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ORIGINAL PAPER

Optimal control of chemical birth and growth processes in a deterministic model

Ramón Escobedo · Luis A. Fernández

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Abstract An optimal control problem for a deterministic model of chemical birth and growth processes is studied. The cost functional takes into account the two main industrial competitive interests: to avoid low temperatures and to shorten the cooling time. Explicit expressions of the optimal controls are obtained by using an analytical approximation of the relation between the instantaneous amount of crystallized polymer and the total amount of cold injected into the sample until this instant of time. Numerical simulations are shown to illustrate the good agreement with the analytical expressions. It is surprising and remarkable that the explicit expressions of the optimal controls can be derived by minimizing an elementary function in one real variable.

Keywords Optimal control · Polymer crystallization · Numerical simulations

1 Introduction

Polymer crystallization can be described as a chemical process of formation of spatially ordered structures resulting from the combination of two main events: the nucleation of small molecules on an initial liquid melt and the growth of the nucleated crystals. The mathematical modelling of these birth and growth processes has been mainly inspired on the classical Avrami–Kolmogorov theory for isothermal crystallization

R. Escobedo (✉)
Departamento de Matemática Aplicada y CC. de la Computación, Universidad de Cantabria,
Av. de los Castros s/n, 39005 Santander, Spain
e-mail: escobedo@unican.es

L. A. Fernández
Departamento de Matemáticas, Estadística y Computación, Universidad de Cantabria,
Av. de los Castros s/n, 39005 Santander, Spain
e-mail: lafernandez@unican.es

[10,1], assuming a spherical shape of crystals when the growth rate is constant. In the general case, non-isothermal crystallization makes the growth rate to depend on both space and time through the temperature field, and the shape of a polymeric crystal is no longer a ball centered at the origin of growth [5]. The *degree of crystallinity*, defined as the mean volume fraction of the space occupied by crystals, is then used as a measure of the state of the crystallization process.

An overview on mathematical models of polymer processing is presented in Ref. [4], and non-isothermal polymer crystallization models have been studied in [11] and [2]. Most of them are stochastic models, accounting for the natural randomness of the nucleation process, both in space and time.

Recently, a deterministic model has been derived [5] from the Avrami–Kolmogorov stochastic model of interacting particles in the limit of *many and small particles*, in which the nucleation and growth processes are characterized by exponential rate functions of the non-homogenous temperature field. As these inherited expressions become unrealistic under deterministic conditions, the model has been modified to avoid the possibility that both nucleation and growth could happen at any room temperature. To do that, two threshold temperatures have been introduced as a cutoff of the respective nucleation and growth rates, above which no birth nor growth can take place. This idea was originally introduced in Ref. [8] and has been used in [6,7] by the same authors in the one dimensional case, having been extended later to rectangular samples [12].

The deterministic polymerization model was studied in Ref. [8] from the numerical viewpoint. There, a simplification (consisting on the linearization of the rate functions) was proposed which yields similar results in terms of the duration of the complete crystallization process. In the present paper the general deterministic model is used in order to obtain realistic and accurate numerical solutions of the polymer problem.

This family of models has been recently used in optimal control problems to approach the fundamental question of the industrial manufacturing of polymers: to determine the best cooling strategy. Three are the main requirements of the industry, one for the final material and two for the cooling process: (1) the material must be spatially homogenous to have good physical properties (elasticity, thermal and electrical conductivity,...), (2) the applied temperature must avoid excessively low values, which are very expensive to reach and maintain, and (3) the duration of the process must be as short as possible. The three industrial needs have been taken into account in the functional cost used in Ref. [9] by controlling the final degree of crystallinity and its spatial uniformity, together with the energy for generating the cooling temperature, expecting that if the degree of crystallinity is uniform and high, the complete crystallization should be close. The mechanical properties related to the morphology of the material are the aim of [3], where the contact-interface density is maximized and its variation is minimized, the duration of the crystallization process being not taken into account in the cost functional.

