other words, within the processing time, the heat exchanges offered by the steep cooling rate of the cure cycle did not effectively offset the extra heat generated by the chemical reactions between this time period.

The distributions of the degree of cure α across the SMC thickness are shown in Figs. 5 and 6 for the cure cycles of iteration 1 and 22. In both cases, the cure of the resin starts from the surface regime and extends into the core of the SMC. The cure cycle of iteration 1 requires the shortest time (60 sec.) to complete the cure reaction of the material with $\alpha > 0.85$. The lower cure temperatures provided by the optimal cure cycle (iteration 22) at the initial 40 seconds into the cure process can hardly trigger the chemo-kinetics reactions of the resin matrix as seen from Fig. 6. However, the rate of the cure speeds up in the next 40 seconds and eventually the degree of cure exceeds the 0.85 limit throughout the SMC material at the end of the cure cycle.

Example 2.

The effect of the length of cure cycle is investigated here. Example 2 is formulated to be identical to the problem stated in Example 1, except that the total processing time, τ , is extended to 150 seconds.

The optimal cure cycle (iteration 22, Fig. 2) of Example 1 is used as the initial trial cure cycle (iteration 1, Fig. 7) studied here. The algorithm needs another 25 iterations to reach the optimal solution for this problem. Table 3 lists the numerical results for the optimal design. The starting (iteration 1) and optimal (iteration 25) cure cycles are compared in Fig. 7. It is noted that the cure cycle of Example 2 possesses even slower initial heating rates, and the cooling period is no longer needed over the entire cure cycle. The value of the objective functions are also compared in Fig. 8, and a drop of maximum temperature deviation from 250 to 110 units is noted. The temperature distributions across the SMC thickness at selected instants during the cure process for the two optimal cure cycles of Examples 1 and 2 are compared in Figure 9. The distribution of the degree of cure for the optimal cure cycle of this example is shown in Fig. 10. The cure behavior basically shows the same characteristics as the optimal solution of Example 1. The extension of the total curing time, therefore, only enables an optimally designed cure cycle to achieve an insignificant improvement in the temperature uniformity.

Example 3.

Example 3 is identical to the problem stated in Example 1, except that a new objective function, Eq. (24) (see below), which defines the maximum deviation in the cure state, is minimized; and the total processing time is extended to 150 seconds.

It was rationalized in the past two examples that a uniform state of cure within the specimen could be achieved when a perfectly uniform temperature distribution at any time of the cure cycle was maintained. However, numerical results showed that the uniform degree of cure was not realized even though the temperature uniformity (as indicated by the objective functions ϕ_0) had been improved greatly. This could be due to the fact that the degree of cure is quite sensitive to the temperature non-uniformity. In this example, the objective function Eq. (19) and the constraint Eq. (22) are therefore replaced by the following forms, respectively;

$$\phi_0 : \text{Maximum} \left[\int_0^h \alpha^2 dz - \left(\int_0^h \alpha dz \right)^2 / h \right], \quad (24)$$

$$\phi_3 : \left[\int_0^h \alpha^2 dz - \left(\int_0^h \alpha dz \right)^2 / h \right] \leq b, \quad 0 \leq t \leq \tau. \quad (25)$$

The optimal cure cycle of Example 2 is used as the initial trial solution in this case. Fifty-five more iterations are required in order for the algorithm to converge to a optimal solution. The optimal cure cycle shown in Fig. 11 has the characteristic of a ramp-hold-ramp feature. Numerical results are listed in Table 4. The values of the objective functions of Eq. (24) for the starting and the (final) optimal cure cycles are compared in Fig. 12. The optimal solution reduces the maximum deviation in the degree of cure, as expected. However, the differences are, again, not significant. The distribution of the degree of cure, shown in Fig. 13, for the optimal cure cycle of Example 3, is similar to that of Example 2 (Fig. 10). The temperature distribution under the optimal cure cycle is shown in Fig. 14. These results indicate that the change of the objective function from Eq. (19) to Eq. (24) has only minor effects on the uniformity in the distribution of the degree of cure, even though the profiles of the optimal cure cycle designs are quite different.

Example 4.

Example 4 is identical to the problem stated in Example 1, except that a new objective function, Eq. (26), which defines the maximum deviation in the state of cure is minimized; the total processing time allowed is 150 seconds and the initial cure temperature is no longer restricted to room temperature, and is regarded as an additional design variable.

In this example, the objective function Eq. (19) and the constraint Eq (22) are replaced by the following forms, respectively;

$$\phi_0 : \text{Maximum } (\alpha_0 - \alpha_c), \quad (26)$$

$$0 \leq t \leq \tau$$

$$\phi_3 : (\alpha_0 - \alpha_c)^2 \leq b, \quad 0 \leq t \leq \tau. \quad (27)$$

Here the objective function becomes the difference of the degree of cure between the outer surface, α_0 , and the center plane, α_c . The optimal cure cycle of Example 3 (iteration 55, Fig. 11) is used as the initial trial solution. The optimal design algorithm does not really achieve convergence. However, after the forty-sixth iteration, the changes of the design variables become relatively small, and all the constraints are satisfied. The feasible design of the forty-sixth iteration is then regarded as the final design. Numerical results of this example are listed in Table 5. This example has five design variables in total that introduce two hundred and eighty-two equations in the form of Eqs. (8) or (15) to be solved. It took the algorithm around 196,800 CPU seconds (54.6 CPU hours) on the IBM 4381 computer to obtain the final solution.

The cure cycles and the values of the objective function of Eq. (26) for iterations 1 (corresponding to the optimal solution of Example 3), iteration 46, and that of reference [2] are shown in Figs. 15 and 16, respectively. Note that the cure cycle given by reference [2] is a constant temperature profile with $T_c = 423$ °K. The cure cycle obtained from the forty-sixth iteration has the characteristic of a ramp-hold-ramp feature similar to that of Example 3. It is recognized that the higher onset cure temperature of the optimal cure cycle benefits the cure cycle design by initiating the chemical reaction as quickly as possible so as to achieve a better uniformity in the state of cure distribution (see Fig. 16). Similar to the previous case, the optimal solution of this example did improve the uniformity of the state of cure, although such improvement is not very significant. The final results of the temperature distribution and the degree of cure distribution at selected instants are shown in Figs. 17 and 18, respectively.

DISCUSSION

After studying four different optimization design formulations, the uniformity of cure was not seen to improve significantly. This result is attributed to the physical aspects of the problem. In the curing of thermosetting resin matrix, there are two heat sources involved, namely, the heat flux from the processing tool platens and the heat generated by the kinetics reactions of the material. The conduction is the dominant mechanism for the heat transfer in this case. It can be seen from Eq. (5) that the objective of current cure cycle design, namely, to have a uniform cure inside the laminate at all times during the entire cure cycle, can be achieved when a uniform temperature profile exists across the laminate. However, a uniform temperature profile without gradient will not yield any heat conduction. Thus the heat generated by the kinetics reactions inside the laminate can not be transferred outward effectively. As a result, the current optimal design algorithm tends to raise the temperature of the laminate, through the heat conduction from the tool platens, at a very slow pace. Such a curing process will require a long cure time τ . For practical considerations, however, only finite lengths of cure time were investigated in this study.

It is interesting to note that the resultant optimal cure cycle in each of the four examples studied above can be broadly divided into three stages as follows:

STAGE 1 This is the initial stage of the cure. The laminate was warmed up uniformly with no apparent reactions triggered. The cure cycle was inclined to raise temperature to expedite the curing process and to meet the design constraints (the cure time τ , for example). The degree of cure at the surface, α_0 , is greater than that at the center, α_c .

STAGE 2 The influx of heat triggered the reactions inside the laminate. However, the small temperature gradient across the laminate prevented effective outward heat conduction, and further kinetics reactions were triggered such that, in a short period of time, large extent of cure was reached. This process is evident by noting a sharp decrease of $(\alpha_0 - \alpha_c)$. For the optimal design of Example 4, this period started at 120 and ended at 140 seconds (Figure 16).

STAGE 3 This is the latter stage of cure. The entire laminate had reached the desired degree of cure. The cure cycle was inclined to lower the temperature to facilitate more effective heat conduction outward from the laminate.

It is evident that the optimal cure design algorithm tends to stretch the period of STAGE 1 as long as possible for the reasons discussed above.

In summary, one may conclude that (i) a uniform cure across the laminate at all time during the cure cycle can only be achieved with an infinite length of cure time. Obviously this is not a practical cure cycle, a more realistic design objective needs to be identified; and (ii) a more uniform temperature profile across the laminate could be achieved if more effective heat transfer mechanisms were adopted in the process. The technique of employing internal heating sources [15] may provide extra flexibilities for the cure cycle design in this aspect.

Finally, it is recognized that SMC molding process has a relatively short cycle time and molding temperature can not be varied easily during curing. The selection of SMC as a preliminary case study for the CAD methodology reported here is simply because of the consideration of the amount of CPU times required to reach convergence of the algorithm. Nevertheless, the optimization technique presented here can be easily applied to the curing process with long cycle time (i.e. Autoclave curing) without any difficulties.

CONCLUSIONS AND REMARKS

A unified CAD methodology is introduced in this paper to design the optimal cure cycle in compression molding operation for a fiber reinforced composite laminate SMC. The process for curing the fiber reinforced composite laminate is simplified as a diffusion-reaction system and a kinetics model is used for the reactive resin system. The proposed CAD methodology consists of three uncoupled modules; namely, Optimization, Analysis, and Sensitivity Calculations. Once the analytical capability of any curing process is established, the Optimization and Sensitivity Calculation modules can be added onto it to constitute a unified CAD methodology which can systematically generate the optimal cure cycle designs subjected to various constraints .

Four optimal design formulations for the optimal cure cycle design for the compression molding of a chopped fiber SMC were studied. The numerical examples showed that the proposed CAD approach was valid in different situations. It did improve the degree of cure uniformity within the laminate. This study also indicated that the total processing time, the initial heating rate and the initial cure temperature have significant effects on the cure uniformity. Enlarging the design space by increasing the total processing time and adding more design variables can further improve the cure cycle design. The long CPU time required in the present optimization formulation was a concern. To shorten the CPU time one may use other numerical algorithms, such as an unconditional by stable one, which can increase the time step. Other kinds of optimization algorithms, such as the I-DESIGN [16] which does not need the line search to accelerate the optimization process, should also be considered.

