A Design Sensitivity Analysis for Crystallinity Control

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Abstract

This paper addresses implementation of a design sensitivity analysis to control crystallinity distribution in semi-crystalline polymers during a film casting process since their properties are shaped by crystallinity distribution during the process. The design sensitivity analysis is used as a control algorithm to determine sub-optimal process inputs to establish the desired crystallinity distribution for the semi-crystalline polymers during the film extrusion process. The simulation result demonstrates the effectiveness of the control algorithm which can efficiently determine a set of process inputs that generate a crystallinity distribution close to the desired one.

1. Introduction

Semi-crystalline polymers are widely used in a variety of applications ranging from household products to high strength structural composites for the aerospace industry. This is because of their attractive properties such as light-weight, high strength and ease of formability. To form a raw polymer in to a usable configuration, quality specifications for the finished product are now determined from the dimensional and structural variables. These variables need to be controlled tightly in order to provide good quality products.

Like chain orientation, crystallinity is one of the most important structural variables since it dictates the properties in the polymeric products. Up to now, control of semi-crystalline polymers has emphasized only product dimension such as thickness, shape and so on. Consequently, dimensional control alone is relatively crude and can result in variations in structural variables. Therefore, crystallinity control during the polymer process is a realistic solution to improve the total quality of semi-crystalline products.

For film extrusion, control of dimensional variables have been studied by many researchers [1], [2]. However, variables that are associated with material properties are not generally used as the process control variables. Crystallinity is an interesting structural variable in semi-crystalline polymer. Properties of polymer depend upon not only the amount of crystallinity but also upon the crystallinity morphology [3]. For example, a lot of small spherulites indicates tight packing. This yields good mechanical properties [4]. During the film extrusion, the process inputs will influence the dynamics of crystallization which affect the structure of film polymer[5]. Therefore, the process controller implements the process inputs in order to control crystallinity distribution inside polymer film during the process.

2. Problem formulation

Figure 1 illustrates the casting film extrusion process.

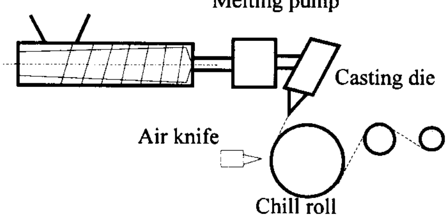

Figure 1 Sketch of film extrusion

The semi-crystalline polymer is melted in the extruder and extruded through the casting die. It is cooled over the chill roll. An air knife is sometimes added to make a good contact between the polymer film and the chill roll. It is found that crystallization takes place on the chill roll[6]. The mathematical model of casting film polymer process is described by kinetic theory and the principle of energy conservation[5], [7], [8]. Here, stress induced crystallization [9] and induction time [10] are ignored. The crystallinity distribution inside the casting film polymer over the chill roll for non-isothermal crystallization can be determined as follows.

The energy equation is written as:

$$\rho v_x\left(C_p\frac{\partial T}{\partial x}-H_\infty\frac{\partial \alpha}{\partial x}\right)=k\frac{\partial^2 T}{\partial z^2} \tag{1}$$

where T is the temperature, α is crystallinity, t is the time, x and z are the coordinates corresponding to the flow direction and film thickness respectively, ρ is the density, C_p is the constant pressure specific heat, H_∞ is the actual specific enthalpy of crystallization and k is the thermal conductivity and v_x is the velocity of polymer film.

The temperature of the surface which is cooled by chill roll T_{sc} is determined by the following equation[11]:

$$T_{sc}=\frac{b_r T_r+b_p T_p}{b_r+b_p} \tag{2}$$

where b_r and b_p are the thermal effusivities of metallic roll surface and of the polymer respectively, T_p is the mean temperature of the polymer at abscissa x and T_r is the temperature of the chill roll.

The other side is cooled by air and air knife convection:

$$-k\frac{\partial T}{\partial z}=h(T_{sa}-T_{a,ak}) \tag{3}$$

where T_{sa} and $T_{a,ak}$ are respectively the temperature of the surface of the film and of the air or air knife.

