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**CHEMICAL PROCESS SYSTEMS
LABORATORY**

Control of Batch Free Radical
Polymerization Reactors

by

K. Y. Choi
D. Butala

RESEARCH REPORT

CHEMICAL PROCESS SYSTEMS ENGINEERING LABORATORY

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POLYMERIZATION REACTORS

K.Y. Choi
D. Butala

**A CONSTITUENT LABORATORY OF
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**THE UNIVERSITY OF MARYLAND
COLLEGE PARK, MARYLAND 20742**

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POLYMERIZATION REACTORS

K. Y. Choi* and D. Butala

Department of Chemical and Nuclear Engineering
and Systems Research Center
University of Maryland
College Park, MD 20742

ABSTRACT

This paper describes a new approximation method to synthesize open loop control schemes for the control of copolymer composition and molecular weight in batch and semibatch polymerization reactors. The open loop control policies are derived from the transient response of the fictitious feedback control system obtained by numerical model simulations. The applicability of the proposed method is demonstrated through numerical examples.

INTRODUCTION

A precise control of polymer properties has become one of the major issues in optimizing industrial polymerization processes recently. Many polymerization processes involve batch or semibatch reactors to produce small-to-medium volume engineering and speciality polymers. Although continuous processes, in general, produce polymers of more consistent quality in large volume than batch processes, the latter are still industrially very important and particularly well suited for the production of polymers of varying grades in a rapidly changing market environment. Since the productivity of any batch polymerization process depends upon consistent reactor operation designed to prepare polymers of desired quality, it is extremely important to develop computational methods of optimal batch reactor control policies. With increasingly stringent product quality specifications, the use of detailed process models in control system design is now becoming more important than ever.

In free radical copolymerization processes, controlling both copolymer composition and molecular weight or molecular weight distribution (MWD) is of primary importance. For instance, two styrene-acrylonitrile (SAN) copolymers differing more than 4 % in acrylonitrile level are incompatible, resulting in poor physical and mechanical properties [1]. It is also well known that significant batch-to-batch variation in product polymer properties is quite common in industrial processes and resulting offspecification products are often wasted.

Many of batch or semibatch polymerization reactor control problems stem from the lack of

accurate on-line polymer property sensors. Although some promising progress has been made in recent years in developing on-line sensors for certain polymerization systems [2], on-line sensors for the measurement of many important polymer properties are not readily available at this time. If such on-line sensors are available the polymer properties can be precisely controlled by using feedback controllers even in batch or semibatch polymerization processes. Unfortunately, this is not quite possible with present sensor technology. Some recent works show that extended Kalman filters can be used satisfactorily for the molecular weight control in batch free radical homopolymerization processes [3,4]. Due to this on-line sensing problems, batch or semibatch polymerization reactor control has always been viewed as an open loop controller design problem which consists of the following general design procedure:

- (i) Synthesis of open loop time varying control policies which minimize properly defined performance index (Task level control). Inevitably, this step requires detailed process model and deep physical insight into the process behavior as a prerequisite.
- (ii) Design of control systems which will drive the plant to follow the open loop control policies obtained in step (i) as closely as possible (Execution level control).

This paper describes the development of the task level control scheme for free radical batch and semibatch copolymerization reactors. Here, the inverse feedback control technique is used with detailed dynamic copolymerization reactor model to derive the time varying monomer addition rate and/or reaction temperature profiles which are required to yield styrene-acrylonitrile copolymers of desired composition and molecular weight.

DESIGN OF OPEN LOOP CONTROL STRATEGY

Let us consider a batch dynamic process represented by the following nonlinear modeling equations:

$$\dot{\underline{x}} = \underline{f}(\underline{x}, \underline{u}) \quad (1)$$

with initial conditions

$$\underline{x}(0) = \underline{x}_0 \quad (2)$$

where \underline{x} is an n -state vector and \underline{u} an m -control vector ($n \geq m$, $m \geq 1$). Suppose that we wish to find open loop controls that would bring certain components of \underline{x} exactly to some fixed desired values (i.e., set points) during the entire processing period: i.e.,

$$x_i(0 \leq t \leq t_f) = x_{i,d}, \quad i=1, \dots, q \quad (q \leq n) \quad (3)$$

or

$$\dot{x}_i = 0, \quad 0 \leq t \leq t_f, \quad i=1, \dots, q \quad (4)$$

where t_f is the total process (batch) time, x_i the state variable being controlled, and $x_{i,d}$ the desired value of the state variable. In batch copolymerization processes, x_i may be bulk phase monomer composition or copolymer molecular weight. The perfect control \underline{u} may be found if there exists \underline{u} which satisfies eq.(4).