The proposed CAD scheme can be extended to design an optimal cure cycle for the fabrication of long fiber reinforced composite laminates (i.e., autoclave processing). It is noted that, to optimally design a realistic cure process for a resin-fiber composite, some modifications in Analysis module over this simplified SMC model are required. For example, the resin flow properties, the material inhomogeneities such as voids, fiber segregation, local fiber orientation, etc., and the heat convection through the surrounding environment should be considered in the

Analysis module. The total processing time should be regarded as a design variable to enlarge the design space. Moreover, some experiments are necessary to verify the results of the optimal cure cycle designed.

In summary, it is clear that the CAD methodology is a valid technique to enhance the capability of designing the optimal cure cycles for the curing process of fiber reinforced composite laminates. However, in order to improve its applicability, further studies in modeling, computational efficiency and optimization formulation are needed to explore.

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Table 1. Material Properties of A Filled Polyester SMC [2]

ρ	=	1900 kg m ⁻³
c	=	1.0 J g ⁻¹ k ⁻¹
k	=	0.53 W m ⁻¹ k ⁻¹
a_1	=	4.9 x 10 ¹⁴ sec ⁻¹
a_2	=	6.2 x 10 ⁵ sec ⁻¹
d_1	=	140 kJ mole ⁻¹
d_2	=	51 kJ mole ⁻¹
m	=	1.3
n	=	2.7
R	=	8.83138 J mole ⁻¹ K ⁻¹
H_r	=	84 J g ⁻¹

Table 2. Convergence History of The Optimum Design (Example 1)

Parameter	Initial Value	Final Value
Design Variables:		
b ₁	100.00	115.74
b ₂	100.00	100.52
b ₃	100.00	-5.16
b ₄	100.00	6.83
Cost Function	2700.00	378.33
l² norm	33.62	0.0054
Computer Time (CPU sec. CDC 855)	---	108,140.00
No. of Iterations	---	22

Table 3. Convergence History of The Optimum Design (Example 2)

Parameter	Initial Value	Final Value
Design Variables:		
b ₁	115.74	162.32
b ₂	100.52	30.98
b ₃	-5.61	-0.18
b ₄	6.83	5.00
Cost Function	250.00	110.86
l² norm	40.88	0.0147
Computer Time (CPU sec. CDC 855)	---	156,370.00
No. of Iterations	---	25

Table 4. Convergence History of The Optimum Design (Example 3)

Parameter	Initial Value	Final Value
Design Variables:		
b ₁	162.32	200.42
b ₂	30.98	25.04
b ₃	-0.18	46.99
b ₄	5.00	-5.28
Cost Function	46.91	33.85
l² norm	5.29	0.19
Computer Time (CPU sec. CDC 855)	---	319,000.00
No. of Iterations	---	55

Table 5. Convergence History of The Optimum Design (Example 4)

Parameter	Initial Value	Final Value
Design Variables:		
T_0	300.00	395.85
b_1	168.39	106.59
b_2	47.41	-46.92
b_3	40.67	23.42
b_4	-0.99	-24.29
Cost Function	0.8032	0.6992
l^2 norm	111.40	4.08788
Computer Time (CPU sec. IBM 4381)	---	196,800.00
No. of Iterations	---	46

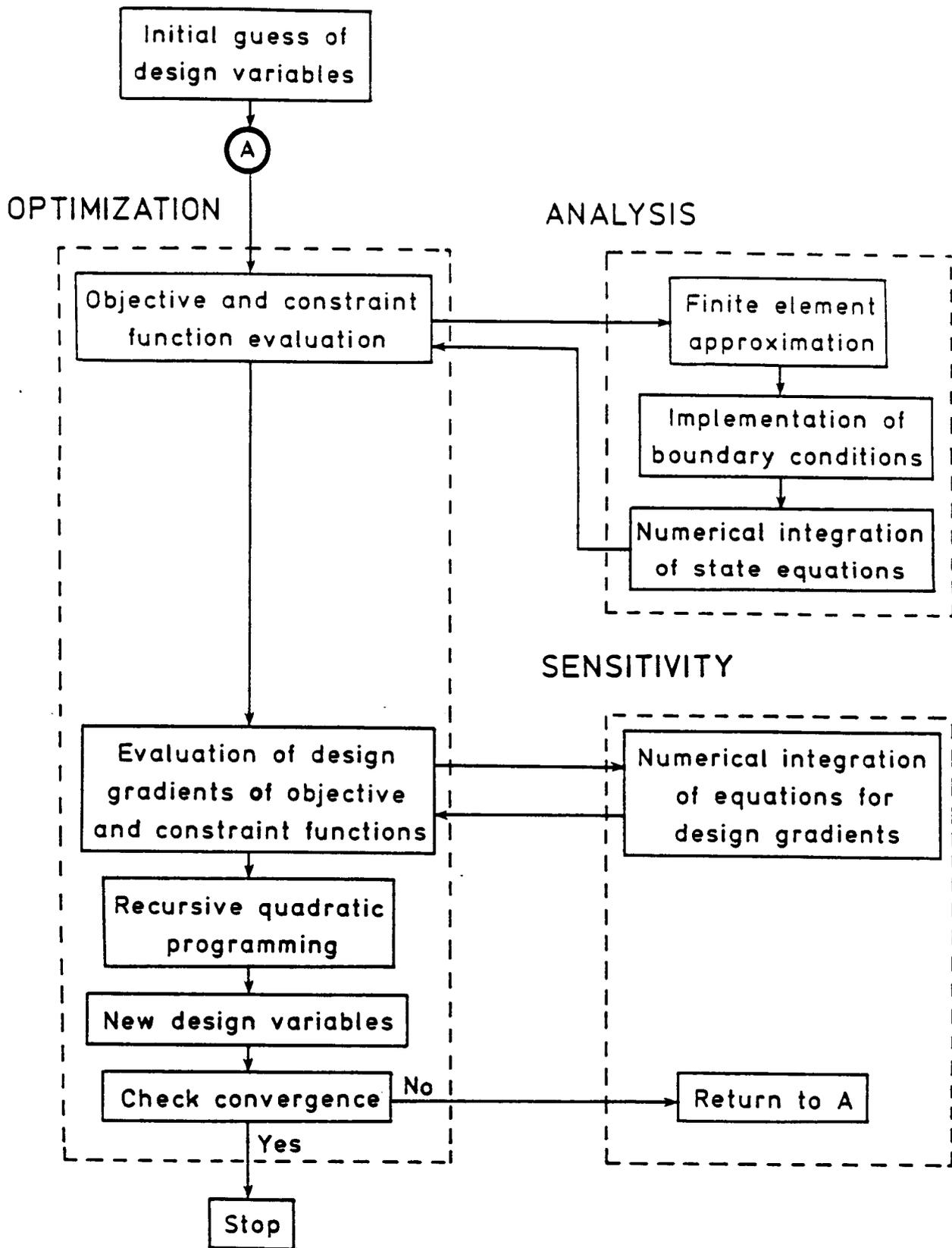


Figure 1. Flow chart of the proposed CAD methodology which includes the Analysis, the Sensitivity and the Optimization modules.

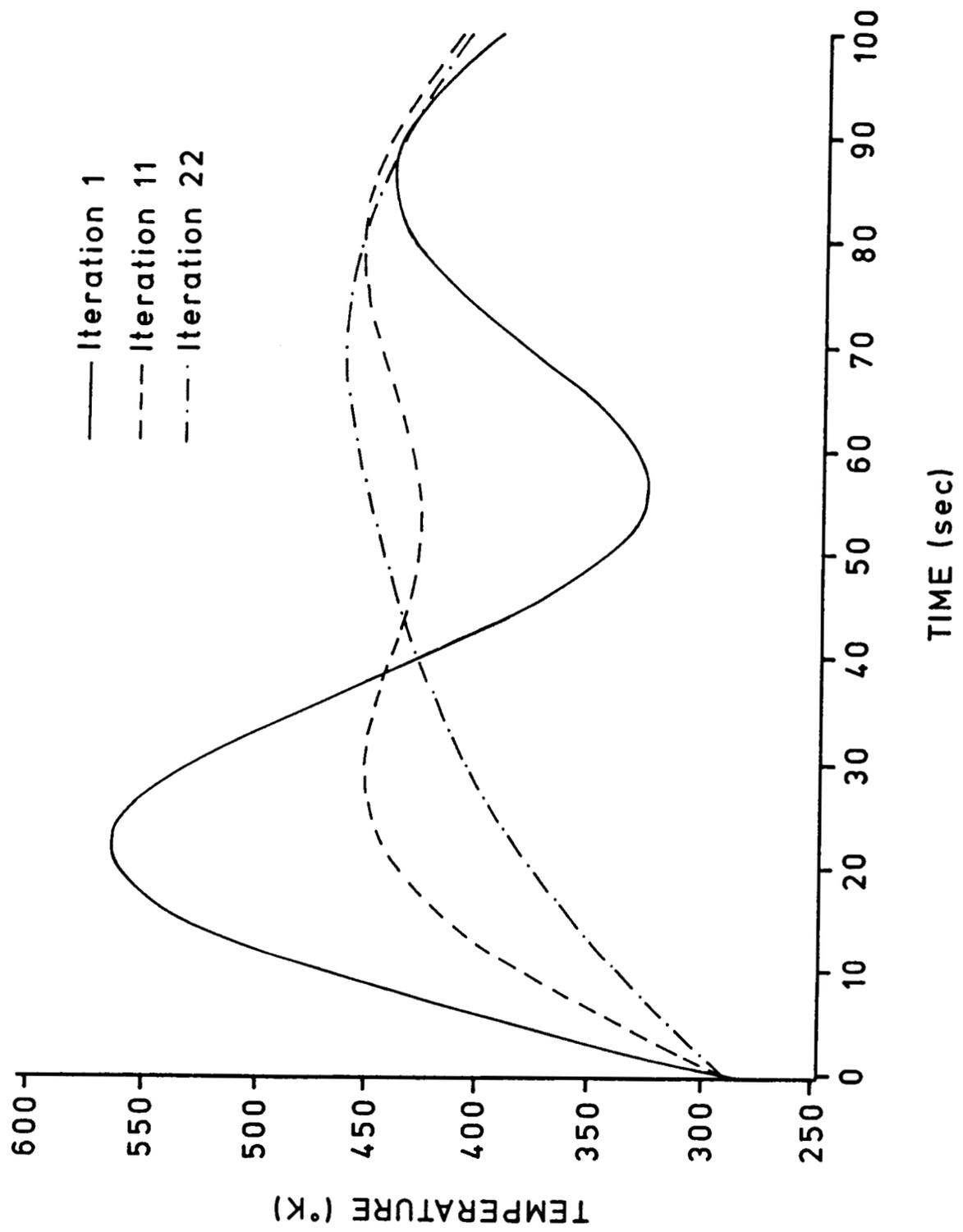


Figure 2. Profiles of the cure cycle (Example 1).

