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EFFICIENT DYNAMIC OPTIMIZATION OF THE MMA CELL-CAST PROCESS FOR PLASTIC SHEET PRODUCTION

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Abstract: The dynamic optimization of a heating process for plastic sheet production is modeled. The formulation is based on the representation of the one-directional heating process taking place in this system, and includes kinetic equations for the reaction of methylmethacrylate. The mathematical model is cast as a Partial Differential Equation (PDE) system, and the optimal profile calculation turns out to be a dynamic optimization problem with distributed parameter systems. A simultaneous approach was selected to solve the dynamic optimization problem. By the full discretization of all variables, a Non-Linear Programming (NLP) model is obtained and solved by using the CONOPT solver. We present results about the dynamic optimization of two plastic sheets of different thickness and compared them against simpler operating policies.

1. INTRODUCTION

It has been widely recognized that the transient operation of processing equipment can be improved by the use of optimal dynamic operation policies (Kameswaran and Biegler, 2006). Up to now, most of the dynamic optimization techniques applications have been aimed at examining the optimal transient behavior of lumped parameter systems. As computing power and better large scale nonlinear solvers become available one of the natural next steps consists in addressing the optimal dynamic behavior of distributed parameter systems (Biegler *et al.*, 2003).

For medium to large scale systems, two optimal control methodologies seem to dominate the numerical solution of optimal control problems. On one hand, the resulting set of differential and algebraic equations (DAE) comprising the dynamic mathematical model of the addressed system is subject to partial discretization of the output variables and the remaining differential system is numerically integrated (Allgor and Barton, 1999).

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This approach is commonly called the sequential optimal control approach. On the other hand, in the simultaneous approach both the set of manipulated and controlled variables are fully discretized leading to a set of algebraic equations. Therefore, the optimal control problem is transformed into a nonlinear program (Kameswaran and Biegler, 2006). Although it has been claimed that the sequential approach is easy to use it has some disadvantages. Presently, it seems to be unable to handle open-loop unstable systems without previous stabilization. Quite the contrary, it has been shown that the simultaneous approach is able to efficiently handle unstable systems (Flores-Tlacuahuac, 2005). Moreover, the simultaneous approach demand good initialization strategies and normally state of the art nonlinear solvers able to handle large systems arising from system discretization. However, with ever increasing advances in computing power and the availability of large scale nonlinear optimization solvers, it seems that the simultaneous approach will be widely used for approaching large scale and highly nonlinear optimal control problems. Traditional poly-Methyl Methacrylate (PMMA) manufacturing process is based on placing the plastic sheets inside warm water baths to allow the polymerization reactions to proceed until monomer and initiators are exhausted. However, industrial experience indicates that the quality of the PMMA plastic sheets tend to display wide variations in molecular weight distributions due to nonuniform heating patterns. To achieve uniform polymerization properties, it has been suggested to carry out the set of PMMA polymerization reactions using an oven where heating proceeds by circulating warm air. Initial pilot plant tests have indicated that by using this last PMMA plastic sheet manufacturing process, uniform molecular weight distributions could be achieved. In this work our aim is to propose optimal heating policies able to produce PMMA plastic sheets with uniform temperature distributions. Even, when presently no molecular weight distributions were computed, forcing optimal heating profiles should lead to PMMA plastic sheets with homogeneous properties. In this paper we use the simultaneous dynamic optimization framework to address the optimal dynamic behavior of the cell-cast process for PMMA plastic sheet manufacture.

2. CELL-CAST PROCESS

Process description. In the typical casting of acrilic sheet, molds formed by two glass plates which are separated by a peripheral gasket sealer and clamped together, are filled with casting syrup through a gap left in the gasket. The casting syrup is made up of partially polymerized monomer (20%) which, once placed in the mold, is inserted into a furnace which is heated by circulating warm air (see Figure1). It will be appreciated that this prior art procedure, which has been generally adopted by the industry with certain variations an modifications for the production of cast sheets of the indicated nature, is relatively cumbersome. It is extremely important to control the progress of the polymerization throughout the procedure and to create suitable mild thermic conditions which, in turn, requires speedy and effective dissipation of excess heat, due to the low heat capacity of air, effective control of the thermal conditions during the operation is very important, so that, the heating is affected by the circulating air (Rivera-Toledo, 2006).