If such \underline{u} can be represented explicitly as a function of state variables

$$\underline{u} = \underline{F}(\underline{x}) \quad (5)$$

then, the perfect open loop control trajectories, $\underline{u}_p(t; 0 \leq t \leq t_f)$ will be determined completely by solving eqs.(1) and (5). In most practical situations, however, such perfect controls (\underline{u}_p) may be extremely difficult or impossible to derive and express in implementable form.

A classical approach to optimal control synthesis is to use the necessary conditions of the Pontryagin's maximum principle [5-7]; however, when the dimension of the process model is large and the process model is very complex (e.g., copolymerization reactor model) it may be impractical and difficult to derive the optimal control policies using the maximum principle. Moreover, when the Hamiltonian is a linear function in control (\underline{u}), one faces a singular control problem which requires special solution techniques. For fed batch type (semibatch) reactor operations such singular control problems occur frequently.

One of the alternative optimal control method is to use approximation methods such as control vector parameterization technique [7] in which the parameterization of the optimal control is made by expanding the control variable(s) in a set of trial functions, ϕ_{ij} , of the following form:

$$u_i(t) = \sum_{j=1}^m a_{ij} \phi_{ij}(t) \quad (6.1)$$

or

$$u_i(\underline{x}) = \sum_{j=1}^m b_{ij} \phi_{ij}(\underline{x}) \quad (6.2)$$

Then, any parameter optimization techniques may be used to determine the optimal set of coefficients a_{ij} or b_{ij} . This approximation method is attractive in that no adjoint equations need to be solved. However, it is

required that the functional form of the optimal control be prespecified and proper weighting factors be chosen when more than one variables are controlled. Obviously, this method needs considerable amount of physical insight into the process behavior being considered.

Therefore, it is desirable to develop the method which may obviate such shortcomings of the standard control vector parameterization technique. Let us first consider a feedback control system shown in Figure 1. Here, the

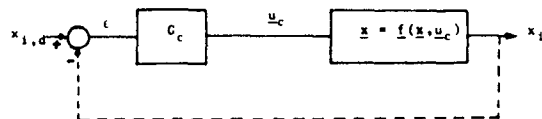


Figure 1 Feedback control system.

variables to be controlled (x_i) are monitored on-line. The objective of this feedback control system is to minimize the set point deviations, $e_1 = |x_{i,d} - x_i|$ ($x_{i,d}$ = fixed, $0 \leq t \leq t_f$), by using the feedback control, \underline{u}_c . Recall that in our original control system (i.e. batch copolymerization process) such on-line sensors do not exist and thus it is not feasible to form a feedback control structure as shown in Figure 1. In fact, this is why we have been considering the open loop controller design problem for the polymerization reactors from the beginning of this discussion. If the feedback control system of Figure 1 represents the batch polymerization reactor control system it is clear that we are now considering a "fictitious" feedback control system with "fictitious" on-line sensors.

If one can design the feedback controller (G_c) which will minimize the servo error (e_1) for this fictitious feedback control system, it is evident that the entire time-varying trajectory of $\underline{u}_{c1}(0 \leq t \leq t_f)$ resulted from the good feedback control of the process is what has been required to attain the desired servo response. Conversely, if the entire trajectory of such control \underline{u}_{c1} is known "a priori", this control \underline{u}_{c1} can be implemented into the actual open loop process where no on-line measurements of variables x_i are available and this control should lead to the same open loop response as the fictitious closed loop response from which \underline{u}_{c1} has been derived. In other words, the open loop control policy is deduced by inverting the transient response of the fictitious feedback control.

Then, our next problem is to obtain such feedback control profiles $\underline{u}_{c1}(t; 0 \leq t \leq t_f)$ "a priori". This can be accomplished by performing dynamic simulation of the process control model. Here, the feedback controller G_c of the fictitious feedback control system is first designed until the best closed loop response is obtained. Then, the transient trajectory of the manipulated variable, $\underline{u}_{c1}(t; 0 \leq t \leq t_f)$, is recorded. Since \underline{u}_{c1} is what has been needed to attain good servo response of the fictitious feedback control system, it

is now clear that the use of this control trajectory in the actual open loop process should result an identical control performance with the fictitious closed loop system.