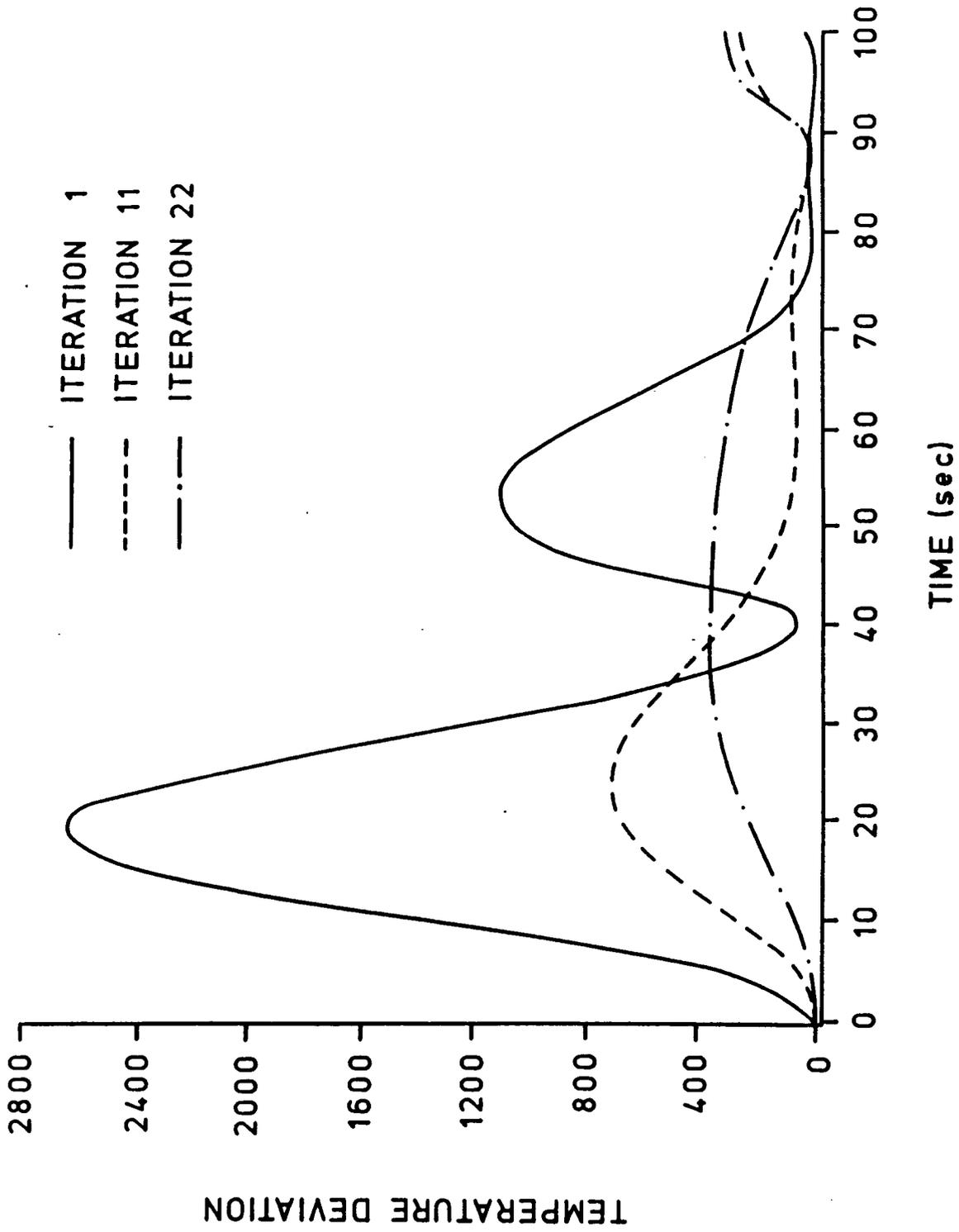
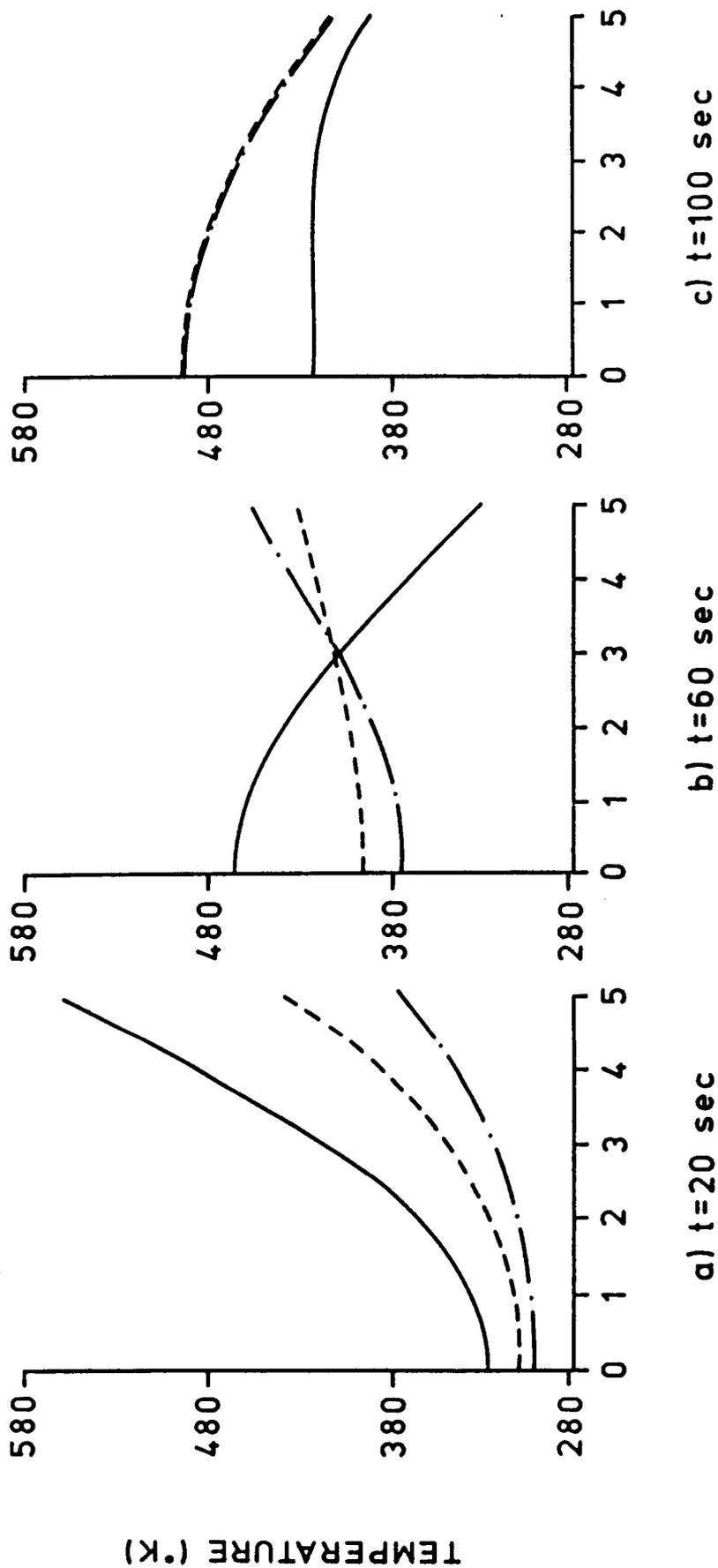


Figure 3. Profiles of the Least-Squared temperature deviation (Example 1).

— ITERATION 1
 - - - ITERATION 11
 - · - ITERATION 22



DISTANCE FROM MIDPLANE (mm)

Figure 4. Profiles of temperature distribution (Example 1).

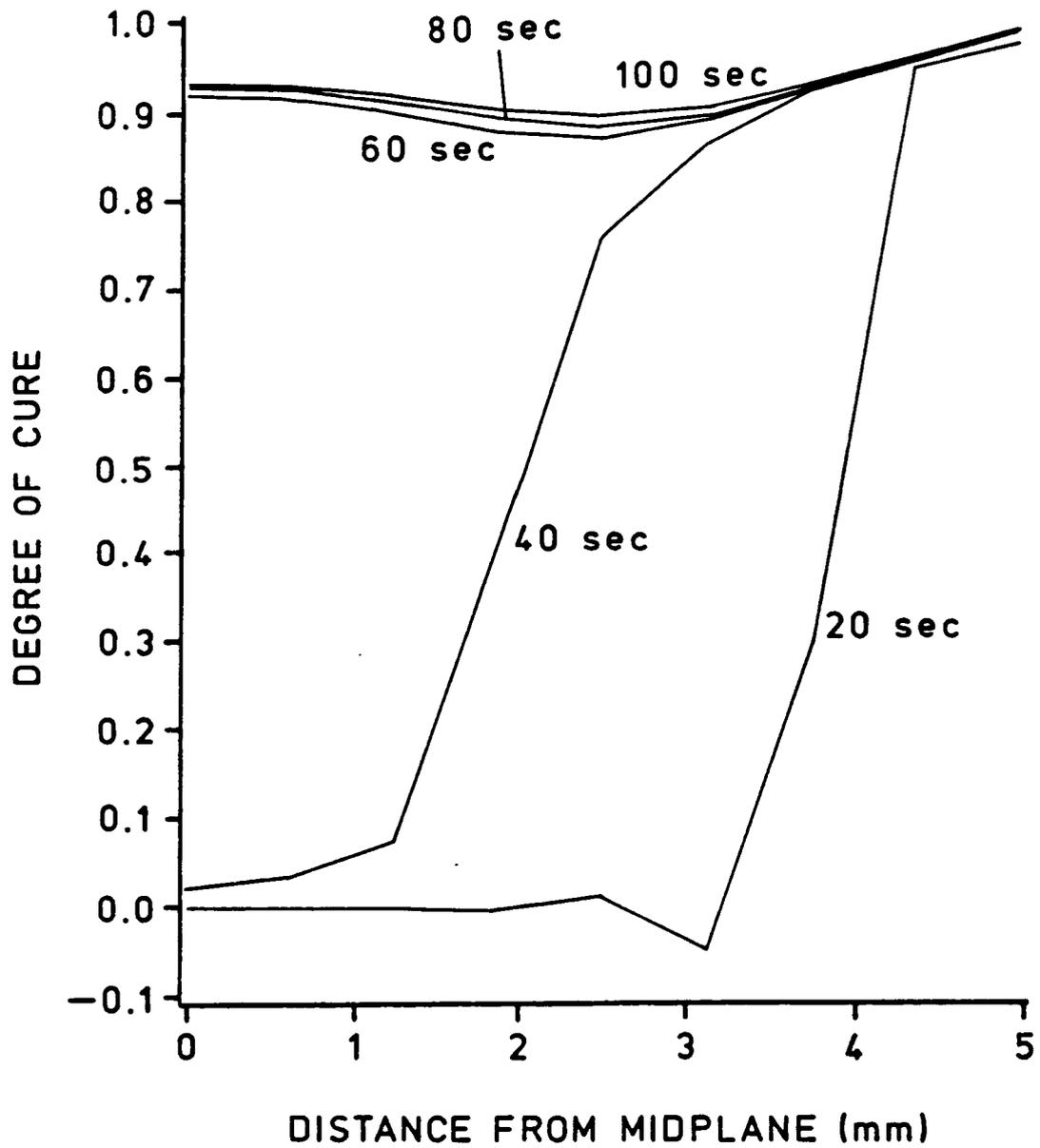


Figure 5. Distribution of the degree of cure at iteration 1 (Example 1).