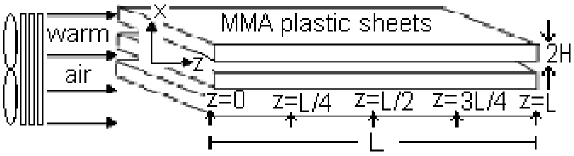

Fig. 1. Cell-cast process for PMMA plastic sheet manufacture

Kinetic model. Besides the conventional chemical kinetics, physical phenomena related to the diffusion of various chemical reactive species are very important in free-radical polymerization reactions. In fact, at high monomer conversions, almost all elementary reactions can become diffusion-controlled. Reactions which are influenced by diffusion phenomena include termination of live macro-radicals (Odian, 1991), propagation of a growing chain, and chemical initiation reactions. Diffusion-controlled termination, propagation, and initiation reactions have been related to the well-known phenomena of gel effect, glass effect, and cage effect, respectively. In the past 30 years, several models have been published dealing with the mathematical description of diffusion-controlled kinetic rate constants in free-radical polymerization (Dubé *et al.*, 1997). The reaction mechanism adopted here consists of a simple approximation for the well know free-radical polymerization kinetics featuring straightforward initiation, propagation, and termination reactions (Achilias and Kiparissides, 1992). The following assumptions are taken (1) the diffusion effect is negligible since we are interested in the thermal process behavior, then the polymer processing is controlled by the chemical kinetic, (2) only the mass balance for the monomer conversion, initiator concentration and the zeroth moment of growing radical distribution are considered. **PMMA mathematical model**. The sheet reactor model is considered for the PMMA plastic sheet production. For lack of space, in this paper we indicate briefly some assumptions on which the mathematical model rests, nevertheless, on previous work (Rivera-Toledo, 2006) we discuss in detail the limitations and scopes of the model. The mathematical model was derived assuming that the heating source resulting from polymer reaction is a function of the local temperature. It was also assumed that polymer properties, like density, heat capacity, thermal conductivity are constants and they are average values for MMA and PMMA. To get the one-dimensional dynamic energy balance, the total heat entering and leaving at the z coordinate was modeled by the Fourier law and the rate of change of energy in the control volume was obtained applying the shell energy balance method. From a reaction engineering point of view, the PMMA plastic sheet process can be considered as taking place in a constant volume batch reactor. Dynamic mass and energy balances coupled trough polymerization kinetics (Achilias and Kiparissides, 1992) describe monomer conversion, initiator concentration and the zeroth moment of growing radical distribution dynamic time evolution. Air is circulated through the forced convection mechanism inside the oven to provide the required energy to rise up the plastic sheet temperature until a point where significant polymerization rates take place. Inside the monomer, the dominant heat transfer mechanism is conduction.

The modeling equations for the sheet reactor consist of the following energy and mass balances:

$$\frac{\partial T}{\partial t} = \alpha\frac{\partial^2 T}{\partial z^2} - \frac{h}{\rho C_p H}(T - T_a) + \frac{Q_{rxn}}{\rho C_p H} \tag{1}$$

$$\frac{dX}{dt} = k_p(1 - X)\lambda_0 \tag{2}$$

$$\frac{d\lambda_0}{dt} = -\frac{\epsilon\lambda_0^2}{1+\epsilon X}k_p(1 - X) + 2fk_dI - k_t\lambda_0^2 \tag{3}$$

$$\frac{dI}{dt} = -k_dI - \frac{\epsilon I}{1+\epsilon X}\lambda_0 k_p(1 - X) \tag{4}$$

