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Semibatch Free Radical Copolymerization
Reactors by Inverse Feedback Control
Technique**

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A DESIGN OF OPTIMAL CONTROL SYSTEM FOR SEMIBATCH
FREE RADICAL COPOLYMERIZATION REACTORS BY
INVERSE FEEDBACK CONTROL TECHNIQUE

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ABSTRACT

A novel Inverse Feedback Control technique is proposed for the design of optimal open loop control system in semi-batch free radical copolymerization process in which on-line polymer property sensors are absent. The open loop polymer quality control problem is converted to the closed loop controller design problem in which "fictitious" on-line polymer property sensors are assumed to be available. The time varying trajectories of reactor manipulated variables in such "fictitious" feedback control system are found to be equivalent to the optimal trajectories of control variables or set point programs required in the original open loop control system. The proposed technique can also be easily applied to the design of multivariable open loop systems for nonisothermal semibatch copolymerization processes. The validity and the applicability of the proposed technique based on "knowledge learning" principle are illustrated through the numerical examples of semi-batch copolymerization process control.

INTRODUCTION

A precise control of polymer properties has become a major issue in process optimization recently in highly competitive polymer industry. Varying and diversifying demands of polymer end users for tailor made polymers are increasingly high. Although continuous polymerization processes are widely used in industry to manufacture high volume commodity resins, many polymerization processes still involve batch or semibatch reactors to produce small-to-medium volume engineering and specialty polymers.

In free radical copolymerization processes, controlling both copolymer composition and molecular weight or molecular weight distribution (MWD) is of primary importance. However, significant batch-to-batch variation in polymer quality is quite common and resulting offspecification products are wasted. For instance, two styrene-acrylonitrile (SAN) copolymers differing more than 4% in acrylonitrile level are incompatible, resulting in poor physical and mechanical properties [Molau, 1965]. Such copolymer composition nonuniformity is due to the preferential depletion of comonomers during the course of polymerization. The molecular weight and MWD of copolymers may also vary significantly with the progress of reaction unless some corrective adjustments of reaction conditions are made. Unfortunately, however, controlling key polymer quality parameters such as copolymer composition and molecular weight is rather difficult in practice.

There are a few reasons why such undesirable product quality

variations occur and why minimizing such variations is difficult. One of the principal obstacles in developing polymer property control system is a lack of adequate on-line sensors required to monitor polymer properties such as copolymer composition distribution, molecular weight, MWD, copolymer chain sequence length distribution, residual monomer concentration, etc. In recent years, some promising progress has been made in developing on-line sensors for certain polymerization systems [e.g. on-line tensiometer and densitometer developed by Shork and Ray (1983)]. However, on-line sensors for the measurement of many other important polymer properties are not readily available at this time. Moreover, when more than one property parameters are to be controlled, defining the proper performance criteria is not also a simple task in the optimal design and control of such polymerization processes. Complexity of polymerization kinetics is also a factor which makes the design of sophisticated polymer reactor control system difficult.

Constructing the control systems for batch or semibatch copolymerization processes consists of the following two steps:

- (i) to develop the optimal time varying profiles of control variables to produce the polymers of desired quality,
- (ii) to develop the control system which will drive the plant to follow the optimal path obtained in (i) as closely as possible.

In order to develop optimal open loop control programs for batch polymerization processes, thorough understanding of

polymerization kinetics and the process dynamics is prerequisite and thus precise dynamic reactor models are required.

Wide literature is available on the modelling of homo- and co-polymerization reactors [for review, see Ray and Laurence (1979), and Hamielec et al. (1982)]. There are also a number of papers on the optimal control of batch free radical co-polymerization reactors [Hanna (1957), Hanson and Zimmerman (1957), Ray and Gall (1969), Gerrens (1969), Snuparek and Krska (1976), Guyot et al. (1981), Tirrell and Gromley (1981), Tsoukas et al. (1982), Garcia-Rubio et al. (1982), Garcia-Rejon et al. (1983), Johnson et al. (1982, 1983)]. In contrast to the requirements for free radical homopolymerization processes, the parameters needed to fully describe copolymerization processes are more numerous and there are more property parameters to be controlled. When only one polymer quality parameter (e.g. copolymer composition or molecular weight or MWD) is controlled by manipulating one control variable such as monomer addition rate or reaction temperature, other uncontrolled property parameters will inevitably deviate from their desired values as reaction proceeds. Thus, the tradeoffs between control policies should be made to achieve the satisfactory control of more than one property parameters. But choosing the weighting factors of objective functional "a priori" is very difficult and is quite often determined somewhat arbitrarily by well experienced process operators. Tsoukas et al. (1982) and Farber (1986) showed that by formulating the polymerization control problem as multiobjective problems and solving for the non-inferior set, all the competitions among the various objectives could be better

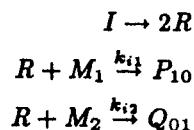
understood. Performance of the dual control scheme (i.e. temperature and monomer addition rate programs) for copolymer composition and molecular weight control was not illustrated, however.

In this paper, we shall present a new approach of determining the optimal open loop control policies for semibatch free radical copolymerization reactors. Unlike classical techniques based on the Maximum principle, Inverse Feedback Control principle is used with detailed dynamic copolymerization reactor model and the time varying monomer addition rate and temperature profiles are derived. The validity and the applicability of the proposed method will be demonstrated through numerical examples.

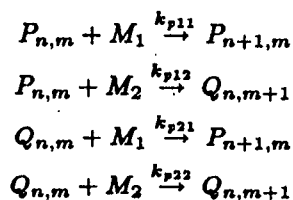
COPOLYMERIZATION KINETICS, AND REACTOR MODELLING

The following kinetic model is used to describe the homogeneous solution free radical copolymerization of styrene with acrylonitrile. When the solvent volume fraction is high, the effect of diffusion controlled termination (gel effect) is weak and thus ignored in the present model. Penultimate effect is also assumed negligible (These assumptions do not restrict the applicability of the proposed controller design technique, however.).