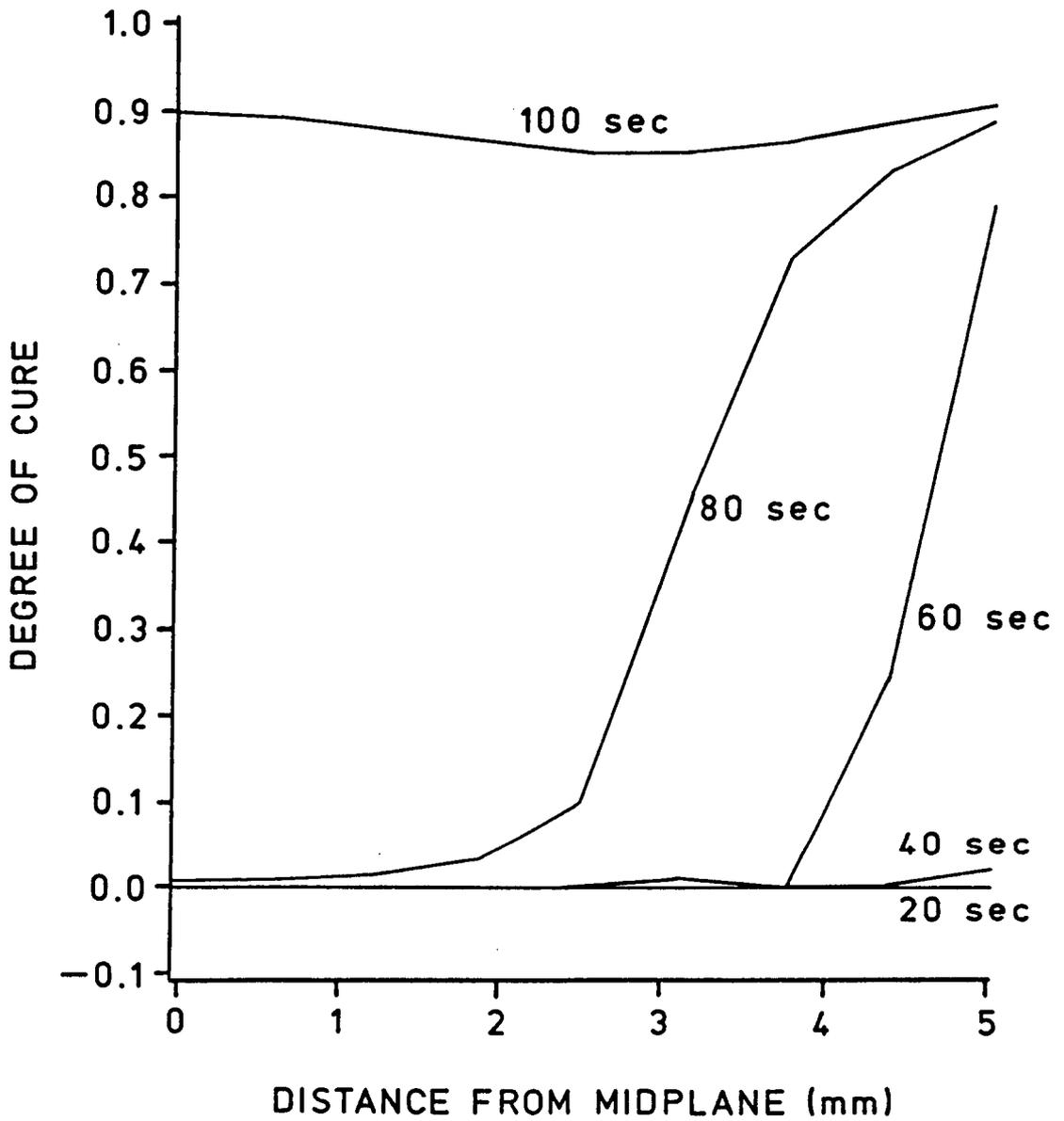


Figure 6. Distribution of the degree of cure at iteration 22 (Example 1).

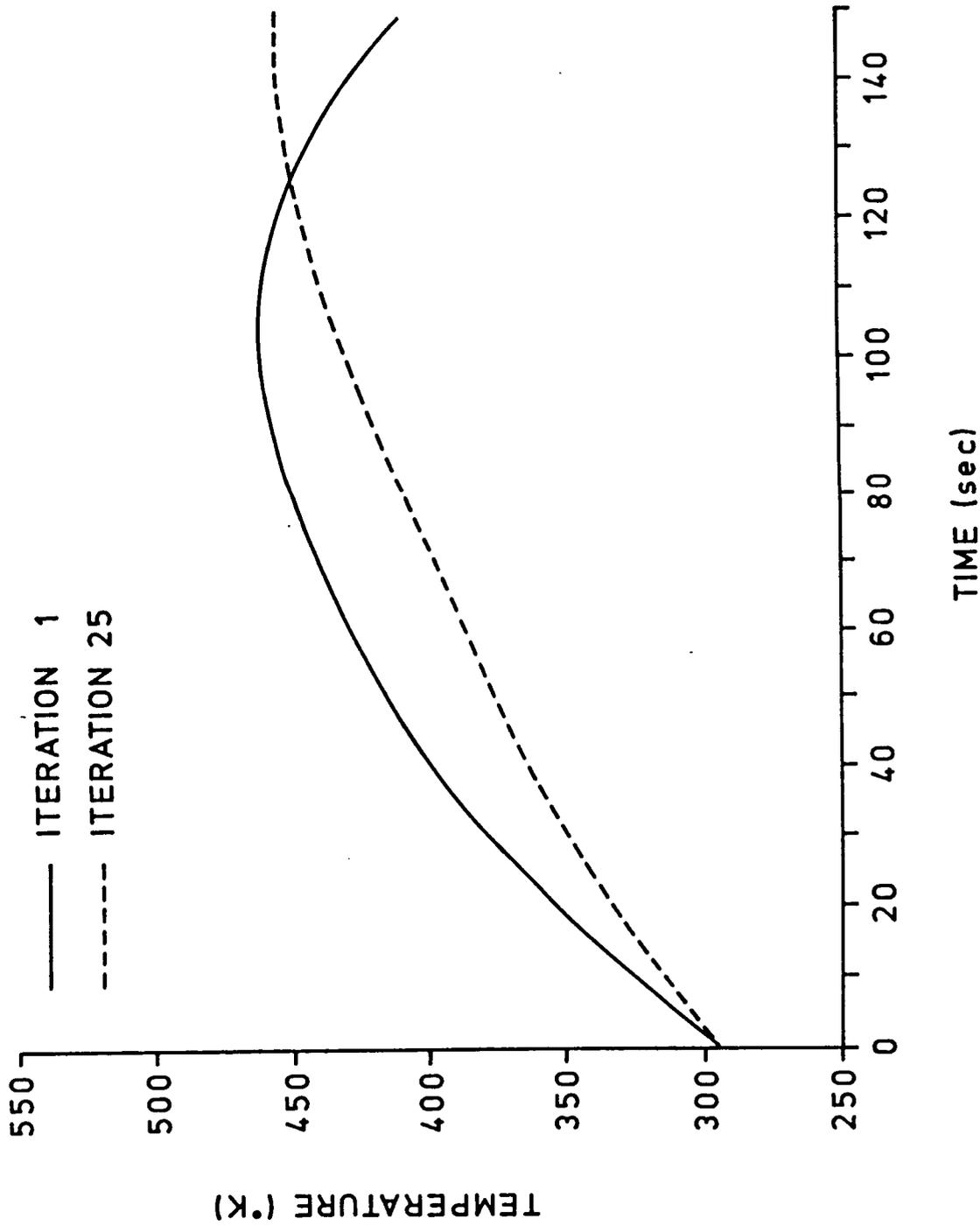


Figure 7. Profiles of the cure cycle (Example 2).