and the initial and boundary conditions are given by

$$\begin{aligned} &t = 0 \quad T = T_0, X = X_0, \lambda_0 = \lambda_{0_0}, I = I_0,\ 0 \leqslant z \leqslant L \\ &z = 0\,, -k\frac{\partial T}{\partial z} = h(T - T_a), \quad \forall t > 0 \\ &z = L\,, \frac{\partial T}{\partial z} = 0, \quad \forall t > 0 \end{aligned} \tag{5}$$

where T is the polymer temperature, T_0 is the initial monomer temperature, T_a is the surrounding temperature, t is the polymerization time, z is the axis for the sheet length, X is the monomer conversion, λ_0 is the zeroth moment of growing radical distribution, I is the initiator concentration, L is the sheet length , H is the sheet thickness, k is the monomer average thermal conductivity, ρ is the density, C_p is the heat capacity, f is the initiator efficiency, ϵ is the volume expansion factor, α is the thermal diffusivity, h is the heat transfer coefficient, k_d, k_p, k_t are the kinetic coefficients for initiator, propagation and termination stages, respectively, and Q_{rxn} stands for the heat released by the polymerization reactions.

The dimensionless variables for the polymer temperature, air temperature, position, time, zeroth moment of growing radical distribution and the initiator concentration are defined as follows:

$$\begin{aligned} &\theta = \frac{T}{T_0}, \quad \theta_a = \frac{T_a}{T_0}, \quad \zeta = \frac{z}{L} \\ &\tau = \frac{\alpha t}{H^2}, \quad \bar{\lambda}_0 = ln(\lambda_0), \quad \bar{I} = \frac{I}{I_0} \end{aligned}$$

the set of energy and mass balances and initial and boundary conditions, are written in dimensionless form as:

$$\frac{\partial\theta}{\partial\tau} = a^2\frac{\partial^2\theta}{\partial\zeta^2} + Bi(\theta_a - \theta) + A_9\frac{(1-X)^2}{1+\epsilon X}e^{\bar{\lambda}_0 - A_2/\theta} \tag{6}$$

$$\frac{dX}{d\tau} = A_1(1 - X)e^{\bar{\lambda}_0 - A_2/\theta} \tag{7}$$

$$\frac{d\bar{\lambda}_0}{d\tau} = -A_4\frac{1-X}{1+\epsilon X}e^{\bar{\lambda}_0 - A_2/\theta} + A_6e^{-\bar{\lambda}_0 - A_5/\theta} - A_7e^{\bar{\lambda}_0 - A_8/\theta} \tag{8}$$

$$\frac{d\bar{I}}{d\tau} = A_4\frac{1-X}{1+\epsilon X}e^{\bar{\lambda}_0 - A_2/\theta} - A_3e^{-A_5/\theta} \tag{9}$$

$$\begin{aligned} &\tau = 0 \quad \theta = \theta_0, X = X_0, \bar{\lambda}_0 = ln(\lambda_{0_0}), \bar{I} = 1, \quad \forall 0 \leqslant \zeta \leqslant 1 \\ &\zeta = 0 \quad \frac{\partial\theta}{\partial\zeta} = Bi(\theta_a - \theta), \quad \forall\tau > 0 \\ &\zeta = 1 \quad \frac{\partial\theta}{\partial\zeta} = 0, \quad \forall\tau > 0 \end{aligned} \tag{10}$$

where $Bi = hH/k$ is the Biot number.

3. DYNAMIC OPTIMIZATION

To compute dynamic optimal heating profiles we use the following objective function:

$$\min\int_0^\tau \left[\left(1 - \frac{\theta}{\theta^d}\right)^2 + \left(1 - \frac{\theta_a}{\theta_a^d}\right)^2\right] d\tau \tag{11}$$

subject to the partial differential and algebraic equations (PDAE) and the initial and boundary conditions (6-10). In the above equation θ^d and θ_a^d stand for the desired values of plastic sheet and air temperatures, respectively. Our aim is to compute the air temperature as time function $\theta_a(t)$ so to drive the plastic sheet temperature θ to its desired profile as soon as possible.

