Dynamic control of crystallinity in polymer film casting process

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Abstract

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This paper presents an approach for dynamic control of crystallinity in polymer film casting process. As known, the transients of crystallization dictate the microstructures of semi-crystalline polymer during solidification. In turn, the properties of finished products can be determined by adjustable variables in polymer film casting process such as temperature of chill roll. In this work, an experimental model of the solidification in film casting process is derived by a system identification technique. This model is used to design a digital feedback controller including a state estimator. The simulation results show the effectiveness of the proposed control technique on an extruded film.

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บทคัดย่อ

ธนัญชัย ลีภักดิ์ปรีดา

การควบคุมเชิงพลวัตของความเป็นผลึกในกระบวนการขึ้นรูปแผ่นฟิล์มพอลิเมอร์

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บทความนี้กล่าวถึงวิธีการแนวทางหนึ่งสำหรับการควบคุมเชิงพลวัตของความเป็นผลึกในกระบวนการขึ้นรูป
แผ่นฟิล์มพอลิเมอร์ เป็นที่ทราบกันว่า การเปลี่ยนแปลงพลวัตในการตกผลึกจะเป็นตัวกำหนดการเรียงและขนาด
โครงสร้างของพอลิเมอร์ระหว่างการเย็นตัวขึ้นรูป สิ่งที่ตามมาก็คือคุณสมบัติของผลิตภัณฑ์สามารถถูกกำหนดได้จาก
ตัวแปรที่ปรับค่าได้ในการขึ้นรูปแผ่นฟิล์มพอลิเมอร์ เป็นต้นว่า อุณหภูมิของตัวล้อหล่อเย็น ในงานนี้ แบบจำลองทาง
คณิตศาสตร์เชิงการทดลองของในกระบวนการเย็นตัวขึ้นรูปถูกหาจากเทคนิคการระบุระบบ   แบบจำลองทาง
คณิตศาสตร์ที่ได้นี้จะถูกนำไปใช้ออกแบบตัวควบคุมสเตตย้อนกลับร่วมด้วยตัวคำนวณสเตท   ผลการจำลองแสดงให้
เห็นประสิทธิภาพของเทคนิคการควบคุมที่นำเสนอบนแผ่นฟิล์ม

ภาควิชาระบบการผลิตและวิศวกรรมเครื่องกล สถาบันเทคโนโลยีนานาชาติสิรินธร มหาวิทยาลัยธรรมศาสตร์ ศูนย์รังสิต ตู้ปณ. 22
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Conventionally, a film casting process is used to produce thin membranes of thermoplastic polymers. Figure 1 illustrates the typical film casting extrusion process. Plastic granules or pellets are fed into a hopper attached to the extrusion machine. From the hopper, the plastic falls through a hole   at the top of the extruder onto the extrusion screw. This screw conveys the plastic forward into a heated region until it exits through a melting pump in the end of the extruder barrel to which a casting die has been attached. The die imparts a shape to the molten plastic film that is cooled by a chill roll in order to solidify the plastic, thus retaining the shape. Normally, an air knife is added to obtain a good contact between the plastic film

Figure 1. Sketch of film extrusion process.

and the chill roll. In the process, adjustable process inputs such as take-up speed, melt temperature, chill roll temperature are to be regulated to maintain desired process outputs such as thickness, shape, and so forth. Up to now, those dimensional variables have mostly been considered the process outputs in polymer processing. To achieve total quality of the finished polymer products, some structural variables, e.g. crystallinity, are also to be controlled in the process. This is because variations in the structural variables dictate the properties of the polymeric films. It is found that properties of the finished polymer product are dictated by the dynamics of crystallinity (Billon *et al.*, 1991 ; and Janeschitz-Kriegl, 1992).