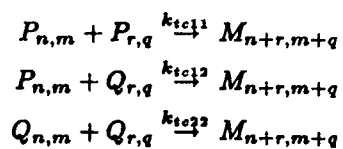
INITIATION



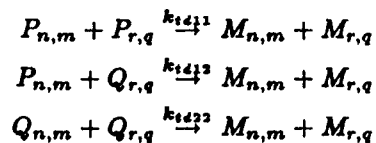
PROPAGATION



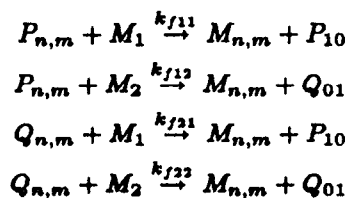
COMBINATION TERMINATION



DISPROPORTIONATION TERMINATION



CHAIN TRANSFER



where $P_{n,m}$ represents a growing copolymer chain with n units of monomer 1 and m units of monomer 2, and monomer 1 on the end. Similarly, $Q_{n,m}$ represents growing copolymer chains with monomer 2 on the end. $M_{n,m}$ denotes inactive (or dead) polymer.

The copolymer molecular weight and MWD, are computed by using three leading moments of the total number average copolymers. The instantaneous k -th moment is given by

$$\lambda_k^d = \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} (nw_1 + mw_2)^k M_{n,m} \quad k = 0, 1, 2, \dots \quad (1)$$

where w_1 and w_2 are the molecular weights of monomer 1 (styrene and monomer 2 (acrylonitrile), respectively. The total number average molecular weight (M_N), the total weight average molecular weight (M_w) and the polydispersity index (PD) which is a measure of molecular weight broadening are expressed as:

$$M_n = \frac{\lambda_1^d}{\lambda_0^d} \quad (2)$$

$$M_w = \frac{\lambda_2^d}{\lambda_1^d} \quad (3)$$

$$PD = \frac{M_w}{M_n} \quad (4)$$

In order to maintain constant monomer mole ratio in the bulk phase, more reactive monomer or monomer mixture is injected with initiator to the reactor. Thus, the modelling equations of the semibatch copolymerization reactor of volume V take the following form:

Reactor Mass Balance:

$$\frac{dM_1}{dt} = \frac{u}{V}(M_{1f} - M_1) - [(k_{p11} + k_{f11})P + (k_{p21} + k_{f21})Q]M_1 \quad (5)$$

$$\frac{dM_2}{dt} = \frac{u}{V}(M_{2f} - M_2) - [(k_{p22} + k_{f22})Q + (k_{p12} + k_{f12})P]M_2 \quad (6)$$

$$\frac{dI}{dt} = \frac{u}{V}(I_f - I) - k_d I \quad (7)$$

$$\frac{dV}{dt} = u \quad (8)$$

Energy Balance

$$\rho C_p V \frac{dT}{dt} = V(\Delta H_r)(R_{cp}) - h_c A_c (T - T_c) + u \rho C_p (T_f - T) \quad (9)$$

where

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

Live Polymer Moments

$$P = \left(\frac{2fk_d I}{(k_{tc11} + k_{td11}) + 2\beta(k_{tc12} + k_{td12}) + \beta^2(k_{tc22} + k_{td22})} \right)^{1/2} \quad (10)$$

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

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 addition rate. Other parameters are defined in the Notation .

Assuming pseudosteady state for live polymers, one can obtain the live polymer moment as follows [Ray and Laurence (1977)]

$$P_1 = \left(\frac{w_1 C_1 \alpha_1 + \frac{\alpha_1 \gamma Q_1}{r_1} + w_1 \left(\alpha_1 P + \frac{\alpha_1 \gamma Q}{r_1} \right)}{(1 - \alpha_1)} \right) \quad (11)$$

$$Q_1 = \left(\frac{w_2 C_2 \alpha_2 + \frac{\alpha_2}{\gamma r_2} P_1 + w_2 \left(\alpha_2 Q + \frac{\alpha_2}{\gamma r_2} P \right)}{(1 - \alpha_2)} \right) \quad (12)$$

$$P_2 = \left(\frac{w_1^2 C_1 \alpha_1 + \frac{\alpha_1 \gamma}{r_1} Q_2 + 2w_1 \alpha_1 P_1 + \frac{2w_1 \alpha_1 \gamma}{r_1} Q_1 + w_1^2 \left(\alpha_1 P + \frac{\alpha_1 \gamma Q}{r_1} \right)}{(1 - \alpha_1)} \right) \quad (13)$$

$$Q_2 = \left(\frac{w_2^2 C_2 \alpha_2 + \frac{\alpha_2}{\gamma r_2} P_2 + \frac{2w_2 \alpha_2}{\gamma r_2} P_1 + 2w_2 \alpha_2 Q_1 + w_2^2 \left(\frac{\alpha_2}{\gamma r_2} P + \alpha_2 Q \right)}{(1 - \alpha_2)} \right) \quad (14)$$

where

$$C_1 = \frac{(k_{f11}P + k_{f21}Q)M_1}{k_{p11}M_1} \quad (15)$$

$$C_2 = \frac{(k_{f22}Q + k_{f12}P)M_2}{k_{p22}M_2} \quad (16)$$

$$r_1 = \frac{k_{p11}}{k_{p12}} \quad ; \quad r_2 = \frac{k_{p22}}{k_{p21}} \quad (17)$$

$$\gamma = \frac{k_{p21}}{k_{p12}}$$

$$\alpha_1 = \left(\frac{k_{p11}M_1}{[(k_{p11} + k_{f11})M_1 + (k_{p12} + k_{f12})M_2 + (k_{tc11} + k_{td11})P + (k_{tc12} + k_{td12})Q]} \right) \quad (18)$$

$$\alpha_2 = \left(\frac{k_{p22}M_2}{[(k_{p22} + k_{f22})M_2 + (k_{p21} + k_{f21})M_1 + (k_{tc22} + k_{td22})Q + (k_{tc12} + k_{td12})P]} \right) \quad (19)$$