Using the simultaneous approach for solving the dynamic optimization problem given by the objective function (Eq.11) and the PDAE (Eqs.6-10) problem, these were converted into a NLP problem by approximating the states $(\theta, X, \bar{I}, \bar{\lambda})$ and control (θ_a) variables by the application of the method of lines (Schiesser, 1991) for the spatial coordinate and orthogonal collocation on finite elements for handling the time coordinate. The discretized NLP problem is given by the objective function:

$$\min_{\theta_{a_{mk}}} \sum_{m=1}^{Ne} \Delta\tau_m \sum_{k=1}^{N_c} \sum_{j=1}^{N_z} \left[\left(1 - \frac{\theta_{mkj}}{\theta^d}\right)^2 + \omega_a \left(1 - \frac{\theta_{a_{mk}}}{\theta_a^d}\right)^2 \right] \tag{12}$$

The discretized energy balance is given by the following two algebraic equations:

$$\theta_{mkj} = \theta_{mj}^0 + \Delta\tau_m \tau_{tr} \sum_{n=1}^{Nc} A_{nk} \dot{\theta}_{mnj} \tag{13}$$

where θ_{mj}^0 is the temperature at starting point at each element, $\Delta\tau_m$ is the length of $m-th$ finite element, τ_{tr} is the operating time, and

$$\begin{aligned} \dot{\theta}_{mnj} = {} & \frac{a^2}{\Delta\zeta^2}(\theta_{mnj+1} - 2\theta_{mnj} + \theta_{mnj-1}) + \\ & Bi(\theta_{a_{mn}} - \theta_{mnj}) + \\ & A_9 \frac{(1 - X_{mnj})^2}{1 + \epsilon X_{mnj}} e^{\bar{\lambda}_{0_{mnj}} - A_2/\theta_{mnj}} \\ & j = 2, 3, ..., N_z - 1 \end{aligned} \tag{14}$$

where $\Delta\zeta$ is the grid spacing for dimensionless length, the following algebraic equations are given by the boundary conditions:

$$\begin{aligned} \frac{-3\theta_{mn1} + 4\theta_{mn2} - \theta_{mn3}}{2\Delta\zeta} + Bi(\theta_{mn1} - \theta_{amn}) = 0 \\ 4\theta_{mnN_z} - 3\theta_{mnN_z-1} + \theta_{mnN_z-2} = 0 \\ m = 1, 2, ..., N_e, \;\; n = 1, 2, ..., N_c \end{aligned} \tag{15}$$

the temperature continuity of each element using the Lagrange orthogonal polynomial:

$$\theta_{mj}^0 = \theta_{m-1j}^0 + \Delta\tau_{m-1} \tau_{tr} \sum_{n=1}^{Nc} A_{nNc} \dot{\theta}_{m-1nj} \tag{16}$$

the starting values are $\theta_{1j}^0 = \theta_0, \;\; \theta_{a11} = \theta_{a0}$

The discretized mass balances for the monomer conversion, initiator concentration and the zeroth moment of growing radical distribution are:

$$\begin{aligned} X_{mkj} &= X_{mj}^0 + \Delta\tau_m \tau_{tr} \sum_{n=1}^{Nc} A_{nk} \dot{X}_{mnj} \\ \bar{\lambda}_{0_{mkj}} &= \bar{\lambda}_{0_{mj}}^0 + \Delta\tau_m \tau_{tr} \sum_{n=1}^{Nc} A_{nk} \dot{\bar{\lambda}}_{0_{mnj}} \\ \bar{I}_{mkj} &= \bar{I}_{mj}^0 + \Delta\tau_m \tau_{tr} \sum_{n=1}^{Nc} A_{nk} \dot{\bar{I}}_{mnj} \end{aligned} \tag{17}$$

where $X_{mj}^0, \bar{\lambda}_{0_{mj}}^0, \bar{I}_{mj}^0$ are evaluated at starting point at each element and