The local convection heat transfer coefficient between air and polymer is calculated by [12]:

$$h=0.402\frac{k_a}{x}\sqrt{\mathrm{Re}}\frac{\mathrm{Pr}^{1/3}}{\left(1+\left(\frac{0.0336}{\mathrm{Pr}}\right)^{2/3}\right)^{1/4}}+0.45 \tag{4}$$

where k_a is the conductivity of air, Pr is the Prandtl Number and Re is the Reynolds Number. x is the distance between the first point of contact polymer-roll and the abscissa x. Between the air knife and polymer film, the convection heat transfer coefficient of a single nozzle is determined from the Nusselt Number Nu [13]:

$$Nu=\frac{3.06}{x_e/W_e+H/W_e+2.78}\mathrm{Re}^s\,\mathrm{Pr}^{0.42} \tag{5}$$

$$s=0.695-\left[\left(\frac{x_e}{2W_e}\right)+\left(\frac{H}{2W_e}\right)^{1.33}+3.06\right]^{-1} \tag{6}$$

where W_e is the width of the slot nozzle, H is the height of the slot-nozzle exit and x_e is distance from the center of the single slot.

The non-isothermal crystallization kinetics can be expressed as:

$$\alpha(t)=1-\exp\left(-E(T)\right) \tag{7}$$

$$E(T) = \left[\sum_{\substack{j=1 \\ j \neq k}}^{j=p+1} \frac{1}{|\dot{T}_j|} \left| Z^{\frac{1}{n}}(T_j) - Z^{\frac{1}{n}}(T_{j-1}) \right| - \frac{\partial Z^{\frac{1}{n}}(T_K)}{\partial T}(t_k - t_{k-1}) \right]^n \tag{8}$$

$$\dot{T}_j = \frac{T_j - T_{j-1}}{t_j - t_{j-1}} \tag{9}$$

$$Z(T) = \exp(C_1 T + C_2) \tag{10}$$

where n is the Avrami index and T_{p+1} is the temperature at the current time. It is assumed that there are $p+1$ constant cooling rate segments. If the cooling rate is zero during any time interval such as $[t_k, t_{k-1}]$, then the second term of summation in [8] is used to determine E during this time. The constants C_1 and C_2 are determined by the differential scanning calorimetry (DSC) for a given material

3. Sensitivity Analysis

Second order sufficient condition [14] for optimization algorithm is stated by the following.

Proposition Let Ω be a subset of n-dimensional space X^n and $f \in C''(\Omega)$second order derivative continuous function in Ω.
If $[\vec{\nabla} f(x^*)] = 0$ and $H_f(x^*) = [\nabla^2 f(x^*)]$ is positive definite Hessian matrix, then x^* is a relative minimum point of f.

Now, consider a scalar performance index P which is a function of the process input vector u. According to second-order sufficient condition, P is minimized by optimal inputs u^* when

$$\vec{\nabla} P(u^*) = 0 \tag{11}$$

and

$$H_p(u^*) > 0 \tag{12}$$

Note that if the conditions in equation (11) and (12) are satisfied, the obtained solutions are the sub-optimal process inputs.

In numerical optimization, the Quasi-Newton's method is considered

$$u^{k+1} = u^k + \mu^k B_P(u^k) \vec{\nabla} P(u^k) \tag{13}$$

where the matrix B_P is the approximation to the inverse of the Hessian matrix, μ^k is the step length. Approximation to inverse of Hessian matrix is determined by BFGS (Broyden-Fletcher-Goldfab-Shanno) method. The step length parameter is evaluated by Fibonacci & Golden section line search method. Note that, to calculate B_f, one needs the first-order derivative. BFGS method and Fibononacci & Golden section line search method are described in [15].

To update the current process inputs u^k in equation [13] needs to be implemented by using the information of the gradient direction vector at iteration k. The gradient direction vector can be obtained by applying the design sensitivity method. To demonstrate the fundamental idea, a steady state linear elliptic system of crystal growth process[16] is presented in order to exemplify the concepts of the design sensitivity analysis.

After the numerical finite element approach is implemented, the system is described by:

$$K(u)T(u) = B(u) \tag{14}$$

where K, B, and T are the stiffness matrix due to conductive heat, forcing vector due to boundary conditions and the temperature vector respectively.

Let us propose a scalar performance index defined as:

$$P(u) = f(T(u), u) \tag{15}$$

The sensitivity of performance index to each process inputs can be obtained by using the derivative chain rule to equation (5).

$$\frac{dP}{du_i} = \frac{\partial f}{\partial T}^T \frac{dT}{du_i} + \frac{\partial f}{\partial u_i} \tag{16}$$

for i=1,2,3,...; components in u.

The main difficulty in evaluating the sensitivity is the fact that $\frac{dT}{du_i}$ is unknown. This term can be evaluated by differentiating the system in equation (14) with respect to the individual process inputs, i.e.,

$$\frac{dK(u)}{du_i}T(u)+K(u)\frac{dT(u)}{du_i}=\frac{dB(u)}{du_i} \quad (17)$$

which can be rearranged to form:

$$K(u)\frac{dT(u)}{du_i}=\frac{dB(u)}{du_i}-\frac{dK(u)}{du_i}T(u) \quad (18)$$

Now, $\frac{dT}{du_i}$ can be determined by solving (18) by the convention numerical scheme. This appoach will be implemented to control the crystallinity distribution in the polymer extrusion process which is described in the next section.