The moment equations for dead polymers are given by

$$\frac{d\lambda_0^d}{dt} = \left(\frac{1}{2}k_{tc11} + k_{td11}\right)P^2 + \left(\frac{1}{2}k_{tc22} + k_{td22}\right)Q^2 + (k_{tc12} + 2k_{td12})PQ + (k_{f11}M_1 + k_{f12}M_2)P + (k_{f22}M_2 + k_{f21}M_1)Q - \frac{\lambda_0^d}{V}u \quad (20)$$

$$\frac{d\lambda_1^d}{dt} = (k_{tc11}P + k_{td11}P + k_{tc12}Q + k_{td12}Q + k_{f11}M_1 + k_{f12}M_2)P_1 + (k_{tc22}Q + k_{td22}Q + k_{tc12}P + k_{td12}P + k_{f22}M_2 + k_{f21}M_1)Q_1 - \frac{\lambda_1^d}{V}u \quad (21)$$

$$\frac{d\lambda_2^d}{dt} = (k_{tc11}P + k_{td11}P + k_{tc12}Q + k_{td12}Q + k_{f11}M_1 + k_{f12}M_2)P_2 + (k_{tc22}Q + k_{td22}Q + k_{tc12}P + k_{td12}P + k_{f22}M_2 + k_{f21}M_1)Q_2 + k_{tc11}P_1^2 + k_{tc22}Q_1^2 + 2k_{tc12}P_1Q_1 - \frac{\lambda_2^d}{V}u \quad (22)$$

The kinetic constants used for the simulation of SAN copolymerization process are listed in Table 1. The instantaneous copolymer composition (F_1) is determined by the relative reactivities of monomers (r_1 and r_2) and bulk phase monomer mole fractions (f_1 and f_2) as follows:

$$F_1 = \frac{r_1 f_1^2 + f_1 f_2}{f_1 f_1 + 2f_1 f_2 + r_2 f_2^2} \quad (23.a)$$

or in terms of monomer mole ratio, ϕ ,

$$F_1 = \frac{r_1 \phi^2 + \phi}{r_1 \phi^2 + 2\phi + r_2} \quad (23.b)$$

Figure 1 shows the copolymer composition curves for different reaction temperatures. Note that copolymer composition is not very sensitive to variation in temperature. There is also an

Table 1: Numerical values of kinetic parameters and reaction conditions for Styrene-Acrylonitrile Copolymerization

	Parameter	Pre-exponential factor	Activation Energy (cal/gmol)	Ref
Initiator AIBN	k_d	$6.02 \times 10^{15} \text{sec}^{-1}$	31.730	(a)
	f	0.6	-	
Monomer Styrene		l/mol-sec		
	k_{p11}	1.06×10^7	7.067	
	k_{t11}	1.25×10^9	1.677	
	k_{f11}	2.31×10^6	12.670	
Acrylonitrile	k_{p22}	3.0×10^7	4.100	
	k_{t22}	3.3×10^{12}	5.400	
	k_{f22}	6.93×10^6	5.837	
	r_1	2.56(-)	1.190	
	r_2	$6.67 \times 10^{-5}(-)$	-4.340	
	k_{f12}	$30 \times k_{f11}$	12.670	
	k_{f21}	$5 \times k_{f22}$	5.837	
	ϕ_i	23(-)	-	
$\Delta H_R = 1.02[1 - (1.393F_1(1 - F_1))^{0.5}] + 16.03F_1 + 16.73(1 - F_1)$				(b)
Reactor Parameters:				
$f_s = 0.5, t_{maz} = 6(\text{hrs}), V_{maz} = 4.0(\text{l}), I_o = 0.05(\text{mol/l})$				
$h_c = 6.5 \times 10^{-4}(\text{kcl/sec} - \text{cm}^2 - ^\circ K), \rho C_p = 0.332(\text{kcl/l} - ^\circ K)$				

(a) Tsoukas, et al. (1982). (b) Miyama and Fujimoto (1961)