In this work, a modern control for polymer film casting process is to be implemented to obtain such tight product specifications. In Batur and Leephakpreeda (1996b) the steady control of crystallinity distribution was implemented by using the sensitivity design analysis where the boundary conditions of the process that correspond to a given desired crystallinity distribution were optimally determined. If there were no disturbances acting on the process, application of these boundary conditions under the open loop control kept the crystallinity at the desired levels. However, in reality,
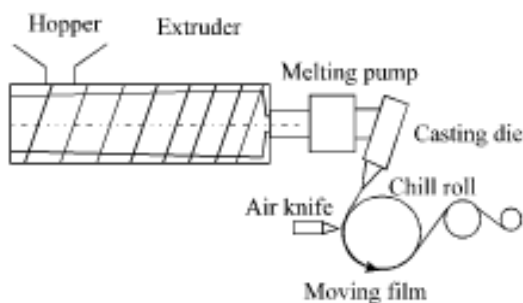
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a variety of random disturbances can change the operating conditions. This paper presents an alternative methodology for dynamic control of the transients of crystallinity. The system identification technique (Soderstrom and Stoica 1989) is applied in order to obtain a transfer function between the process inputs and the resulting crystallinity. With experimental data driven model, the obtained transfer function can then provide a knowledge of transient behavior for designing a digital feedback control system in order to control the crystallinity of the extruded product on-line.

This paper is organized as follows. The mathematical model, which is regarded as a real process, describes the dynamics of crystallinity over the chill roll in section 1. In the section 2, the system identification technique is applied to obtain the transfer function of the process. Section 3 presents the application of digital controller based the obtained transfer function. Finally, conclusion is stated.

1. Crystallization over the chill roll

A mathematical model is established for crystallization of a polymer film over a chill roll (Cotto *et. al.*, 1989). According to Figure 1 a semi-crystalline polymer is melted in the extruder and extruded though the casting die. It is cooled over the chill roll. An air knife is added to make a good contact between the polymer film and the chill roll. The mathematical model for this polymer film casting process can be described by the principle of energy conservation and the crystallization kinetic model (Duffo *et al.*, 1991; Avrami, 1939 ; and Nakamura *et al.* 1972). The crystallinity distribution inside a casting polymer film over the chill roll for non-isothermal crystallization can be determined as follows.

The energy equation is written as:

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

(1)

where T is the temperature, $\alpha \in [0,1]$ is relative crystallinity, t is the time, x and z are the coordinates corresponding to the flow direction and film thickness respectively, ρ is the polymer density, C_p is the constant pressure specific heat, H_∞ is the specific enthalpy of crystallization for 100 % crystallinity, k is the thermal conductivity and v_x is the velocity of polymer film.

The temperature of the surface which is cooled by the chill roll T_{sc} is determined by the following equation (Billon *et al.* 1991) :

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

(2)

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

The other side is cooled by air convection or air knife convention:

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

(3)

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

The local convection heat transfer coefficient between air and polymer (Churchill and Ozoe, 1973) is calculated by:

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

(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 and the abscissa x. Between the air knife and polymer film, the convective heat transfer coefficient of a single nozzle is determined from the Nusselt Number Nu (Incropera and Dewitt, 1990):

$$Nu = \frac{3.06}{x_e / W_e + H / W_e + 2.78} \text{Re}^s \text{Pr}^{0.42}$$

(5)

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

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 (Isayes *et al*. 1994):

$$\frac{d\alpha}{dt} = nK(T)(1-\alpha)[-\ln(1-\alpha)]^{(n-1)/n} \quad (7)$$

$$K(T) = \ln 2^{1/2} \left(\frac{1}{t_{\frac{1}{2}}} \right) \exp\left(-\frac{U^*/R}{(T-T_\infty)} \right) \exp\left(-\frac{K_g}{T\Delta Tf} \right) (8)$$

$$\Delta T = T_m^0 - T \quad (9)$$

$$f = \frac{2T}{(T+T_m^0)} \quad (10)$$

$$T_\infty = T_g - 30 \quad (11)$$

where n is the Avrami index, T_m^0 is the equilibrium melting temperature, $t_{\frac{1}{2}}$ is the isothermal crystallization half time, U^* is the activation energy for molecular trasport across the melt/solid interface, R is the universal gas constant, K_g is the nucleation exponent and T_g is the glass transition temperature. The induction time can be determined by:

$$\bar{t} = \int_0^{t_1} \frac{1}{b(T_m^0 - T)^{-\gamma}} d\tau = 1 \quad (12)$$

where $\bar{t}$ is the dimensionless induction time index, γ and b are the constant values. As soon as this time index reaches unity, the corresponding length of delay time t_I is the non-isothermal induction time. It should be noted that the integration of Eq. (12) starts when the temperature of polymer is below the equilibrium melting temperature.