The advantages of this inverse feedback control technique are: (i) The open loop control policy is determined by directly solving the model of corresponding fictitious closed loop control system; (iii) Since this method is based on the dynamic simulation of the process model, very sophisticated and thus more accurate process model can be used for the controller design; (iii) There is no need to introduce any weighting factors into the control objective. This is particularly attractive when more than one control variables are used; (iv) The effect of various operating conditions such as initial reactor conditions and manipulative variable characteristics can be analyzed easily by performing dynamic model simulations.

The factors which influence the quality of the open loop control policies determined by this inverse feedback control technique are (i) the accuracy of the process model and (ii) the goodness of the feedback controllers used in the numerical simulation of the fictitious closed loop control system. The interactions between the feedback loops for multivariable systems may also affect the quality of control policies. In the next section, we shall use this technique to find open loop control policies for the control of batch and semibatch SAN copolymerization reactors.

CONTROL OF SEMIBATCH COPOLYMERIZATION REACTORS

The control objective here is to maintain constant copolymer composition and constant polymer molecular weight during the course of polymerization by manipulating reactor variables such as monomer addition rate and reactor temperature. The modeling equations of the semibatch copolymerization reactor of volume V take the following form:

$$\frac{dM_1}{dt} = \frac{u}{V}(M_{1f} - M_1) - [(k_{p11} + k_{f11})P + (k_{p21} + k_{f21})Q]M_1 \quad (7)$$

$$\frac{dM_2}{dt} = \frac{u}{V}(M_{2f} - M_2) - [(k_{p22} + k_{f22})Q + (k_{p12} + k_{f12})P]M_2 \quad (8)$$

$$\frac{dI}{dt} = \frac{u}{V}(I_f - I) - k_d I \quad (9)$$

$$\frac{dV}{dt} = u \quad (10)$$

$$\rho C_p V \frac{dT}{dt} = V(\Delta H_r)(R_{sp}) - h_c A_c(T - T_c) + u\rho C_p(T_f - T) \quad (11)$$

$$R_{sp} = [(k_{p11} + k_{f11})P + (k_{p21} + k_{f21})Q]M_1 + [(k_{p22} + k_{f22})Q + (k_{p12} + k_{f12})P]M_2$$

where

$$P = \left(\frac{2fk_d I}{(k_{t11} + k_{t12}) + 2\beta(k_{t11} + k_{t12}) + \beta^2(k_{t11} + k_{t12})} \right)^{1/2}$$

$$\beta = \frac{k_{p12} + k_{f12}}{(k_{p21} + k_{f21})\phi} ; \phi = \frac{M_1}{M_2} ; Q = \beta P \quad (12)$$

M_1 and M_2 are the molar concentrations of monomer 1 (styrene) and monomer 2 (acrylonitrile), respectively, I the initiator concentration, and u the volumetric monomer-initiator feed rate. Subscript f denotes the feed conditions. Quasisteady state approximation has been applied to live polymers (P, Q). The copolymer molecular weight and MWD are computed by solving molecular weight moment equations. Notice that the reactor volume V varies with time due to the addition of monomers. The instantaneous copolymer composition is described by the Mayo-Lewis equation:

$$F_1 = \frac{r_1 \phi^2 + \phi}{r_1 \phi^2 + 2\phi + r_2} \quad (13)$$

which can be held constant by maintaining constant bulk phase monomer mole ratio (ϕ). In eq.(13), r_1 and r_2 represent reactivity ratios.

Isothermal copolymerization: To justify the necessity of the proper control system for the production of SAN copolymers of desired composition and molecular weight, the numerical simulation of isothermal batch solution copolymerization was conducted and the results are shown in Figure 2. The reactor was initially charged with initiator (AIBN) and monomers with mole ratio $\phi = 1.0$. As expected, significant variations in composition and molecular weight occur as monomer conversion increases. Such nonuniformity in polymer properties is not acceptable for practical application of the polymers.

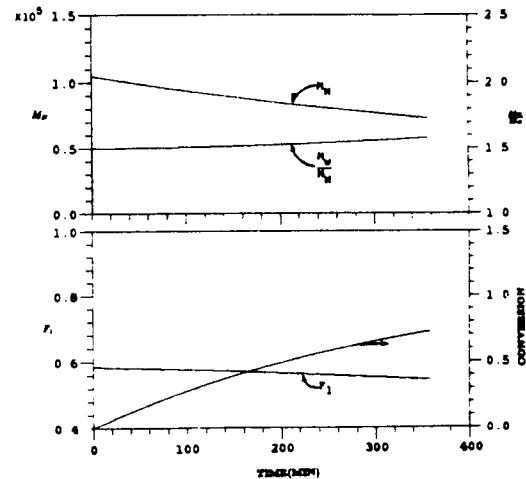


Figure 2 Isothermal batch copolymerization (70°C , $f_g = 0.5$, $I_0 = 0.05 \text{ mol/l}$).