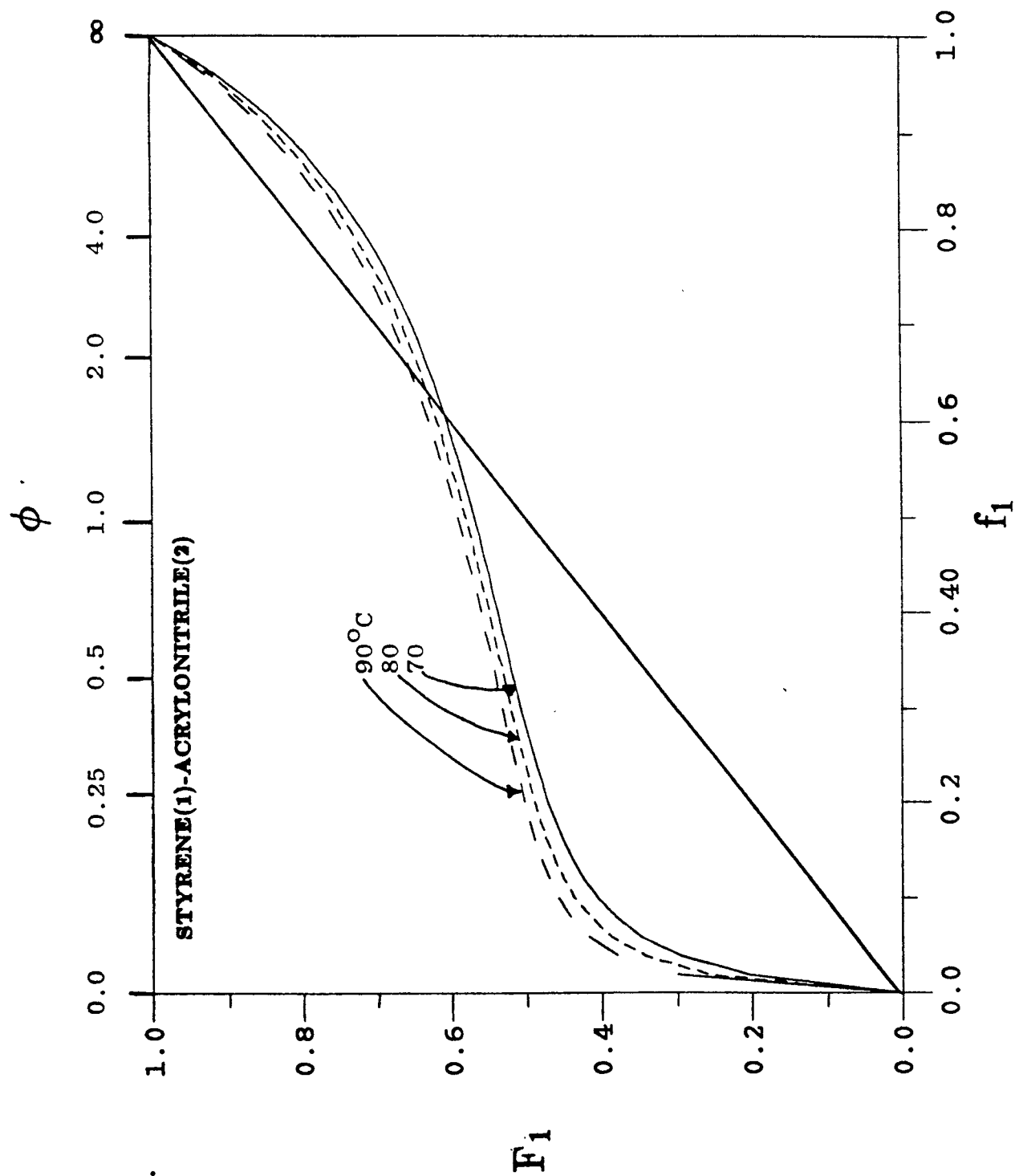


Figure 1: Styrene-acrylonitrile copolymer composition curves for different temperatures.

"azeotrope" at which the copolymer composition is identical with bulk phase monomer composition (i.e. $F_1 = f_1$). The ideal objective of any copolymer composition control scheme is to maintain the instantaneous copolymer composition F_1 (or instantaneous molar ratio of bulk monomers, ϕ) constant at some desired value.

PROBLEM FORMULATION

To justify the necessity of the proper control system to produce copolymers of desired quality, the numerical simulation of isothermal batch solution copolymerization process was carried out for styrene-acrylonitrile system. The reactor was initially charged with initiator (AIBN; azobisisobutyronitrile) and monomers with mole ratio corresponding to the $\phi = 1.0$ (acrylonitrile rich copolymer). The results are shown in Figure 2. As expected, significant variations occur in copolymer composition, number average molecular weight and MWD as conversion increases. Here, the conversion of monomer 1 is defined as follows:

$$X_1 = \frac{V_0 M_{10} + \int_0^t u(t) M_{1f} dt - V M_1(t)}{V_0 M_{10} + \int_0^t u(t) M_{1f} dt} \quad (24)$$

where V_0 is the initial reaction volume, M_{10} the initial monomer concentration in the reactor, M_{1f} the feed monomer concentration, and $u(t)$ the monomer feed rate. Such nonuniformity in polymer properties is not acceptable for practical application of the polymers.

Unlike in continuous polymerization processes, the feedback control of polymer properties is extremely difficult in batch or semibatch copolymerization processes. As mentioned earlier, this is due mainly to the lack of adequate on-line sensors for rapid measurements of copolymer composition and molecular weight.

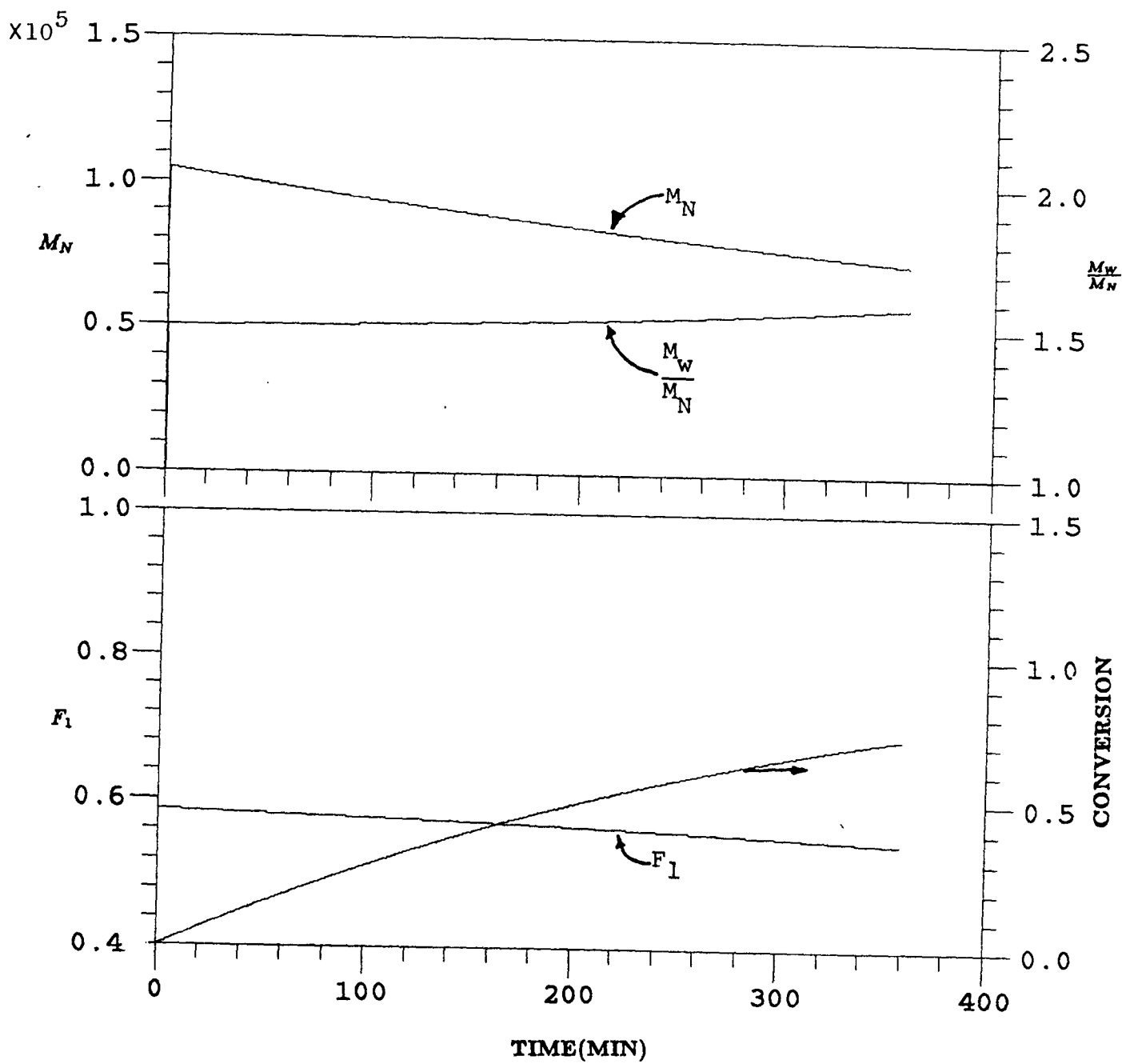


Figure 2: Isothermal batch copolymerization of styrene with acrylonitrile ($I_s = 0.05 \text{ mol/lit}$, 70° C , $f_s = 0.5$, $\phi_s = 1.0$).