4. Control design algorithm

Controller design problem is to determine the process inputs such as extruder temperature T_0, chill roll temperature T_r, air knife temperature T_{ak} and air temperature T_a to establish the desired crystallinity distribution. To achieve this objective, the performance index P is defined in terms of the error difference between the actual crystallinity distribution and the desired one, for example, as:

$$P(u)=\frac{1}{2}\left(\alpha(u)-\alpha_d\right)^T w\left(\alpha(u)-\alpha_d\right) \quad (19)$$

where u is a set of the process inputs, α is the actual crystallinity distribution, α_d is the desired crystallinity distribution and w is the weighting matrix.

The performance index is minimized by applying the design sensitivity analysis[17]. This minimization means the controller determines the process inputs that generate the actual crystallinity distribution close to the desired one.

5. Simulation results

Figure 2 shows the performance of the controller obtained by the proposed control scheme.

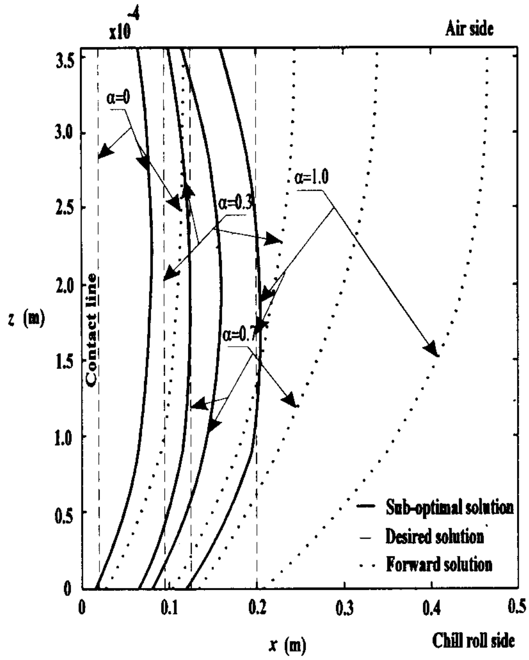

Figure 2. Performance of the proposed control algorithm.

Forward solution is presented for crystallinity distribution corresponding to arbitrary process inputs T_r, T_a, T_{ak} and T_0. After the desired crystallinity distribution is specified by uniform crystallinity across the flow direction, the controller determines the new process inputs and the sub-optimal solution shows the crystallinity distribution which is close to the desired one according to Table 1. The performance index P reduces from 38.12 to 3.70.

Table 1 Performance index

Process inputs	Forward solution	Sub-optimal solution
T_r (K)	373	370.24
T_a (K)	293	292.99
T_{ak} (K)	Not used	282.77
T_0 (K)	553	552.87
P	38.12	3.70

6. Conclusion

Control of crystallinity distribution during sheet extrusion is important in practice because crystallinity affects the final material morphology which determines the properties of the finished product. As the simulation results demonstrate, the control technique can determine the process inputs in order to generate the crystallinity distribution close to the desired one.

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8. Appendix

b_p: Thermal effusivity of polymer, b_p= 640.88 (J $s^{-1/2}$ K^{-1} m^{-2}).
b_r: Thermal effusivity of chill roll, b_r= 7520 (J $s^{-1/2}$ K^{-1} m^{-2}).
C_p: Constant pressure specific heat, C_p=2550 (J kg^{-1} K^{-1}).
e: Thickness of the polymer sheet, e=3.56×10^{-4} (m).
H: Height of the slot air-knife jet to material on the chill roll, H=0.1 (m).
H_∞: Actual specific enthalpy of crystallization, H_∞= 87800 (J kg^{-1}).
k: Conduction heat transfer coefficient of polymer, k= 0.21 (J m^{-1} s^{-1} K^{-1}).
n: Avrami index, n=3.
T_0^m: Equilibrium melting temperature, T_0^m =481 (K).
v_x: Polymer velocity, v_x=0.064 (m s^{-1}).
V_e: The velocity of air jet at slot nozzle exit, V_e=30 (m s^{-1}).
W_e: The width of a slot nozzle, W_e=0.02 (m).
W: The weight matrix, W is identity matrix.
$Z(T)$: Parameter Z, $Z(T)$=exp(-0.73591T+278.749) (K^3s^{-3}).
ρ: Density, ρ=767 (kg m^{-3}).