We would like to notice that the spatial homogeneity of the material does not depend exclusively upon the degree of crystallinity; as it has been shown in Ref. [8], different space segments of same length can have different times of crystallization, affecting to the physical properties of the material. This should be taken under consideration in future optimal control problems about crystallization processes.

Two main results are presented in this paper: first, a numerical relation between two important magnitudes of the polymer problem, the amount of crystallized polymer and the amount of cold injected in the sample, and second, the optimal values of the main industrial controls of the polymerization process, the cooling time and the applied temperature profile.

2 The birth and growth model

Following [8], we consider the system of equations for the degree of crystallinity $y(x,t)$ and the temperature $T(x,t)$:

$$y_t(x,t) = \left[Gy(x,t)\,(1-y(x,t)) + v_0 N\,(1-y(x,t))^2\right]\theta(T(x,t)), \tag{1}$$

$$T_t(x,t) = \sigma T_{xx}(x,t) + a_G G y(x,t)\,(1-y(x,t))\,\theta(T(x,t)), \tag{2}$$

for $(x,t) \in\ Q_\tau = (0,L)\times(0,\tau)$, where L is the length of the sample and τ is the final time, when the cooling process is stopped.

Equations (1) and (2) must be solved together with the following boundary conditions for all $t \in\ (0,\tau)$,

$$T(0,t) = u(t), \quad T_x(L,t) = 0, \tag{3}$$

and the following initial conditions for all $x \in (0,L)$:

$$y(x,0) = y_0 \in [0,1), \quad T(x,0) = T_0. \tag{4}$$

Nucleation and growth rate functions are $N\theta(T(x,t))$ and $G\theta(T(x,t))$, with

$$\theta(T) = \begin{cases} \exp(-\beta T) & \text{if } T < T_f, \\ 0 & \text{if } T \geq T_f. \end{cases} \tag{5}$$

The positive real constants $G, v_0, N, \sigma, a_G, \beta$ and T_f denote the growth factor, the initial mass, the nucleation factor, the heat diffusion coefficient, the non-isothermal factor, the nucleation and growth exponent and the critical phase transition temperature (from liquid to solid), respectively.

Figure 1 shows the solution of (1)–(5) when a constant cooling temperature $u = 40°\mathrm{C}$ is applied during a cooling time $\tau = 2.2\times10^4\,\mathrm{s}$ to a sample such that $T_f = 70°\mathrm{C}, \sigma = 0.002\,\mathrm{m}^2/\mathrm{s}, a_G = 2500°\mathrm{C}, N = 20\,\mathrm{s}^{-1}, G = 5\,\mathrm{s}^{-1}, v_0 = 0.01, \beta = 0.1(°\mathrm{C})^{-1}, L = 1\,\mathrm{m}, y_0 = 0$, and $T_0 = 100°\mathrm{C}$. The complete crystallization time is denoted by t_{cryst}, defined by $t > t_{\mathrm{cryst}} \Rightarrow 1 - y(L,t) < \varepsilon$, where $\varepsilon > 0$ is a small tolerance; here $t_{\mathrm{cryst}} = 1.95\times10^4$ s for $\varepsilon = 10^{-8}$.

As in other birth and growth processes, the solution exhibits an advancing front in the degree of crystallinity $y(x,t)$. The singularity of this model is that the front is more a band than a boundary layer, and that, under constant cooling