This implies that closing the feedback control loop for property control is not possible. Thus, the batch or semibatch copolymerization reactor control problem has always been viewed as the open loop feedforward control problem which requires special temperature and reactant (monomer) feed programs. Such control policies are determined by conducting repeated experiments or by using the detailed physical model of the polymerization process.

For the system with n state variables (\underline{x}), m control variables (\underline{u}) and with dynamic behavior described by the ordinary differential equations

$$\dot{\underline{x}} = \underline{f}(\underline{x}, \underline{u}), \quad \underline{x}(0) = \underline{x}_0 \quad \text{for } 0 \leq t \leq t_f \quad (25)$$

the classical approach of finding the optimal time varying policies is to use the Maximum principle with properly defined objective functional such as

$$I[\underline{u}(t)] = G(\underline{x}(t_f)) + \int_0^{t_f} F(\underline{x}, \underline{u}) dt \quad (26)$$

which we wish to maximize or minimize. There may also arise constraints of the form

$$\underline{u}_* \leq \underline{u} \leq \underline{u}^* \quad (27)$$

which is quite common in many practical problems. For an isothermal semibatch copolymerization system with the objective

functional

$$I[u(t)] = \int_0^t (\phi - \phi_s)^2 dt \quad (28)$$

where ϕ_s denotes the desired copolymer molar ratio in the bulk phase, the Hamiltonian is defined by

$$H = (\phi - \phi_s)^2 + \underline{\lambda}^T \underline{f}(\underline{x}, u) \quad (29)$$

where $\underline{\lambda}$ refers to adjoint variables. When the control variable (u) is the monomer feed rate, it is easy to show that the Hessian matrix is singular (i.e. $\frac{\partial^2 H}{\partial u^2} = 0$) and thus we encounter a singular optimal control problem. For such systems, the coefficient of the linear control term in H vanishes identically on a singular arc. Thus, the control is not determined in terms of the state and adjoint variables by the necessary condition $\frac{\partial H}{\partial u} = 0$ (or minimizing H) along the singular arc but determined by the requirement that the coefficient of these linear terms remains zero on the singular arc. [Bryson and Ho (1975), Ray (1981)]. Due to the complexity and nonlinearity of the copolymerization reactor modelling equations, it is extremely difficult, if not impossible, to derive the optimal singular control policy for semibatch copolymerization problems. Designing the optimal control policies to maintain both copolymer composition and molecular

weight is even more challenging. In what follows, we shall present an alternative approach to solve such problems.

Inverse Feedback Control Principle

Let us first consider a simple single input single output (SISO) open loop control system as shown in Figure 3 where operator G denotes the plant, u_0 the open loop control, and x the plant output (or desired response). Here, our goal is to find the time varying optimal control u_0 which will force the system to follow the desired process trajectory, x . Clearly, such controller will be given by

$$u_0 = G^{-1}x \quad (30)$$

[In the isothermal semibatch copolymerization process, u_0 is the addition rate of more reactive monomer and x the monomer mole ratio in the bulk polymerizing mixture (i.e. ϕ).] Here, the desired trajectory of x will be the constant monomer mole ratio (x_d).

In general, the advantage of the open loop scheme is that the controller is easy to design; however, the disadvantages of the open loop scheme are the sensitivity of the controller performance to modelling errors and the inability to cope with unmeasured disturbance. Moreover, the plant should be invertible (eg. (30)). Now with the feedback control arrangement as shown in Figure 4, modelling errors and unmeasured disturbance can be dealt with effectively if properly tuned feedback

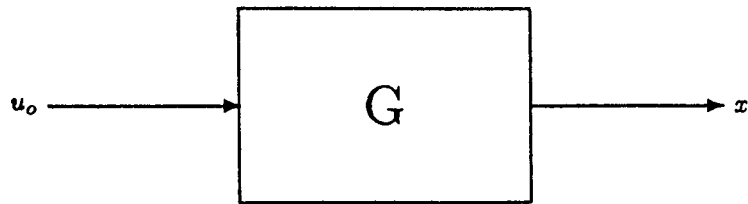


Figure 3 Open loop single input-single output control system.

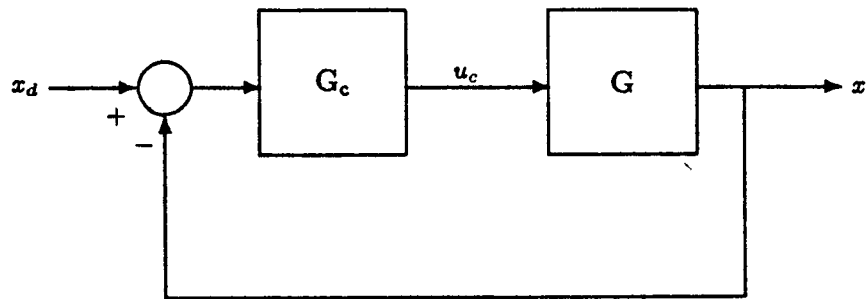


Figure 4 Closed loop single input-single output control system.

controller (G_c) is used.

It is easy to show that for the feedback control system shown in Figure 4 the closed loop trajectory of control u_c is given by

$$u_c = \frac{G_c}{1 + G_c G} x_d \quad (31)$$

Thus, the plant inverse becomes

$$G^{-1} = \frac{G_c u_c}{G_c x_d - u_c} \quad (32)$$

where $G_c x_d \neq u_c$.

If we can make the controller gain (G_c) as large as possible without causing system instability, then the plant inverse becomes

$$G^{-1} \approx \frac{u_c}{x_d} \quad (33)$$

Therefore, if the approximate plant inverse obtained from the feedback control system is used in Equation (30) the following equation is obtained:

$$u_0 \approx u_c \quad (34)$$

In other words, closed loop control (u_c) which is resulted from the feedback control system of Figure 4 is equivalent to the open loop feedforward control policy (u_0) which is being sought. Thus, if perfect feedback controller which can maintain the

output at its target value is found, the time varying trajectory of manipulative variable $u_c(t)$ from $t=0$ to $t=t_f$ will become the optimal open loop feedforward control ($u_o(t)$) and it will function perfectly even in the absence of on-line measurement device.