Control of copolymer composition in isothermal semibatch reactor: The inverse feedback control method has been applied to control isothermal copolymerization process. Here, monomer addition policy is sought to maintain constant instantaneous copolymer

composition by holding the constant monomer mole ratio (ϕ) in the bulk phase. The controller design procedure is as follows:

- (i) Assuming perfect fictitious copolymer composition sensor is available, perform the dynamic simulation of the fictitious feedback control system and find the best feedback controls which minimize the servo errors. Any constraints on the control variables of the form $u_{\min} < u < u_{\max}$ can be incorporated into the simulation scheme. The final batch time is reached as the desired conversion is attained or as the reactor is full.
- (ii) Record the time varying trajectory of resulting control variable ($u_c(t)$) from $t=0$ to $t=t_f$.
- (iii) Implement the entire trajectory of u_c obtained from (ii) into the actual open loop control system where no on-line composition sensor is available.

In practice, the original control signal $u_c(t)$ resulted directly from the dynamic simulation of the fictitious feedback control system tends to be noisy and may be impractical for implementation into the actual open loop process. Such problem can be circumvented by smoothing or filtering the original feedback signals and fitting them into the properly parameterized forms. The above procedure can be easily applied to multivariable systems. Figure 3 illustrates

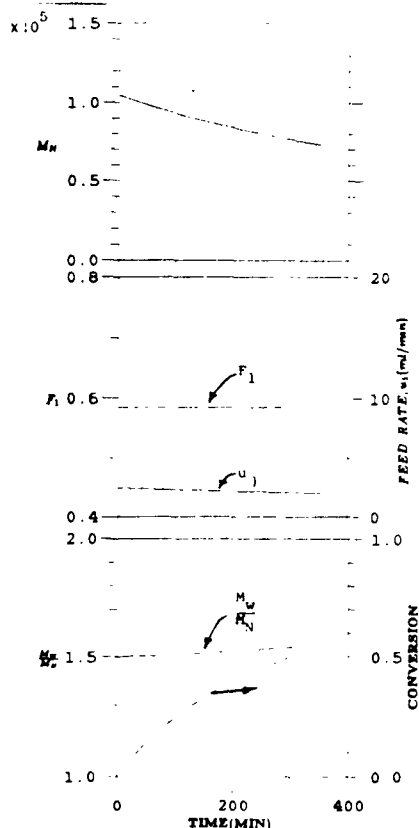


Figure 3 Open loop composition control in isothermal semibatch reactor (70°C , $f_s = 0.5$, $I_0 = 0.05 \text{ mol/l}$).

the performance of the open loop controller designed by the proposed method. Note that copolymer composition control is excellent. Although the total amount of added monomers is not great, such composition adjustment results in a significant improvement of the copolymer composition control. However, molecular weight varies considerably with conversion. Clearly, this is because only one manipulated variable was used for the nonlinear multivariable interacting system.

Control of copolymer molecular weight in batch reactor: If the copolymer molecular weight is the property parameter to be controlled, reaction temperature will be a better control variable than monomer addition rate. In this case, the copolymerization reactor is operated in a batch mode and time varying temperature set point program is required. Figure 4 shows the performance of the batch reactor controller designed by the inverse feedback control method. Again, very good molecular weight control is obtained with only slight offset. The temperature profile shown in Figure 4 can be easily implemented into actual reactor systems. Note, however, that this time the polymers of poor copolymer composition are obtained.

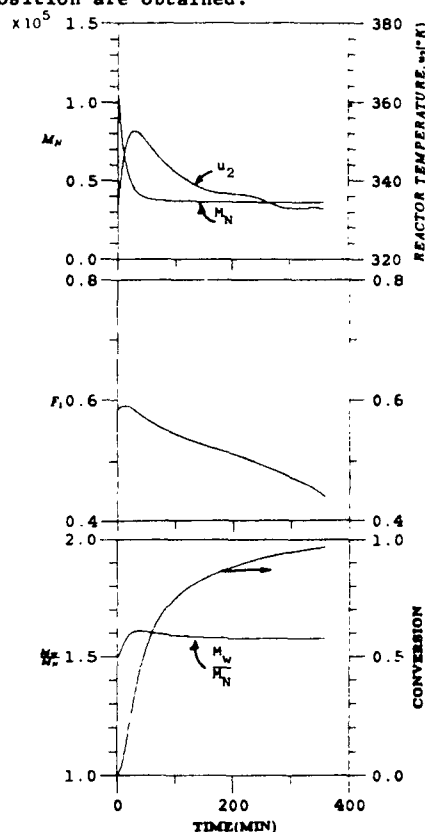


Figure 4 Open loop copolymer NW control in a batch reactor.