2. System Identification

The non-linear model which consists of energy equations in Eqs (1)-(6) and crystallization kinetics in Eqs. (7)-(11) including the non-isothermal induction time in Eq. (12) is applied to describe the film casting process. The system identification technique (Soderstrom and Stoica 1989) is performed at $x = 25$ mm and $z = 3.56$ mm, and this position is illustrated in Figure 2. The parametric values for polypropylene are given in the Appendix. Initially, the chill roll temperature is 350 K and the air temperature is 293 K. Figure 3 shows the change of crystallinity when the chill roll temperature is changed following a Pseudo Random Binary Sequence (PRBS) of magnitude 1K around the operating condition of 350. The clock period of the PRBS signal is 5 seconds. From simulation,
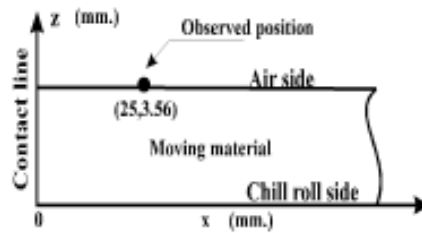

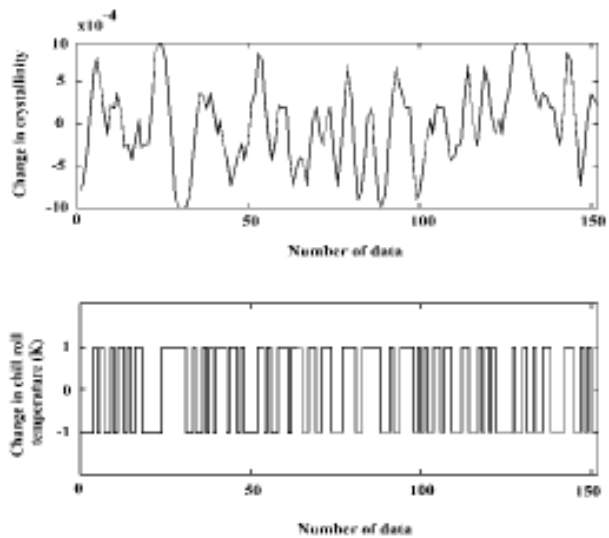**Figure 2. Model of crystallinity in a given position.**

Figure 3. Changes in crystallinity following PRBS changes in chill roll temperature.

total 150 input/output data are obtained. The first 100 data are selected for building a model. The remaining 50 data are used to validate the model. The Box-Jenkins model (Eykhoff, 1974) is used in Eq. (13) for representing the linear model of the polymer film casting process around an operating condition. The numerical optimization algorithm is available in System Identification Toolbox of MATLAB (Ljung, 1993). The transfer function can be expressed as:

$$\alpha(z) = \frac{H(z)}{G(z)} T_r(z) \qquad (13)$$

with the denominator

$$G(z) = 1 + g_1 z^{-1} \qquad (14)$$

and the numerator of the transfer function as

$$H(z) = h_0 + h_1 z^{-1} + ... + h_5 z^{-5} \qquad (15)$$

Corresponding to the fitting of those 100 input/output data, the numerical values of model parameters in Eqs. (14) and (15) are shown in Table 1. Figure 4 shows how well the output of model fits the independent remaining 50 output data which are not used to generate the model. In this procedure, the remaining 50 input data was fed to the identified model in Eq. (13). The dotted line shows the output data that are obtained from the identified model whereas the solid line presents the remaining 50 output data. The dynamic response of outputs from the model in Eq. (13) is fitted to

Table 1. Numerical values obtained by the process identification technique.