Now, we return to our semibatch copolymerization reactor control problem. As mentioned earlier, the key obstacle in implementing feedback control system for the polymer property control in the copolymerization process is a lack of on-line sensors to measure the polymer properties such as copolymer composition and molecular weight. This implies that implementing the feedback polymer property control scheme is not feasible in practical situation. Now, let us assume that there exists a perfect "fictitious" on-line copolymer composition sensor; then, the feedback copolymer composition control loop can be closed as in Figure 4 in which monomer addition rate is the control variable. If we can design the feedback controller G_c based on the accurate process model in such a way as to minimize the deviation of output (x) from the desired value (x_d), then the resulting trajectory of $u_c(t)$ from $t=0$ to $t=t_f$ (final batch time) will be the open loop control policy u_o which is being sought. Therefore, the original open loop controller design problem has been converted to the closed loop controller design problem which can be solved without significant difficulty.

The design procedure for open loop copolymer composition control system can be summarized as follows:

- (i) Develop first a detailed dynamic model of the

copolymerization process. Since the complexity of the process model does not cause any particular computational difficulties in solving the ordinary differential equations, one can use very detailed model which describes the polymerization behavior more precisely.

- (ii) Assume that perfect "fictitious" property measurement sensor is available to measure the copolymer composition. Perform the dynamic simulation of the "fictitious" feedback control system with "fictitious" sensor and find the best feedback controller which minimizes the deviation of the output from the set point value (i.e. desired copolymer composition). Any constraints on the control variable can be incorporated into the simulation scheme. The final batch time (t_f) is arrived as the desired conversion or reaction time is reached or as the reactor volume reaches its full capacity.
- (iii) Save the time varying trajectory of resulting control variable ($u_c(t)$) from $t=0$ to $t=t_f$.
- (iv) Implement the entire trajectory of $u_c(t)$ obtained in step (iii) into actual open loop control system where no on-line sensor is available. If $u_c(t)$ is the result of "perfect" feedback control, then $u_c(t) = u_o(t)$ and $u_o(t)$ should also result in a perfect open loop control in the absence of any measurement devices in

real copolymerization process.

In practice, the control signal $u_c(t)$ resulted from the simulation of "fictitious" feedback control system tends to be noisy and may be impractical for implementation into the "real world" open loop process. Thus, step (iv) is modified as follows so that easy implementation of $u_c(t)$ into the open loop system can be achieved; once the time varying noisy trajectory $u_c(t)$ is found, it can be smoothed or filtered and can also be fitted into any parameterized form such as

$$u_0 = \sum_{i=0}^N a_i t^i \quad (35)$$

Note that the principal factors influencing the efficiency of the proposed Inverse Feedback Control technique are (i) the accuracy of the process model, and (ii) the goodness of the feedback controller which maintains the output (copolymer composition) at its target value as closely as possible.

This technique can also be easily applied to multivariable control system. Let us consider a semibatch copolymerization process which requires the tight control of both copolymer composition and molecular weight or MWD. First, we need to pair the manipulated variables with output variables properly. In most free radical copolymerization, copolymer composition is very sensitive to bulk phase monomer composition, and molecular weight to reaction temperature. Moreover, very high polymer molecular

weight is obtained even at low monomer conversion in free radical polymerization. This characteristic leads us to fix set point of the molecular weight at its desired value from the beginning of the process without making significant errors. Thus, the multivariable "fictitious" feedback control system can be constructed as shown in Figure 5. Here, the fixed set points of ϕ_s and M_{Ns} are used throughout the polymerization period, $0 \leq t \leq t_f$.

Again, the original open loop control problem has been transformed to the multivariable feedback control problem. Now, any multivariable feedback control design technique may be used to achieve the best control performance. The resulting time histories of control variables (u_{c1} = monomer addition rate, u_{c2} = reactor temperature or jacket temperature) are equivalent to the desired open loop control policies. Any constraints on the control variables of the form

$$\underline{u}_{c,min} \leq \underline{u}_c \leq \underline{u}_{c,max} \quad (36)$$

can also be included in the model simulation without causing any computational difficulties. Once u_{c1} and u_{c2} are found through the numerical simulation of "fictitious" model based feedback control system, those control signals are filtered and parameterized in an appropriate manner for easy implementation into the open loop "real" process (Figure 6).