$$\begin{aligned} \dot{X}_{mnj} &= A_1(1 - X_{mnj}) e^{\bar{\lambda}_{0_{mnj}} - A_2/\theta_{mnj}} \\ \dot{\bar{\lambda}}_{0_{mnj}} &= -A_4 \frac{1 - X_{mnj}}{1 + \epsilon X} e^{\bar{\lambda}_{0_{mnj}} - A_2/\theta_{mnj}} + \\ & A_6 e^{-\bar{\lambda}_{0_{mnj}} - A_5/\theta_{mnj}} - \\ & A_7 e^{\bar{\lambda}_{0_{mnj}} - A_8/\theta_{mnj}} \\ \dot{\bar{I}}_{mnj} &= A_4 \frac{1 - X_{mnj}}{1 + \epsilon X_{mnj}} e^{\bar{\lambda}_{0_{mnj}} - A_2/\theta_{mnj}} - \\ & A_3 e^{-A_5/\theta_{mnj}} \end{aligned} \tag{18}$$

The continuity constraints between each element are:

$$\begin{aligned} X_{mj}^0 &= X_{m-1j}^0 + \Delta\tau_{m-1} \tau_{tr} \sum_{n=1}^{Nc} A_{nNc} \dot{X}_{m-1nj} \\ \bar{\lambda}_{0_{mj}}^0 &= \bar{\lambda}_{0_{m-1j}}^0 + \Delta\tau_{m-1} \tau_{tr} \sum_{n=1}^{Nc} A_{nNc} \dot{\bar{\lambda}}_{0_{m-1nj}} \\ \bar{I}_{mj}^0 &= \bar{I}_{m-1j}^0 + \Delta\tau_{m-1} \tau_{tr} \sum_{n=1}^{Nc} A_{nNc} \dot{\bar{I}}_{m-1nj} \end{aligned} \tag{19}$$

the starting values are $X_{1j}^0 = X_0, \;\; \bar{I}_{1j}^0 = 1, \;\; \bar{\lambda}_{0_{1j}}^0 = ln(\lambda_0)$, at $j = 1, 2, ..., N_z$. At this mathematical formulation, the notation for subscripts indices were used to represent points in time and position. For example, θ_{mkj} denotes the value of dimensionless temperature at the $m-th$ element, $k-th$ collocation point at the temporal mesh, and the $j-th$ position point for the sheet length. N_e is the number of finite elements, N_c is the number of collocation points, and N_z is the number of points for discretization on ζ-direction dimensionless.