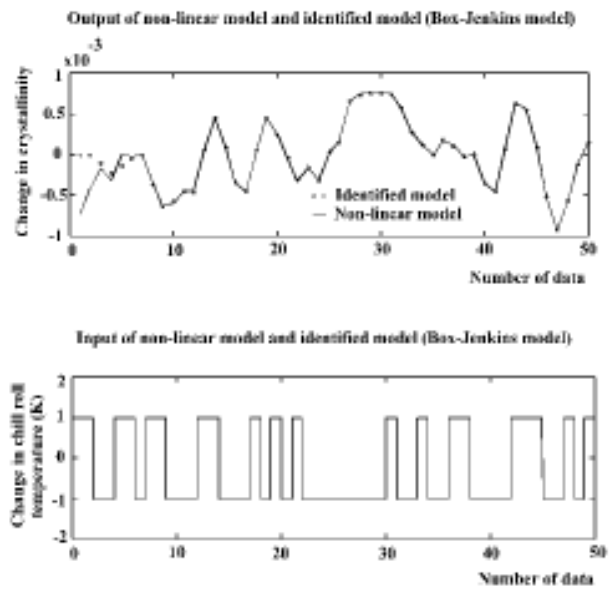| Identified parameters	Values
h_0	0
h_1	-4.86×10^{-6}
h_2	-8.77×10^{-5}
h_3	-2.21×10^{-4}
h_4	-2.67×10^{-4}
h_5	-2.36×10^{-4}
g_1	-1.72×10^{-1}

Figure 4. Comparison between the output of non-linear model and identified model around operating condition of $\alpha = 3.9 \times 10^{-2}$ and $T_r = 350$ K.

the dynamics of output data. The error at beginning is due to the lack of initial input data points at initial iteration for Eq. (13).

3. Control of crystallinity

The application of the obtained model in section 3 is illustrated by incorporating it in a state feedback control design. The obtained model is used to conventionally design a digital controller and a state estimator (Ogata, 1995) as shown in Figure 5. The performance of the digital controller and state estimator is shown as Figure 6 and 7. Figure 6 shows the resulting response of crystallinity for a step change. This means the digital controller is requested to increase the crystallinity to next level with an amount of 1×10^{-3}. During eleven sampling time steps, the digital controller regulates the temperature of the chill roll in order to drive the crystallinity to the desired level. It can be seen that the temperature of the chill roll is to decrease as expected and the crystallinity reaches the desired level eventually. Figure 7 shows the disturbance responses of the same controller. A disturbance is
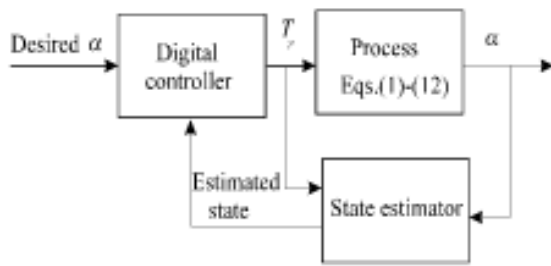
Figure 5. Diagram of a digital feedback control system based on identified model.
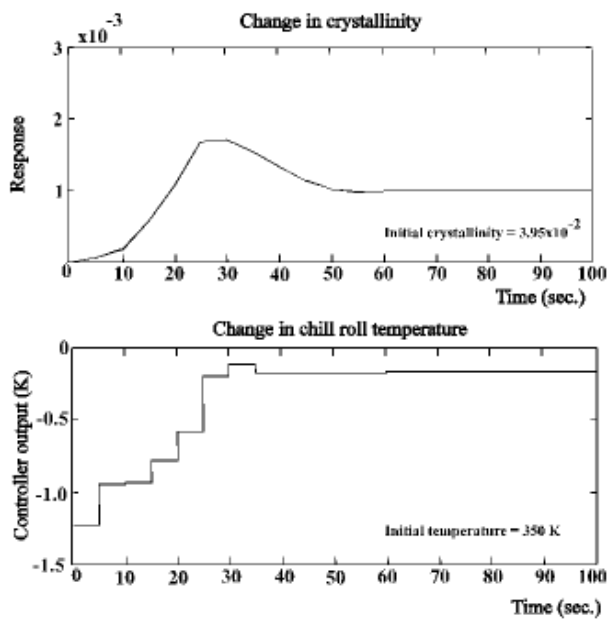

Figure 6. Performance of the digital controller in case of reference point.