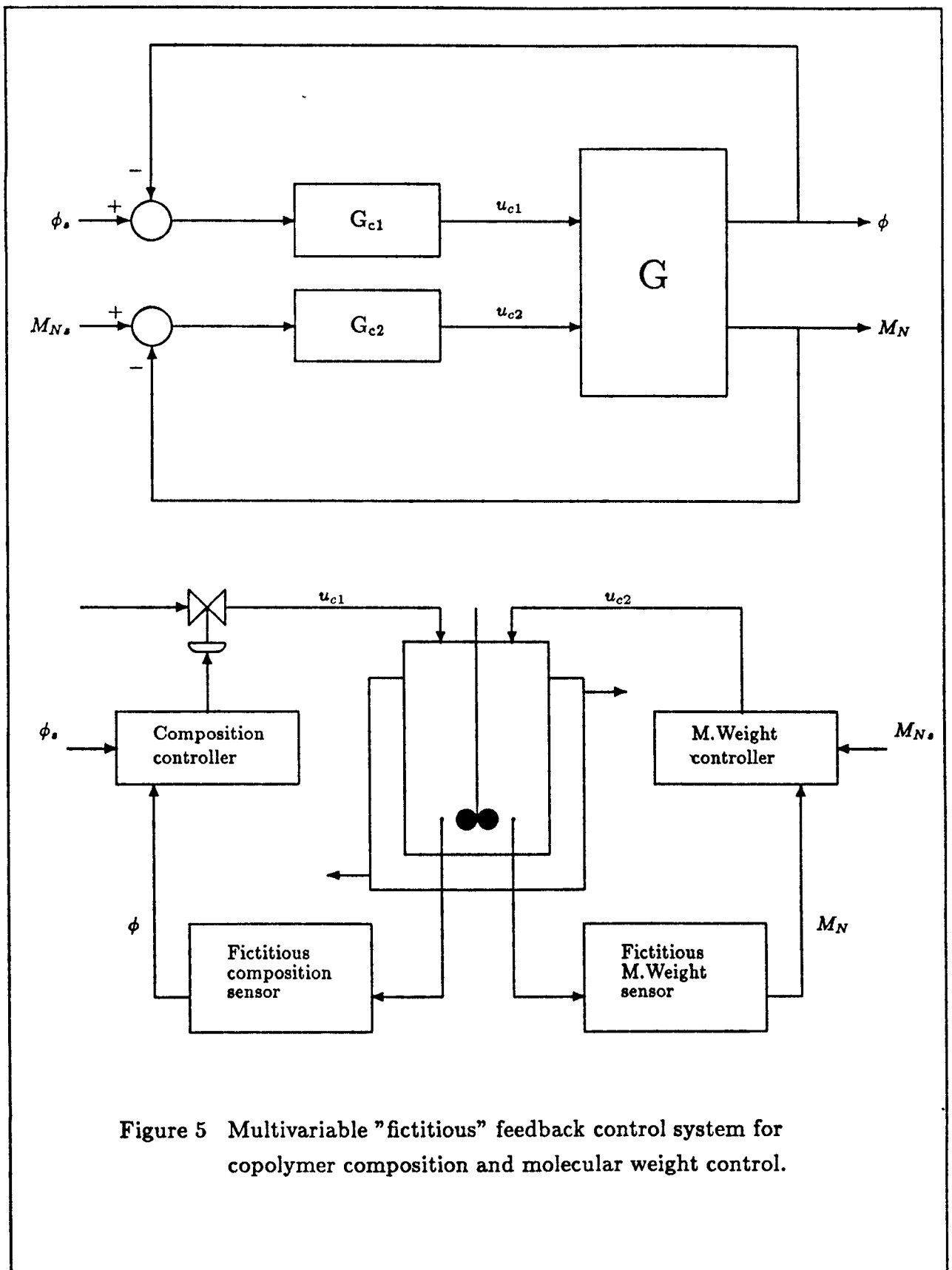
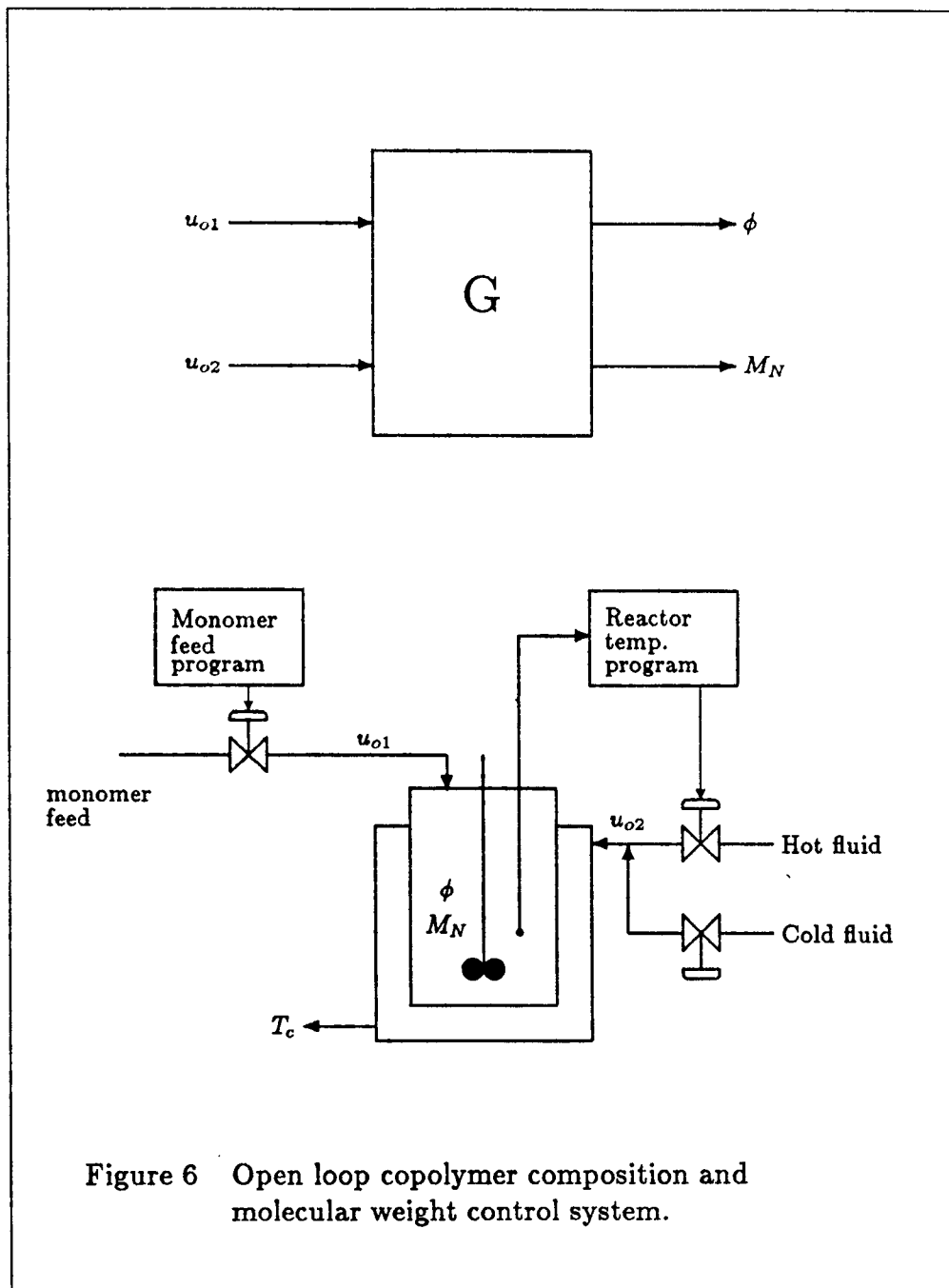


Figure 5 Multivariable "fictitious" feedback control system for copolymer composition and molecular weight control.