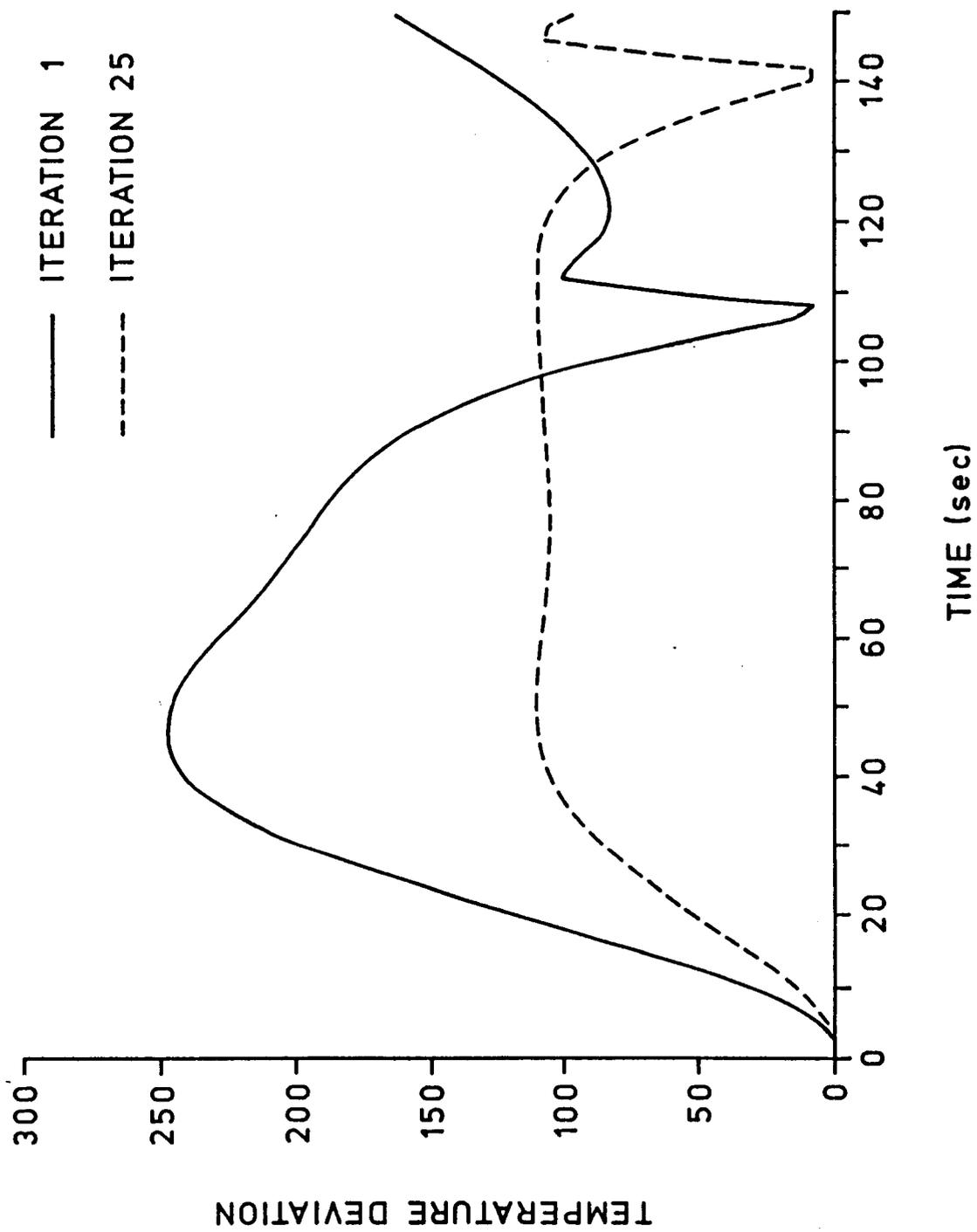


Figure 8. Profiles of the Least-Squared temperature deviation (Example 2).

— ITERATION 1 (Optimal soln. of Example 1)
 - - - - ITERATION 25 (Optimal soln. of Example 2)

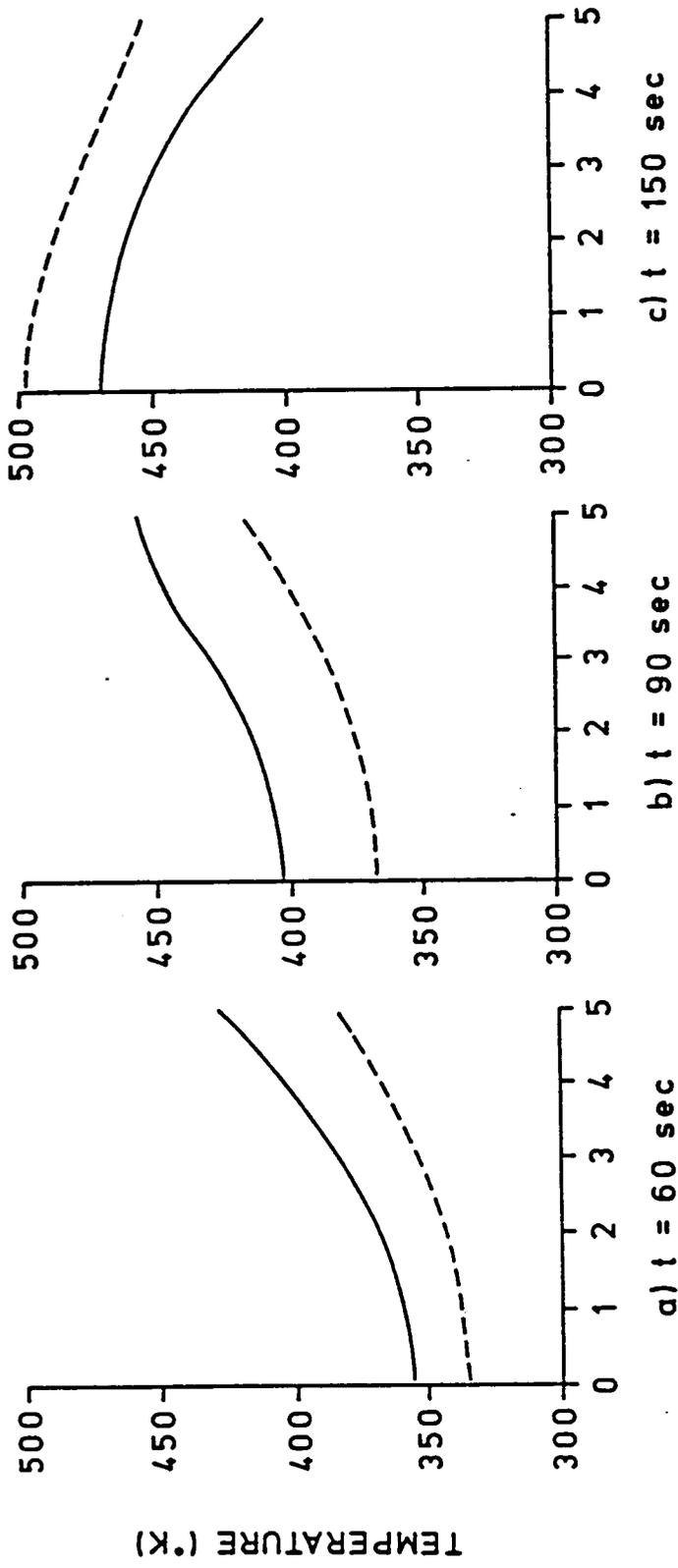


Figure 9. Profiles of temperature distribution (Example 2).

9

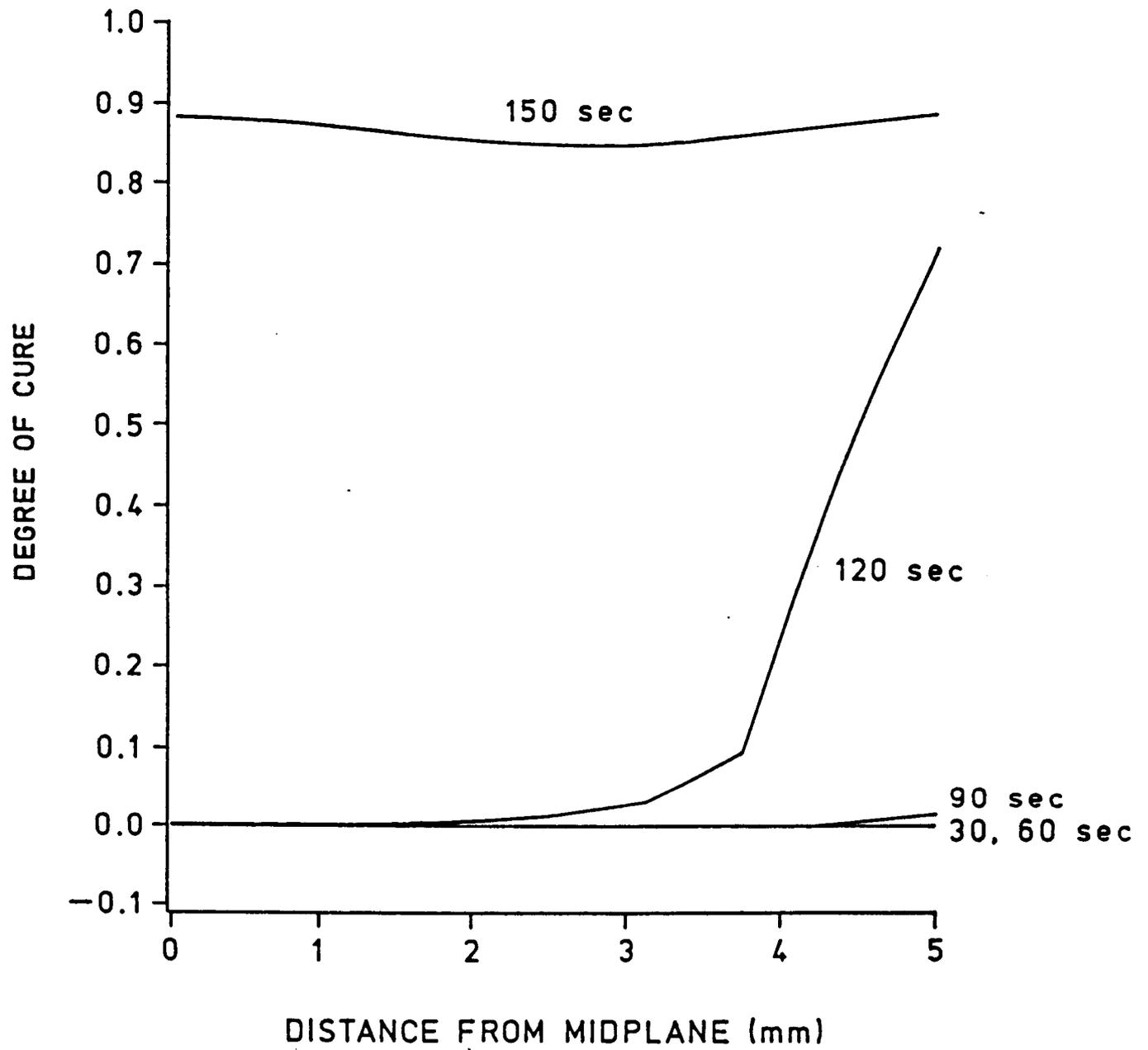


Figure 10. Distribution of the degree of cure at iteration 25 (Example 2).