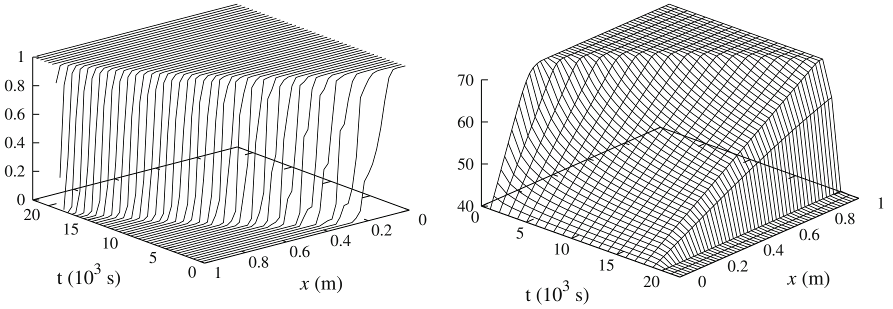

Fig. 1 Time evolution of the degree of crystallinity $y(x, t)$ (*left*) and the temperature field $T(x, t)$ (*right*) for $u(t) \equiv 40°\mathrm{C}$ and $\tau = 2.2 \times 10^4$ s. The complete crystallization is reached at $t_{\mathrm{cryst}} = 1.95 \times 10^4$ s. Note that the $x - t$ axes have been inverted to show clearly the evolution of the temperature field

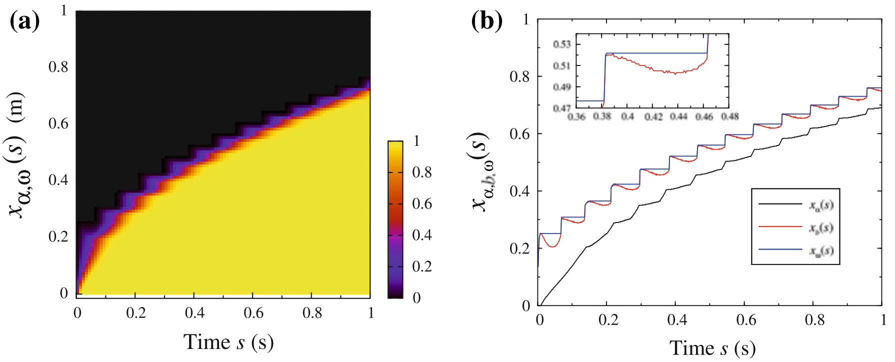

Fig. 2 **a** Color map of the crystallinity $y(x, s)$ for $u \equiv 40^{\mathrm{o}}\mathrm{C}$, $\tau = 10^4$ s. **b** Numerical characterization of the crystallinity distribution with abscissas $x_\alpha(t)$, $x_b(t)$ and $x_\omega(t)$ as defined in [8]. Time axis is normalized to $s \in [0, t/\tau]$

temperatures, the (averaged) velocity of the band is not constant but decreasing in time. See Fig. 2.

The solution can be characterized by means of three abscissas $x_\alpha(t) \leq x_b(t) \leq x_\omega(t)$, where $x_b(t)$ is the critical point where the temperature $T(x, t)$ reaches the phase transition threshold, $T(x_b(t), t) = T_f$, and $x_\alpha(t)$ and $x_\omega(t)$ divide the sample in three regions: an almost fully crystallized region $[0, x_\alpha(t)]$ in which $1 - \epsilon \leq y(x, t) \leq 1$, a region empty of nucleii $[x_\omega(t), L]$ where $y(x, t) = 0$ and nucleation has not started, and a third region $[x_\alpha(t), x_\omega(t)]$ in between where $0 < y(x, t) < 1 - \epsilon$ (ϵ small and positive); see [8].

Depending on the parameter values, another important feature can be observed in addition to the decreasing velocity: the advance of the band is not regular but *by jumps*, a feature that appears usually in nucleation and growth models where the size of the nucleii (the growth) acts on the probability of new nucleations. Particle models are especially suitable to analyze this kind of interaction between nucleii, which are not easy to reproduce with deterministic models, in spite of what the present model is able to exhibit this behavior by means of oscillations in the crystallinity and temperature distributions; see again Fig. 2. However, it is not our present aim to analyze this oscillatory behavior, and we will not take it into account in what follows.