4. RESULTS

The dynamic optimization model for PMMA given by Eqs. (6-11) was cast as a NLP problem by Eqs. (12-19), using the simultaneous approach. In this section, we present a comparison between dynamic optimal heating profiles against simpler operating policies. The analysis was done for two plastic sheet thicknesses, 3 mm and 6 mm. It should be stressed that "simpler" operating policies mean that the air temperature was always kept constant at 50 ^{o}C. The numerical results for the two cases presented here were obtained on a Pentium IV computer with 2.8 GHz clock speed and 512 KB RAM. Table 1 summarizes the parameters used for the two addressed cases. The NLP problem was solved using the CONOPT NLP solver embedded in the GAMS algebraic modelling system (Brooke *et al.*, 1998). In Figures 2 and 3, the results of using dynamic optimal heating profiles for two different thicknesses are compared against results obtained using the simpler operating policy. The last results were obtained by the numerical integration of Eqs. (6-10) imposing constant air temperature. Using the method of lines, the discretization scheme required five points along the position and 30 finite elements and two internal collocation points for the time coordinate for a plastic sheet of 3 mm thickness. The objective function is 0.8983, the optimal solution was obtained in 10.4 minutes CPU time. The desired temperature value along the plastic sheet is $50^{o}C$ or $\theta = 1$ (dimensionless temperature). The dynamic optimal air temperature profile to achieve the desired temperature value is depicted in Figure 2(a). As it can be seen from Figure 2(b), a good conversion is obtained, 92% at 50 minutes, while keeping constant the air temperature required 200 minutes to achieve the same conversion value. The zeroth living moment gradually increases with the polymerization temperature, but suddenly, a peak appears and it is related to the thermal effects due to contribution from the heat of reaction. At this moment, the air temperature fell down to remove the heat generated by the polymer reaction. In Figure 2(c) the polymer plastic sheet temperature profile is shown along the time at five points on the plastic sheet (see Figure 1). Note that, using the optimal heating pattern, the desired temperature value is achieved for all points in less time compared to the the simpler operating policy. As depicted in this Figure, due to the warm air flow direction, the temperature increases gradually along the plastic sheet; at $z = 0$ the dynamic temperature behavior is almost the same as the air temperature, but at $z = L$, the biggest peak temperature is located there, because the heat removal capacity is decreased. At Figure 3, the dynamic optimization responses are shown for a 6 mm thickness plastic sheet. The discretization scheme required 40 finite elements with two internal collocation points for the time coordinate. The objective function value is 2.8419. The optimal solution profiles were computed in around 18 minutes CPU time. It can be seen that the maximum value of the air temperature was higher than for the 3 mm thickness plastic sheet because the heat conduction resistance is two times greater. The dynamic behavior for the conversion and zeroth moment are similar to above case. The time value to get the same monomer conversion (92%) is around 40 minutes for the optimal operation and 123 minutes for the case where a constant heating profile was imposed. We think that the model can represent of reliable way the process of polymerization because the oven gets ready of the devices for to achieve the manipulation of the temperature. However, the main advantage of using dynamic optimal heating profiles is that undesired effects, such as bubble formation, might be avoided.

Table 1. Coefficients for PDE system

Coefficient	3 mm	6 mm
A_1	$6.1539 \text{x} 10^{8}$	$2.4616 \text{x} 10^{9}$
A_2	$6.7786 \text{x} 10^{0}$	$6.7786 \text{x} 10^{0}$
A_3	$1.3184 \text{x} 10^{18}$	$5.2736 \text{x} 10^{18}$
A_4	$-1.4626 \text{x} 10^{8}$	$-5.8503 \text{x} 10^{8}$
A_5	$4.76514 \text{x} 10^{1}$	$4.76514 \text{x} 10^{1}$
A_6	$1.3812 \text{x} 10^{18}$	$5.5248 \text{x} 10^{18}$
A_7	$1.2266 \text{x} 10^{11}$	$4.9064 \text{x} 10^{11}$
A_8	$1.0916 \text{x} 10^{0}$	$1.0916 \text{x} 10^{0}$
A_9	$4.4036 \text{x} 10^{8}$	$1.7614 \text{x} 10^{8}$
a	$5.555 \text{x} 10^{-4}$	$1.111 \text{x} 10^{-3}$
Bi	5.3366	10.6731

5. CONCLUSIONS

The computation of dynamic optimal heating profiles for the cell-cast MMA process was addressed. A comparison of model response, using two plastic sheet thicknesses, between the optimal heating profiles and an heuristic operating policy, consisting in keeping constant the air temperature, was done. It was found that the dynamic optimal heating profiles achieved the desired plastic sheet temperature in less time compared to the heuristic operating policy. Even when the thermal and kinetic effects of the addressed systems are complex, the computational load for finding optimal solutions was relatively modest. Therefore, real time control of the addressed polymerization system seems to be feasible and it will be addressed in a future work.