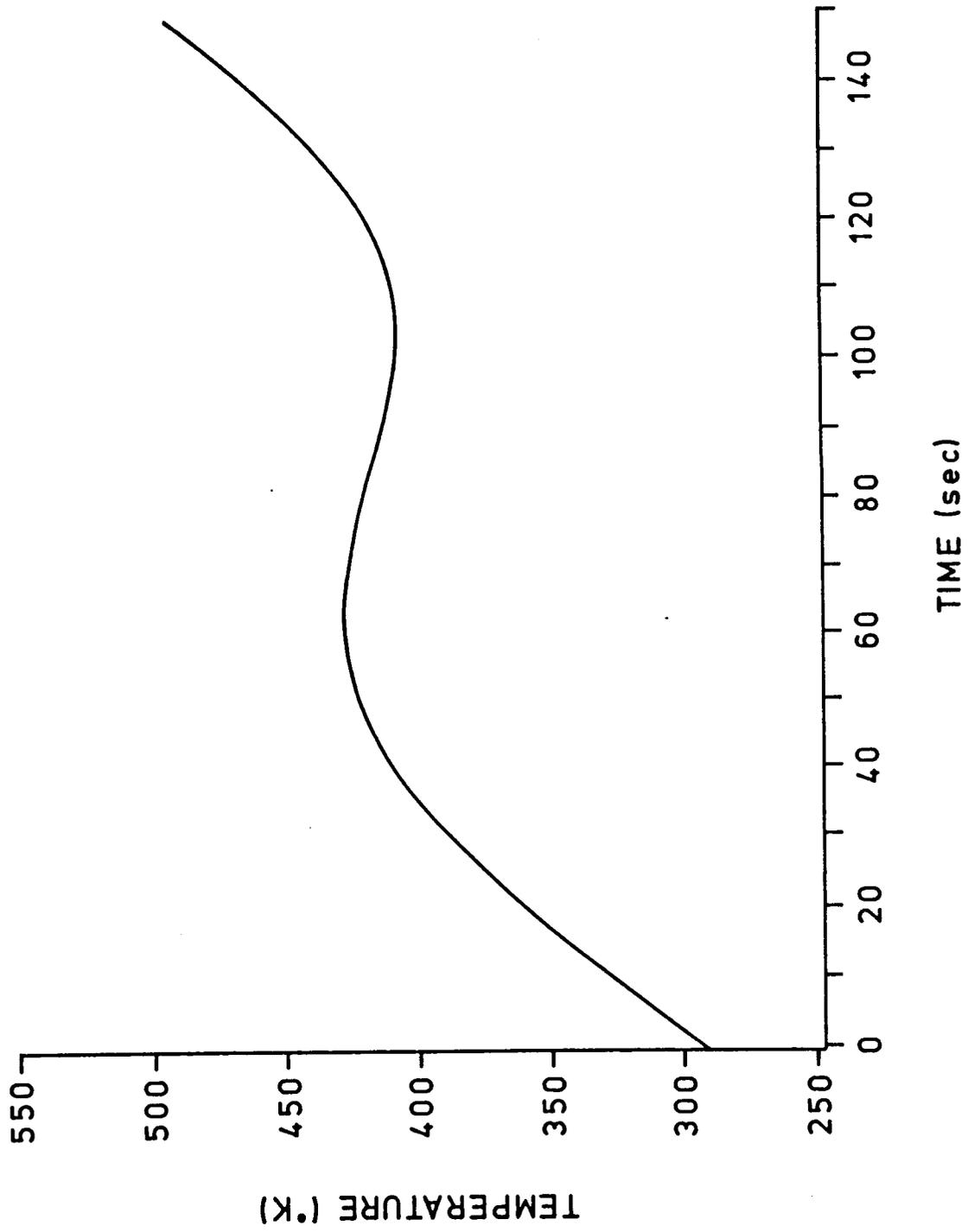


Figure 11. Profile of the optimal cure cycle at iteration 55 (Example 3).

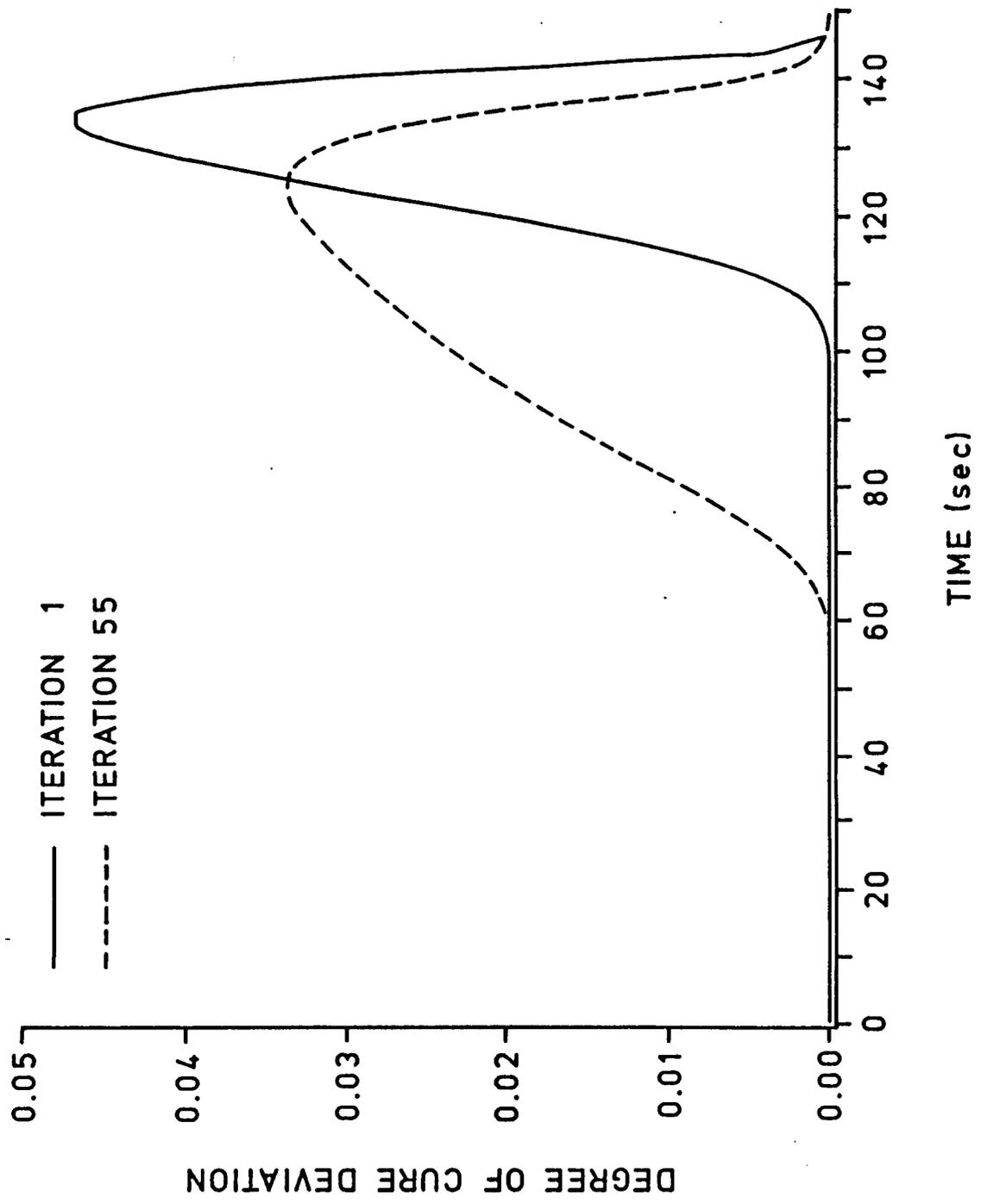


Figure 12. Profiles of the Least-Squared degree of cure deviation (Example 3).

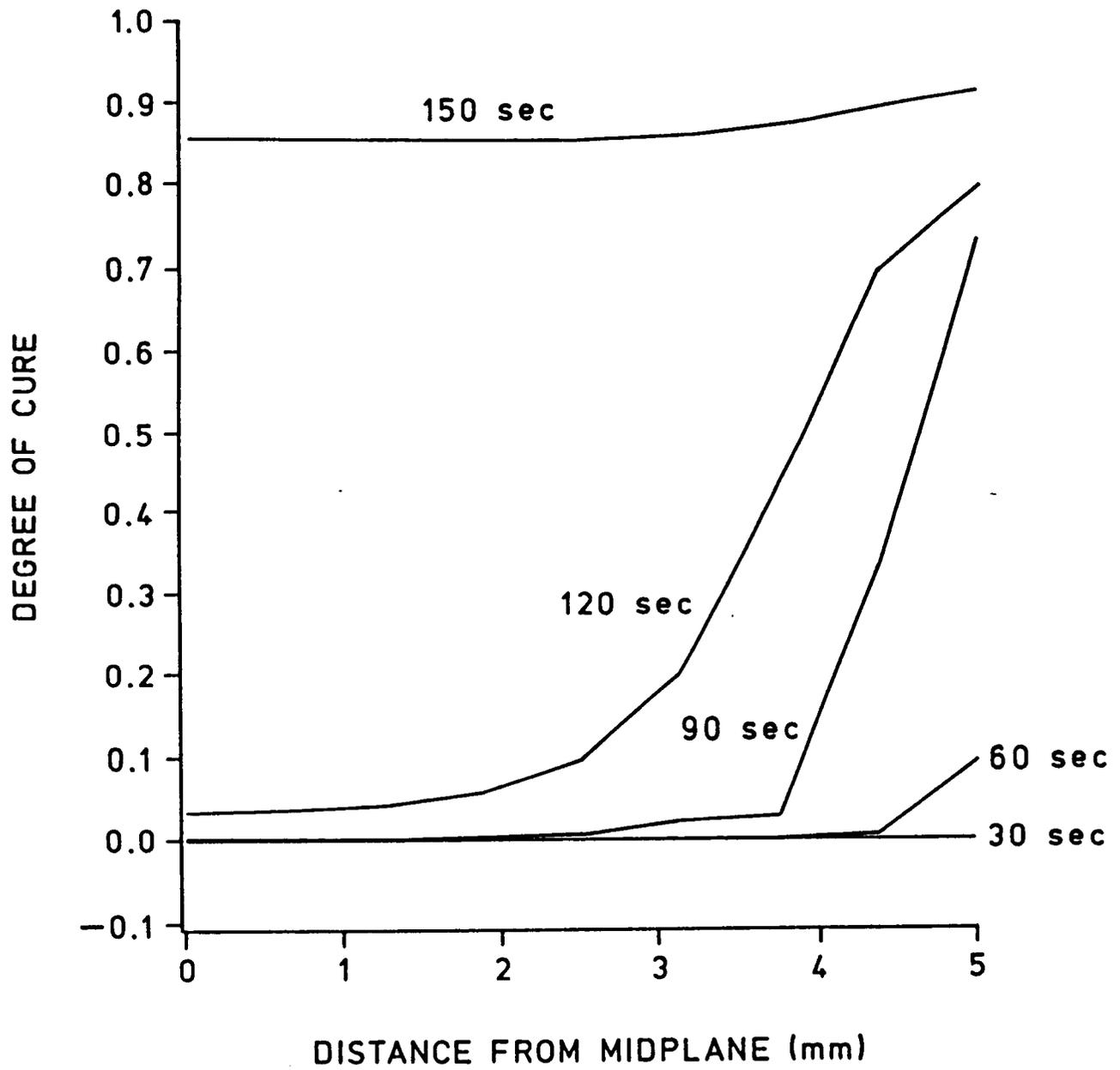


Figure 13. Distribution of the degree of cure at iteration 55 (Example 3).