3 The optimal control problem

The effectiveness of the cooling process depends mainly upon the total cooling time τ and the applied temperature profile $u(t)$ for $t \in (0, \tau]$. From the industrial viewpoint, it is well known that, in the one hand, the shorter cooling processes are cheaper and preferable, but in the other hand, low temperatures are expensive to reach and to maintain (in fact, the cost does not grow linearly but at an increasing rate as the temperature decreases). Hence, an equilibrium compromise must be adopted in each particular situation. Notice again that the spatial homogeneity of the resulting material is not taken into account.

The industrial strategy consists then in tuning the competitive control parameters τ and $u(t)$ to reach an agreement yielding the minimal value of the cost of the cooling process. We can then formulate an optimal control problem (CP), introducing two non negative weights σ_1 and σ_2 to balance the contribution of each term, and a set U_{ad} of admissible controls, where the condition for complete crystallization at time τ in the whole sample is written in an equivalent integral form:

$$\text{(CP)} \begin{cases} \text{Min} \;\; J(u, \tau) = \sigma_1 \displaystyle\int_0^{\tau} \left(T_f - u(t)\right)^2 \mathrm{d}t + \sigma_2 \tau, \\ (u, \tau) \in U_{\mathrm{ad}}, \end{cases}$$

$$U_{\mathrm{ad}} = \left\{ (u, \tau) \in L^2(0, \tau) \times [0, \infty) : u(t) \in [0, T_f], \text{ a.e.}, \int_0^L y(x, \tau)\, \mathrm{d}x = L \right\}. \tag{6}$$

The choice of $\sigma_{1,2}$ is based upon the rate σ_2/σ_1, which is a measure of the relative cost of the two competitive terms of the cost functional: to avoid low temperatures we will select σ_2/σ_1 small enough, and to shorten the cooling time we will pick up σ_2/σ_1 sufficiently large.

In order to solve the optimal control problem (CP), we have first obtained numerically an approximation of an important magnitude of the polymer problem, which is the amount of crystallized polymer at a given time, that has allowed us to derive explicit expressions of the optimal controls.

4 Main results

Let us define $P(t)$ and $Q(t)$ as the amount of crystallized polymer and the amount of cold injected into the sample at time t, respectively:

$$P(t) = \int_0^L y(x,t)\,\mathrm{d}x, \quad Q(t) = \int_0^t \big(T_f - u(r)\big)\,\mathrm{d}r. \tag{7}$$

4.1 Estimation of the quantity of crystallized polymer

Our first claim is that $P(t)$ does not depend on the history of the cooling process but on the amount of cold injected, that is, $P(t)$ depends upon $u(t)$ through $Q(t)$. We have obtained a numerical approximation of this relation:

$$P(t) \approx c\sqrt{Q(t)}, \tag{8}$$

for some positive constant c. Expression (8) is an important result by itself; the rest of Sect. 4.1 is devoted to illustrate that this relation compares very well with numerical simulations, and can then be used in Sect. 4.2 to solve the optimal control problem.

To illustrate the validity of the expression (8), we have tried six qualitatively different cooling strategies (u, τ) which inject the same amount of cold $Q_0 = 35 \times 10^4$ °Cs. We have used four $u(t)$-profiles for a fixed $\tau_1 = 1.5 \times 10^4$ s, denoted by (A–D) and depicted in Fig. 3, and two values of τ for a constant applied temperature u, denoted by (E,F):

$$\text{(A)}\quad u_A(t) = T_f\left[1 - \left(1 - \tfrac{t}{\tau_1}\right)^2\right], \quad \text{(B)}\quad u_B(t) = T_f\left[1 - \left(\frac{t}{\tau_1}\right)^2\right],$$

$$\text{(C)}\quad u_C(t) = \frac{2}{3}T_f, \qquad \text{(D)}\quad u_D(t) = \begin{cases} 0 & \text{in } [0, \tau_1/3] \\ T_f & \text{in } [\tau_1/3, \tau_1] \end{cases},$$

$$\text{(E)}\quad \tau_2 = 3 \times 10^4\,\text{s},\ u_E = \frac{5T_f}{6}, \qquad \text{(F)}\quad \tau_3 = 0.6 \times 10^4\,\text{s},\ u_F = \frac{T_f}{6}.$$

Figure 4 shows the time evolution of the amount of crystallized polymer $P(t)$ with respect to the amount of cold injected $Q(t)$ for the six strategies, together with the approximation (8) for $c \approx 1.33 \times 10^{-3}\,\text{m/(s °C)}^{-1/2}$.