Control of copolymer composition and molecular weight in semibatch reactor: Figures 3 and 4 clearly illustrate that both composition and molecular weight should be controlled by manipulating monomer feed rate (u_1) and reactor temperature (u_2). The open loop control policies have been determined by the proposed method in which PI-controllers were used to obtain satisfactory fictitious feedback control. Figure 5 shows that excellent open loop control of composition and molecular weight is obtained. Slowly varying monomer addition rate and temperature set point profiles shown in Figure 5 can be easily implemented into the modern high speed process control computers. Notice that MWD is also maintained at constant level. In implementing the transient response of the control variables from the fictitious feedback control system, the noisy control signals were

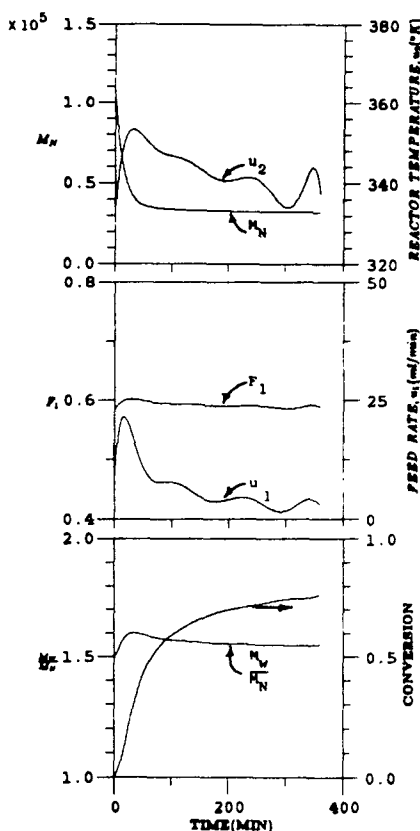


Figure 5 Open loop composition and MW control in a semibatch reactor.

filtered and fit into the ninth order polynomials. Figure 6 illustrates how the original fictitious closed loop signals (u_1 and u_2) are conditioned according to the procedure described earlier. Also shown in Figure 6 (insets) are the output responses obtained from the fictitious feedback control system.

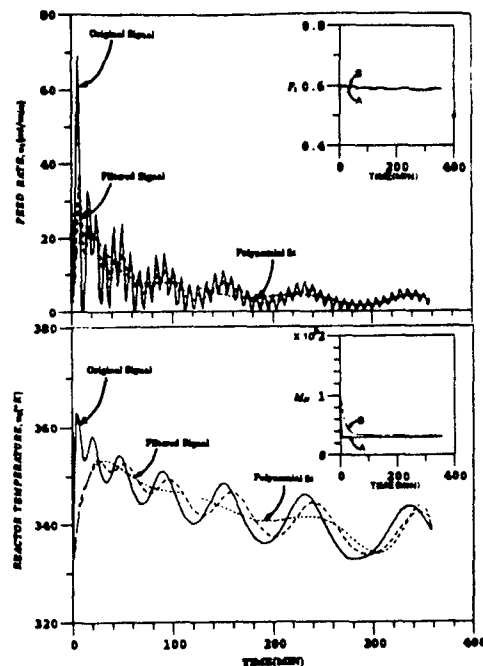


Figure 6 Conditioning of control variables for Figure 5: A, with original feedback signals; B, with polynomial fitted signals.

CONCLUSION

In this paper, a new approximation method has been introduced for the synthesis of open loop controls for batch and semibatch free radical copolymerization processes. This method is based on the discovery that the time varying trajectories of the manipulated variables obtained from the fictitious feedback control system are equivalent to the trajectories of the control variables or time varying set point programs required for the control of open loop control system in which no on-line sensors are available. Our simulation results indicate that this method is very powerful to synthesize the open loop control policies for the control of both copolymer composition and molecular weight in batch and semibatch reactors.

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