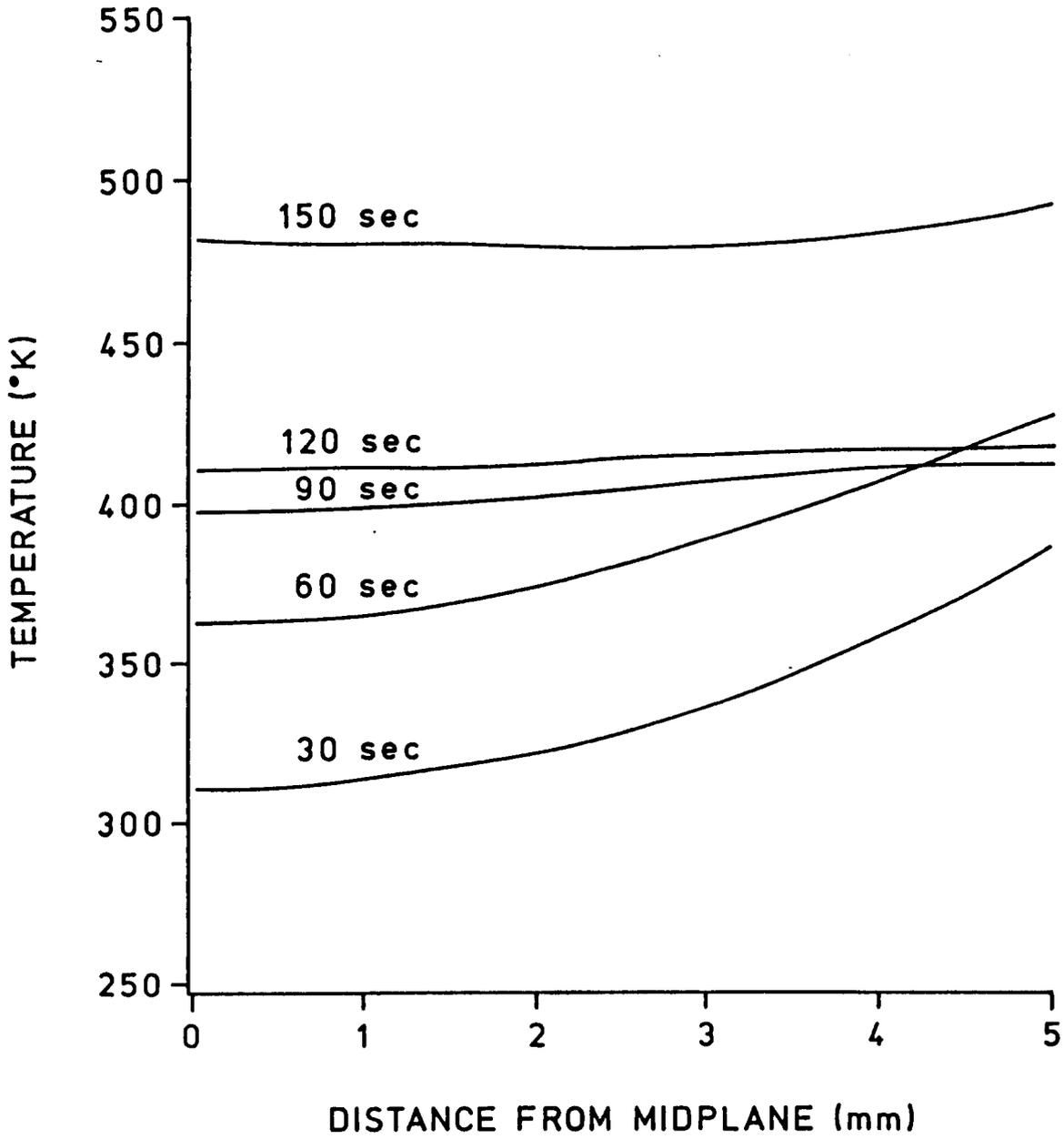


Figure 14. Profiles of temperature distribution (Example 3).

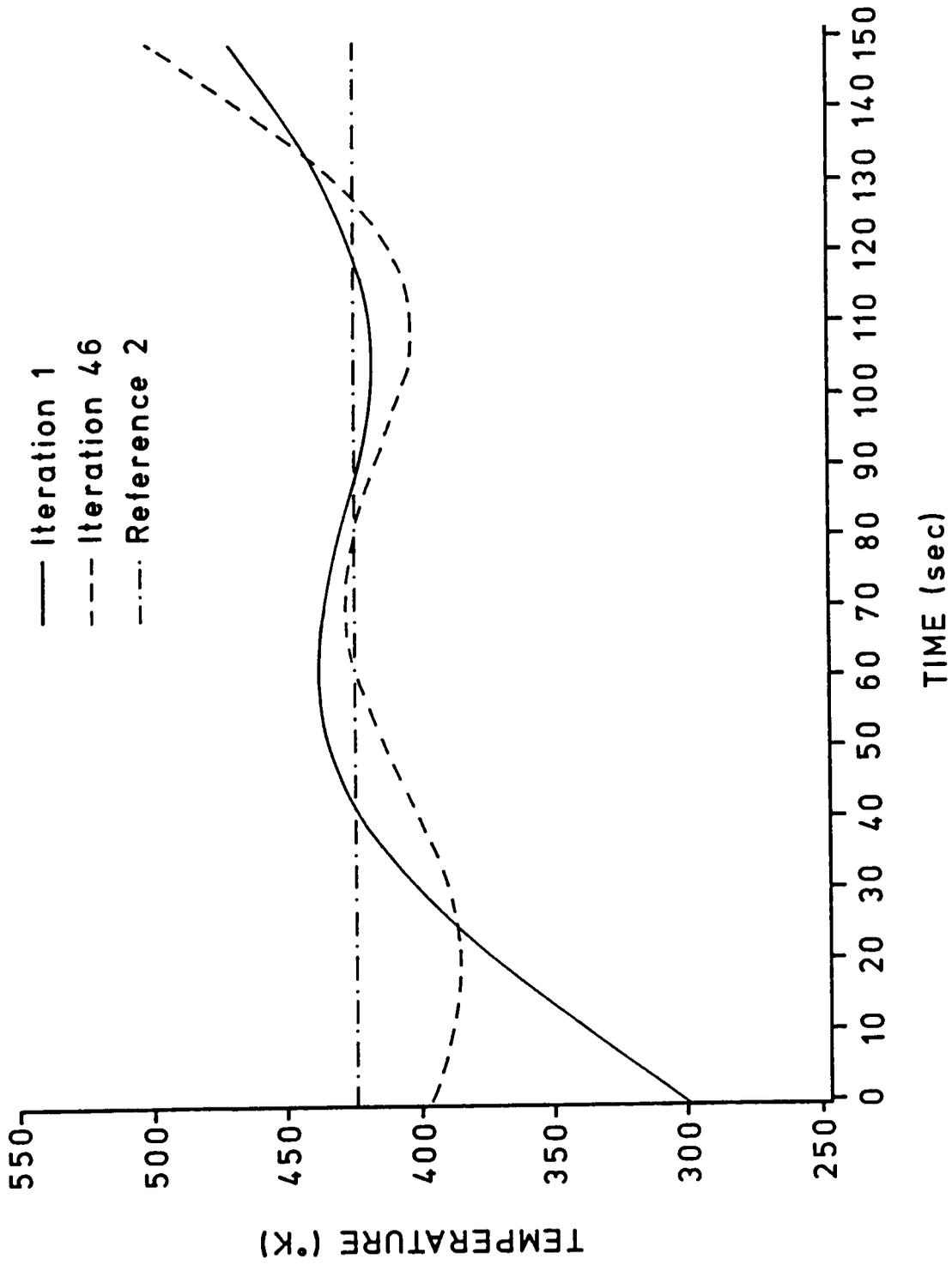


Figure 15. Profiles of the cure cycle (Example 4).