Figure 7. Performance of the digital controller in case of acting disturbance.

Conclusion

An experimental process model can be obtained for a given position. The data does not actually come from a physical polymer sheet over the chill roll, instead the dynamics are simulated by the partial differential equations for the process. The same technique can also be extended to real process and multi-point measurements. As the results demonstrate, the model is quite capable of describing the process around the chosen operating points. This linear model can be used to design a dynamic crystallinity controller. The controller is applied for step changes in reference point and disturbance. The simulation results demonstrate that the proposed control can be implemented to real-time control crystallinity in polymer casting film process.

introduced to the process by reducing the temperature of polymer film to 1 K at contacting line on the chill roll. This situation may be caused by the temperature variation of the extruder. With this cause, the crystallinity increases from the desired level. The digital controller thus increases the temperature of the chill roll so as to reduce the crystallinity back to the desired level. As the resulting simulation shows, the controller can regulate the crystallinity despite the effect of disturbance. However, it should be noted that effects of a delay on the chill-roll temperature are not included in simulation.
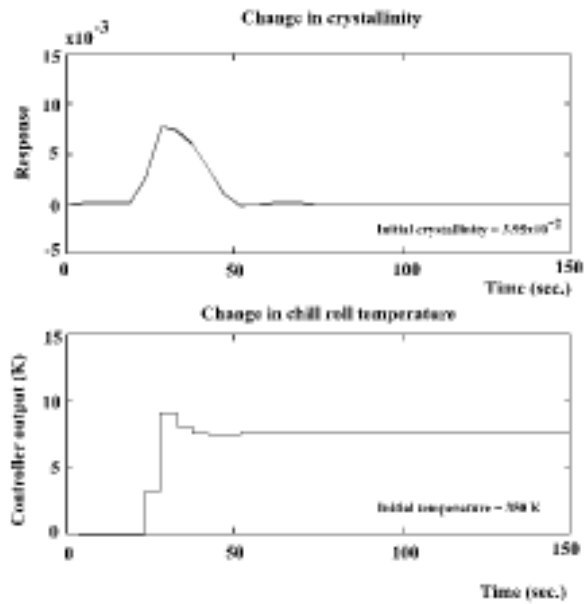
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Appendix

b, γ :   Constant material for induction time, $b = 8.0 \times 10^{11}$, $\gamma = 6$.

b_p :   Thermal effusivity of polymer, $b_p = 609.69$ (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, $C_p = 2140$ (J kg^{-1} K^{-1}).

e :   Thickness of the polymer sheet, $e = 3.56 \times 10^{-3}$ (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, $H_\infty = 83800$ (J kg^{-1}).

K_g :   Nucleation exponent, $K_g = 2.99 \times 10^5$ (K^2).

k :   Conduction heat transfer coefficient, $k = 0.193$ (J m^{-1} s^{-1} K^{-1}).

n :   Avrami index, $n = 3$.

T_g :   Glass transition temperature, $T_g = 255$ (K).

T_0^m :   Equilibrium melting temperature, $T_0^m = 445$ (K).

t_m :   Material constant for induction time, $t_m = 8.0 \times 10^{11}$.

$(1/t_{1/2})_0$:   Isothermal crystallization half time, $(1/t_{1/2})_0 = 2.07 \times 10^6$ (s^{-1}).

R :   Universal gas constant, $R = 0.831$ (J mol^{-1} K^{-1}).

U^* :   Activation energy, $U^* = 6284$ (J mol^{-1}).

v_x :   Polymer velocity, $v_x = 0.001$ (m s^{-1}).

W_e :   The width of a slot nozzle, $W_e = 0.02$ (m).

ρ :   Density, $\rho = 900$ (kg m^{-3}).