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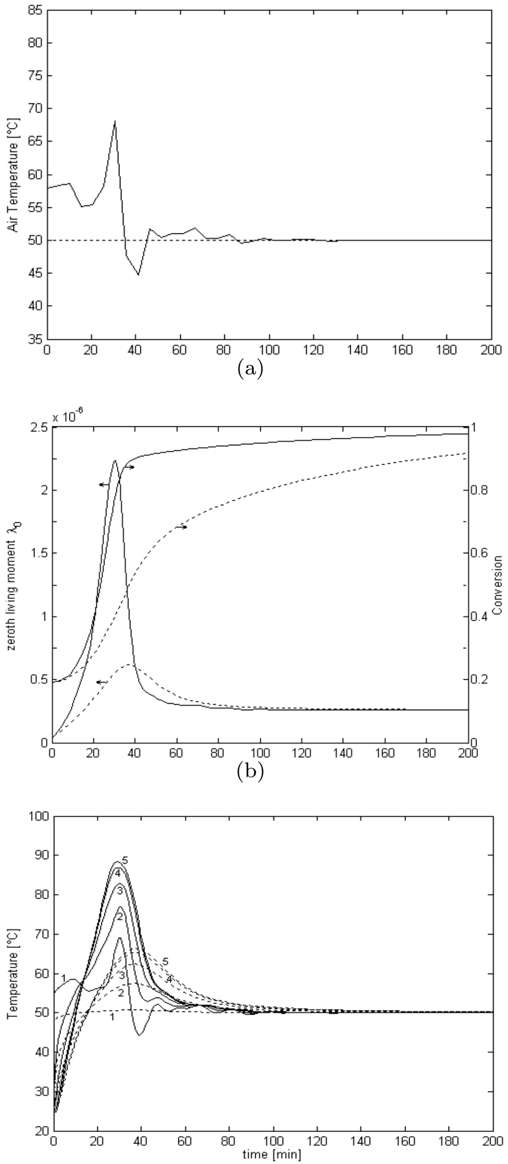
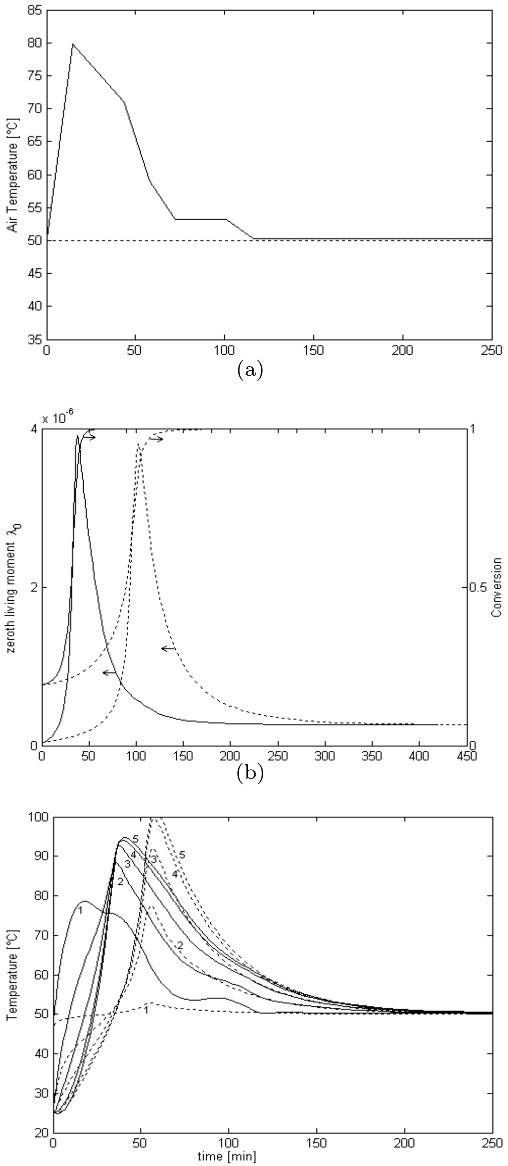


(a)

(b)

(c)

Fig. 2. The continuos line stands for dynamic optimization responses while the dashed line represents dynamic simulation results for (a)air temperature, (b)conversion and zeroth living moment and (c)temperature profile at [1]$z = 0$, [2]$z = L/4$, [3]$z = L/2$, [4]$z = 3L/4$, [5]$z = L$ (see Figure 1) for along the plastic sheet of 3 mm thickness.



(a)

(b)

(c)

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