The constant c is obtained numerically to fit the curves of $P(Q)$ and is obviously the same for the six strategies. Exhaustive numerical simulations show that c remains constant if a_G, σ, v_0, N, G and β are not changed.

The agreement between the six strategies and the approximation (8) is very good, as shown in Fig. 4 and by the two error estimates we have used, the L_2-norm error $E_1(\tau)$ and the maximum relative error $E_2(\tau)$, defined as

$$E_1(\tau) = \frac{1}{\tau}\int_0^\tau \left|P(t) - c\sqrt{Q(t)}\right|^2 \mathrm{d}t, \quad E_2(\tau) = \max_{t\in[t^*,\tau]}\left(\frac{|P(t) - c\sqrt{Q(t)}|}{P(t)}\right),$$

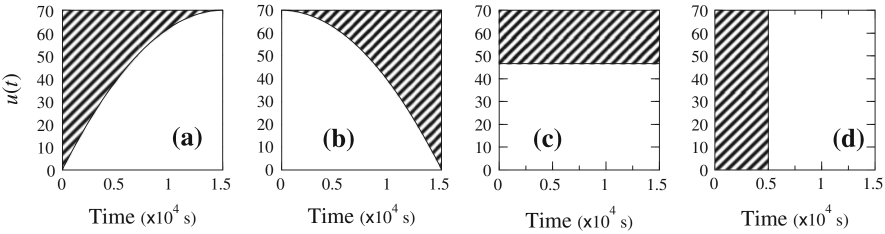

Fig. 3 Four cooling strategies $u(t)$ for $T_f = 70°\text{C}$ and $\tau_1 = 1.5 \times 10^4$ s, with a total amount of injected cold $Q_0 = Q(\tau_1) = \tau_1 T_f/3 = 35 \times 10^{4}°\text{Cs}$

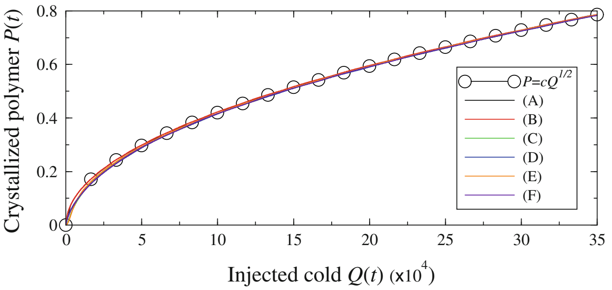

Fig. 4 Time-parametric representation of the amount of crystallized polymer $P(t)$ versus the amount of cold injected $Q(t)$ for the six strategies (A)–(F), denoted by the six overlaping *solid lines*, showing the complete agreement between them and with the relation $P \approx c\sqrt{Q}$ with $c \approx 1.33 \times 10^{-3}$ (*circles*)

where $t^* \approx 2 \times 10^3$ s is used to filter the effects of the initial numerical transient, whose orders of magnitude are $E_1(\tau_1) \approx 10^{-5}$ and $E_2(\tau_1) \approx 10^{-2}$.