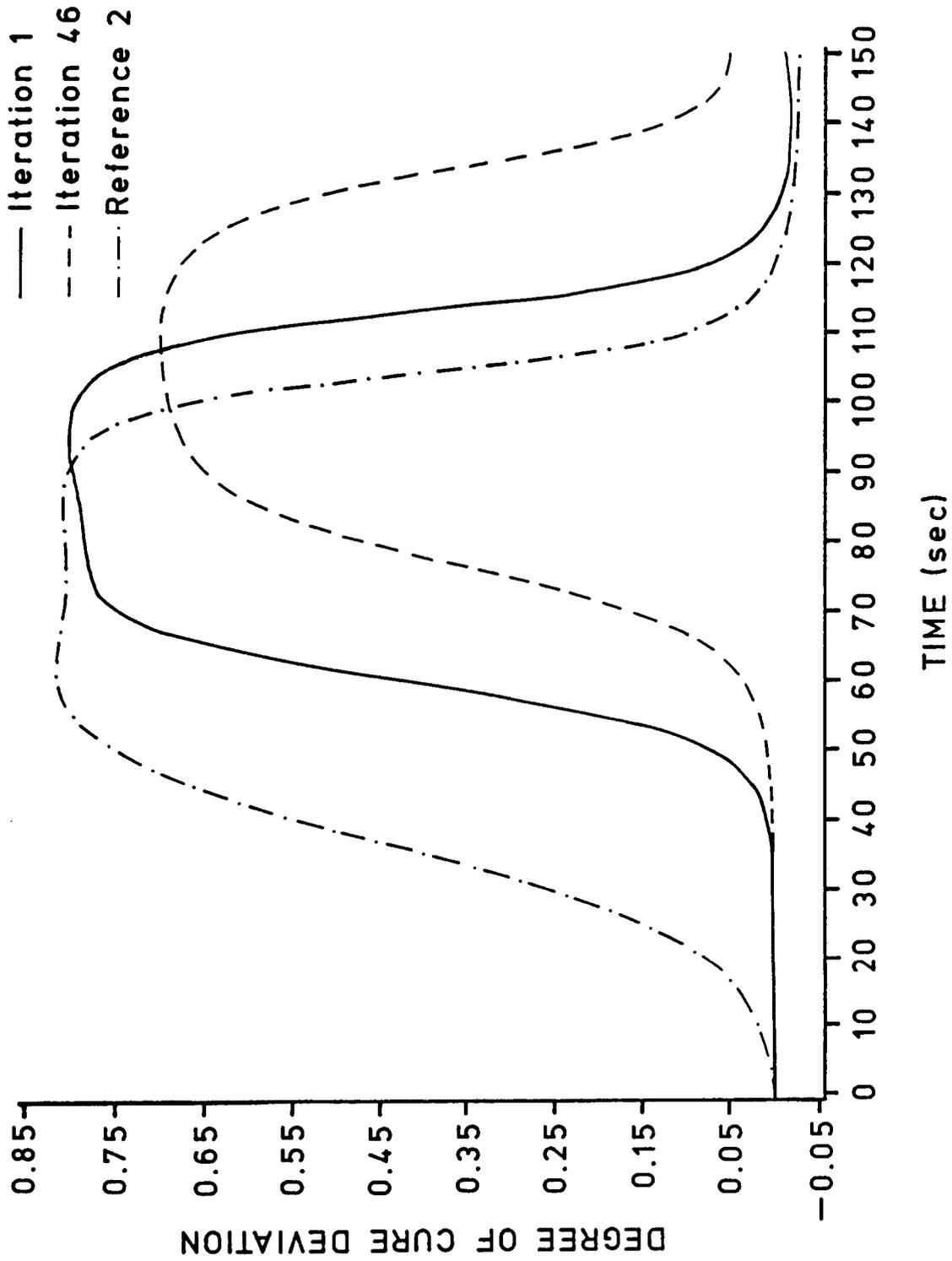


Figure 16. Profiles of the degree of cure deviation between the outer surface and the center plane of the SMC.

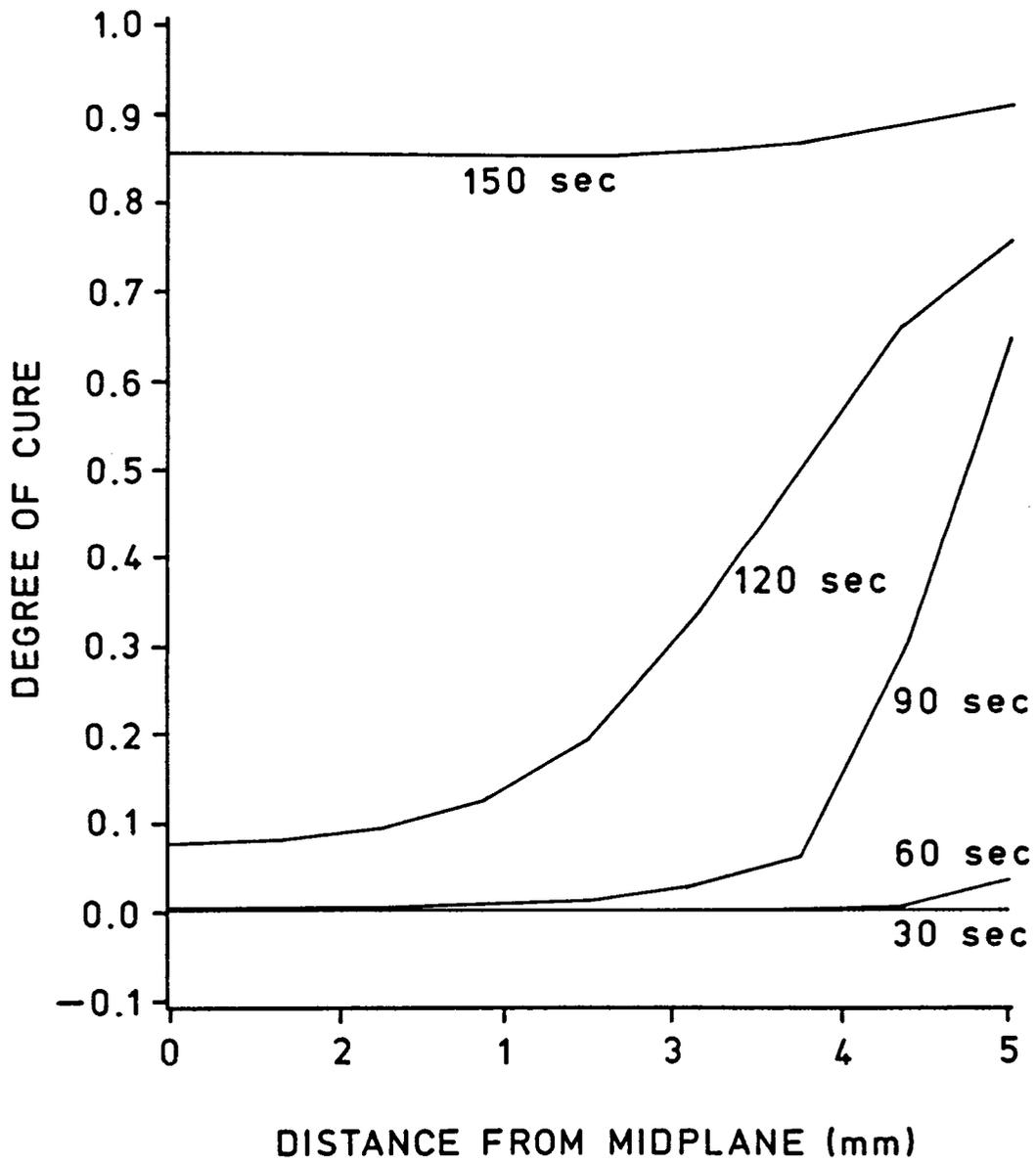


Figure 17. Distribution of the degree of cure at iteration 46 (Example 4).

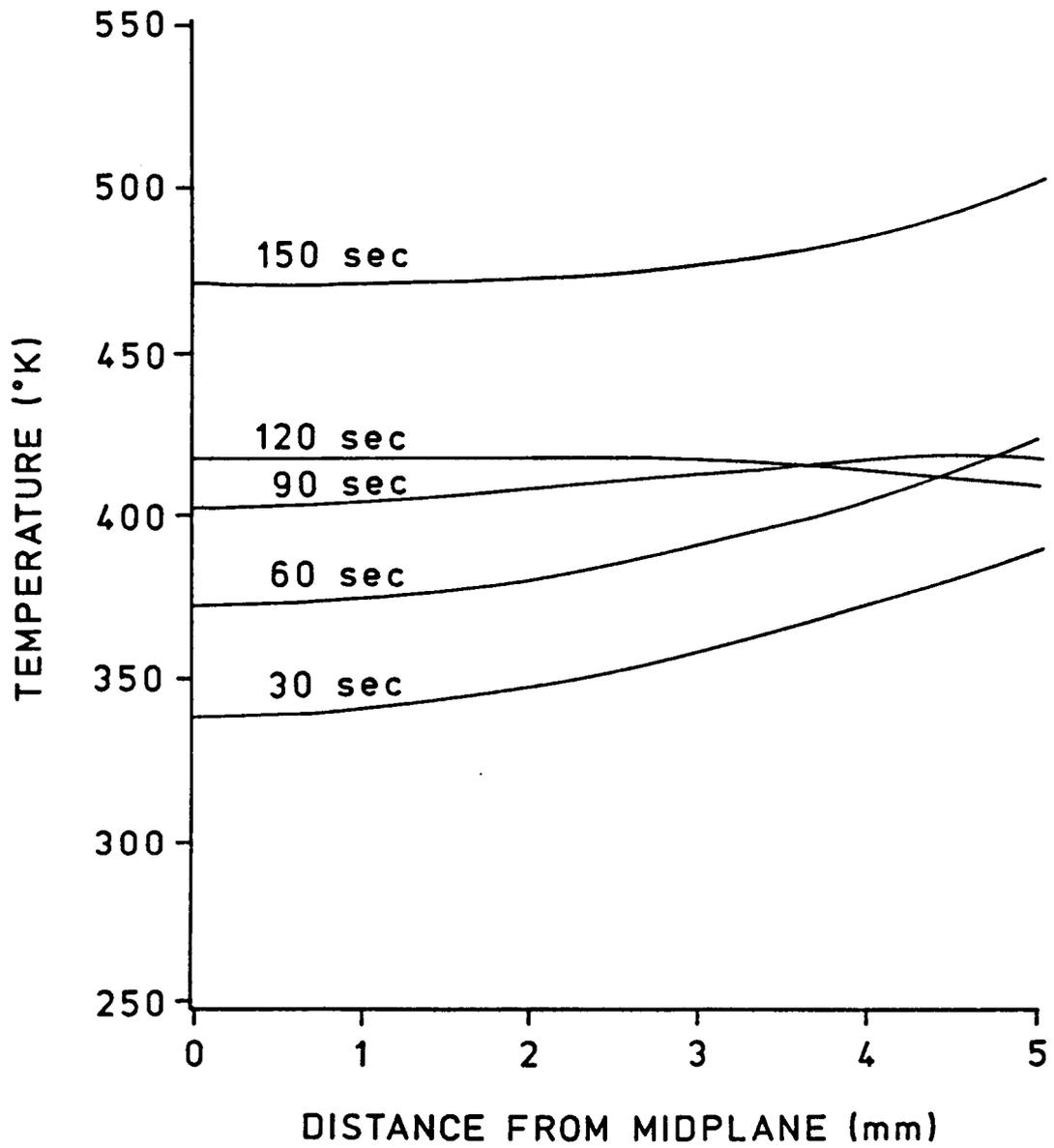


Figure 18. Profiles of temperature distribution at iteration 46 (Example 4).



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16. Abstract Fiber-reinforced composite laminates are used in many aerospace and automobile applications. The magnitudes and durations of the cure temperature and the cure pressure applied during the curing process have significant consequences for the performance of the finished product. The objective of this study is to exploit the potential of applying the optimization technique to the cure cycle design. Using the compression molding of a filled polyester Sheet Molding Compound (SMC) as an example, a unified Computer Aided Design (CAD) methodology, consisting of three uncoupled modules, (i.e., Optimization, Analysis and Sensitivity Calculations), is developed to systematically generate optimal cure cycle designs. Various optimization formulations for the cure cycle design are investigated. The uniformities in the distributions of the temperature and the degree of cure within the specimen cured under the optimal cycles are compared with those resulting from conventional isothermal processing conditions with pre-warmed platens. Recommendations with regards to further research in the computerization of the cure cycle design are also addressed.					
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