4.2 Optimal controls

The expression (8) is now used to solve the optimal control problem. If (u, τ) is an admissible control, the complete crystallization is reached at time τ, so $P(\tau) = L$ and (8) yields $L \approx c\sqrt{Q(\tau)}$. Then, $Q(\tau) \approx L^2/c^2$ and the following expressions can be considered equivalent:

$$P(\tau) = \int_0^L y(x, \tau)\,\mathrm{d}x = L \quad \text{and} \quad Q(\tau) = \int_0^\tau \big(T_f - u(r)\big)\,\mathrm{d}r = \frac{L^2}{c^2}. \tag{9}$$

Then, we can reformulate (CP) as the following optimization problem (OP):

$$\text{(OP)} \begin{cases} \text{Min } \; J(u, \tau) = \sigma_1 \displaystyle\int_0^\tau \big(T_f - u(t)\big)^2\,\mathrm{d}t + \sigma_2 \tau, \\ (u, \tau) \in V_{\text{ad}}, \quad \text{where} \end{cases} \tag{10}$$

$$V_{\text{ad}} = \left\{ (u,\tau) \in L^2(0,\tau) \times [0,\infty) : u(t) \in [0, T_f],\ \text{a.e.},\ Q(\tau) = \frac{L^2}{c^2} \right\}. \quad (11)$$

The admissibility condition $(u,\tau) \in V_{\text{ad}}$ implies a lower bound for τ, $\tau \geq \hat{\tau}$, where $\hat{\tau} = L^2/(T_f c^2)$. On the other hand, Hölder's inequality yields

$$\int_0^\tau (T_f - u(r))^2 \mathrm{d}r \geq \frac{1}{\tau}\left(\frac{L^2}{c^2}\right)^2, \quad \forall (u,\tau) \in V_{\text{ad}}, \quad (12)$$

and consequently, $J(u,\tau) \geq (L^2/c^2)^2\sigma_1/\tau + \sigma_2\tau, \forall(u,\tau) \in V_{\text{ad}}$. The following auxiliary real function ψ has a unique global minimum at $\overline{\tau}$ in $[\hat{\tau},\infty)$,

$$\psi(\tau) = \frac{\sigma_1}{\tau}\left(\frac{L^2}{c^2}\right)^2 + \sigma_2\tau, \quad \tau \in [\hat{\tau},\infty), \quad (13)$$

where $\overline{\tau} = \hat{\tau}$ if $\sigma_2 > \sigma_1 T_f^2$, and $\overline{\tau} = L^2\sqrt{\sigma_1/\sigma_2}/c^2$ if $\sigma_2 \leq \sigma_1 T_f^2$. Therefore, $\sigma_2 > \sigma_1 T_f^2$ implies that $J(u,\tau) \geq \psi(\tau) \geq \psi(\hat{\tau}) = J(0,\hat{\tau}), \forall(u,\tau) \in V_{\text{ad}}$, and $\sigma_2 \leq \sigma_1 T_f^2$ implies that $J(u,\tau) \geq \psi(\tau) \geq \psi(\overline{\tau}) = J(T_f - \sqrt{\sigma_2/\sigma_1}, \overline{\tau})$, $\forall(u,\tau) \in V_{\text{ad}}$. Previous argumentations allow us to derive the following theorem:

Theorem 1 *Assume that* $\sigma_1 \in [0,\infty)$ *and* $\sigma_2 \in (0,+\infty)$. *Then,*

(*a*) *If* $\sigma_2 > \sigma_1 T_f^2$, *the unique solution of (OP) is given by*

$$\overline{u}(t) \equiv 0, \quad \overline{\tau} = \frac{L^2}{c^2 T_f}. \quad (14)$$

(*b*) *If* $\sigma_2 \leq \sigma_1 T_f^2$, *the unique solution of (OP) is given by*

$$\overline{u}(t) \equiv T_f - \sqrt{\frac{\sigma_2}{\sigma_1}}, \qquad \overline{\tau} = \sqrt{\frac{\sigma_1}{\sigma_2}}\frac{L^2}{c^2}. \quad (15)$$

The following remarks are in order:

- In the case $\sigma_2 > \sigma_1 T_f^2$, the main interest is to shorten the crystallization process, without taking care about the cost of reducing the temperature to very low values. It is therefore easy to see that the optimal strategy consists in applying the lowest possible temperature ($\bar{u} \equiv 0$) during the whole process.
- In the case $\sigma_2 \leq \sigma_1 T_f^2$, the goal is to obtain a complete crystallization retaining the applied temperature close to the cheapest value T_f, even if the duration of the process increases. This is shown by the expressions (15): as σ_2/σ_1 decreases to zero, we see that $\overline{u} \to T_f$ and $\overline{\tau} \to +\infty$; see Fig. 5.
- The effect of small variations in σ_2/σ_1 over the optimal controls (normalized with their corresponding typical values) can be seen in Fig. 5. The figure shows two different regimes: in $\sigma_2/\sigma_1 \in [0,1]$ (inset of the figure), the cooling time has a

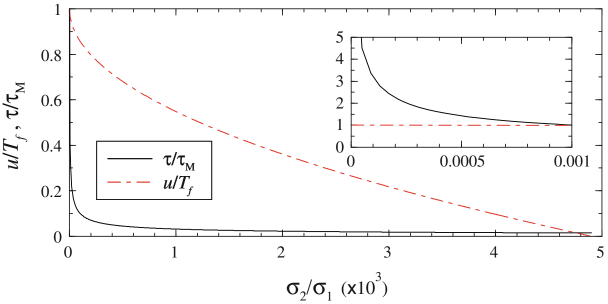

Fig. 5 Variation of the optimal controls $\overline{u}$ and $\overline{\tau}$ with respect to σ_2/σ_1 in $(0, 5 \times 10^3]$, normalized with T_f and τ_M, respectively. The value of $\tau_M \approx 5.6 \times 10^5$s corresponds to $\sigma_2 = \sigma_1$. Note that $T_f^2 = 4.9 \times 10^3 °C^2$ is the upper bound of σ_2/σ_1 corresponding to the condition $\overline{u} \geq 0$

higher relative sensitivity to small changes in σ_2/σ_1 than the applied temperature, whereas the situation is the opposite in $[1, 4.9 \times 10^3]$.

We have solved the optimization problem with $\sigma_2/\sigma_1 = 900$, getting $\overline{u} = 40°$C and $\overline{\tau} = 1.88 \times 10^4$ s, which compares very well with the complete crystallization time $t_{\text{cryst}} = 1.95 \times 10^4$ s that was obtained applying the temperature $u(t) \equiv 40°$C to the model (1)–(5) for the given values of the parameters. For these data, the relative error is in accordance with the previous estimations of $E_2 : |(t_{\text{cryst}} - \overline{\tau})/t_{\text{cryst}}| \approx 3.6 \times 10^{-2}$.

5 Conclusion

We have analyzed an optimal control problem for a deterministic model of chemical birth and growth processes in polymer crystallization, where the cost functional takes into account two (of the three) main industrial interests: to avoid low temperatures and to shorten the cooling time.

We have shown numerically that the amount of crystallized polymer $P(t)$ is proportional to the square root of the amount of cold injected into the sample $Q(t)$, obtaining the constant of proportionality for a given set of values of the physical parameters.

The important result (8) of the polymer problem has allowed us to obtain explicit expressions of the optimal controls, namely, the duration of the cooling process until complete crystallization $\bar{\tau}$ and the temperature applied to the sample $\bar{u}(t)$. Surprisingly, the optimal cooling strategy is constant, something a priori not expected because the velocity of the crystallization front is not constant. Despite of the complicated appearance of the initial optimal control problem (CP) under study, it is surprising to discover that the optimal control values can be derived through the minimization of an elementary function in one real variable. This means a huge reduction in the complexity of the problem. The good agreement of our results has been checked with numerical simulations and the L_2-norm and relative error estimates.

Future work should take into account the spatial homogeneity of the resulting material by controlling the crystallization speed, in order to avoid the characteristic oscillations of the temperature at the crystallization front.

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