The major advantages of this Inverse Feedback Control (IFC) technique are as follows:

- (i) The open loop optimal feedforward control policy is determined by transforming the problem into the closed loop feedback controller design problem; various techniques are available for the design of feedback control systems.
- (ii) Very sophisticated and more accurate model can be used without causing any computational difficulties.
- (iii) Computational scheme is straightforward; one only needs an ordinary differential equation solver for numerical simulation of the dynamic process model.
- (iv) The effects of various operating conditions such as initial reactor conditions and reactant feed conditions can be analyzed easily.
- (v) Multivariable open loop control system can also be dealt with.

The underlying principle of this proposed technique is analogous to the knowledge learning principle used for the design of industrial robots; the knowledge of human experts on the complex process is implemented into the robots, which execute the process satisfactorily in the absence of the experts.

NUMERICAL EXAMPLES

In order to demonstrate the validity and applicability of the proposed Inverse Feedback Control technique, we shall present the numerical examples of both single input-single output open loop control problem (e.g. composition or molecular weight control) and two input-two output open loop control problem (i.e. composition and molecular weight control).

Control of a Single Variable

The proposed design technique is first applied to isothermal semibatch SAN copolymerization process. Here, optimal monomer addition policy is sought to maintain constant instantaneous copolymer composition (F_1) by maintaining constant monomer molar ratio, ϕ , in the bulk phase. The optimal open loop control policy has been determined according to the procedure in the previous section. Figure 7 shows the monomer addition rate ($u_1(t)$) which was used for the simulation and the resulting copolymer composition, copolymer molecular weight, MWD and conversion. Here, the feed stream is the mixture of initiator (AIBN) and two comonomers with $\phi_f > \phi_s$ where ϕ_s is the desired monomer mole ratio in the reactor. Note that excellent composition control is achieved with the policy of slowly decreasing monomer addition rate. Although, the total amount of added monomers is not great, such composition adjustment results in a significant improvement of copolymer composition control. Simple proportional-integral (PI) controller was used in the

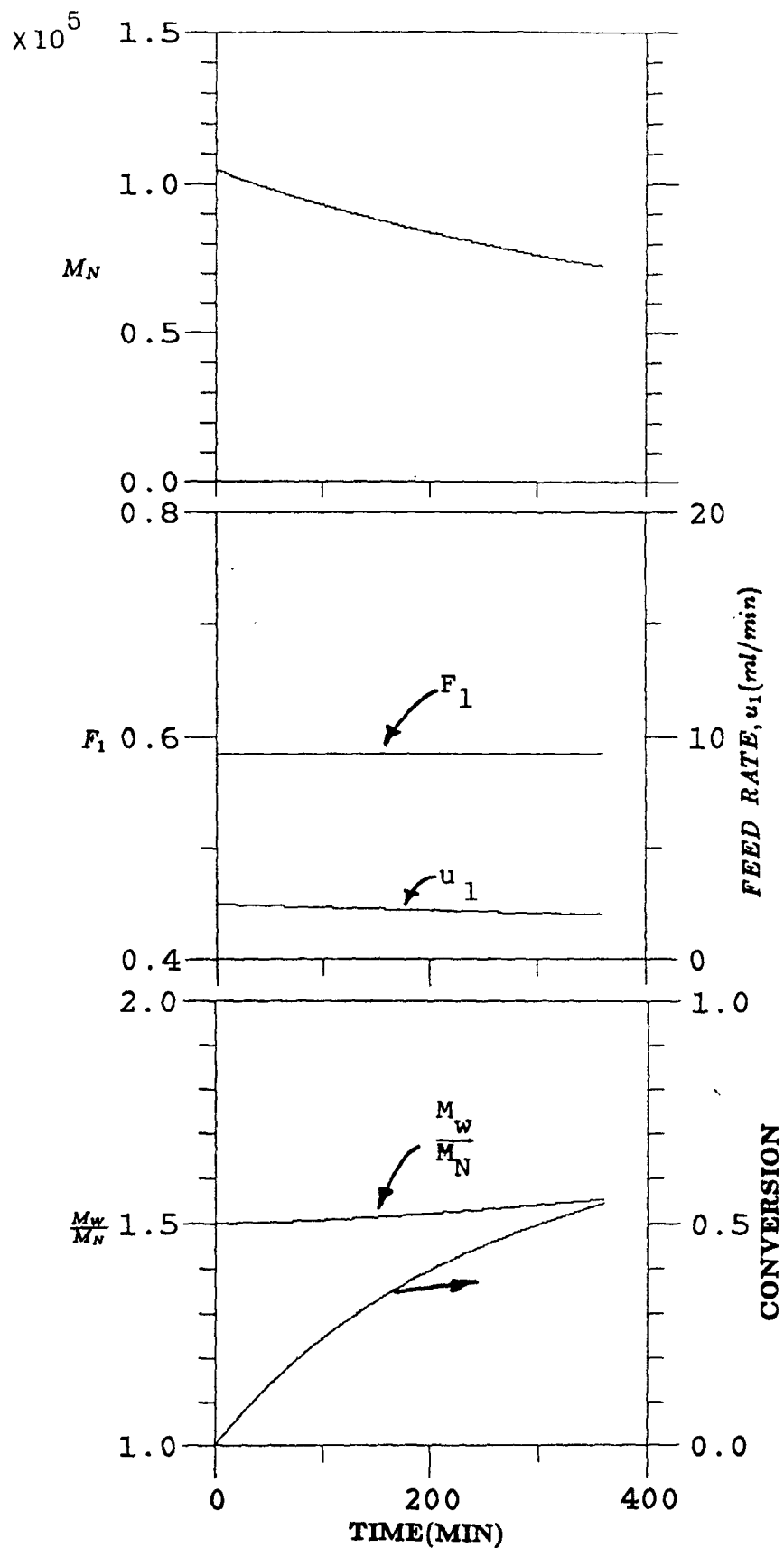


Figure 7: Open loop copolymer composition control in isothermal semibatch copolymerization reactor ($I_s = 0.05 \text{ mol/lit}$, 70°C , $f_s = 0.5$, $\phi_s = 1.0$).

"fictitious" closed loop control system and the resulting control trajectory was filtered and parameterized in ninth order interpolating polynomial in time. Although excellent control of copolymer composition is obtained, significant variation still occurs in molecular weight with slight variation in polydispersity as was observed by other workers [Tsoukas et al. (1982)] for similar systems. Clearly, this is because only one manipulative variable was used for this non-linear interacting system.

If the copolymer molecular weight instead of copolymer composition is the property parameter to be controlled, reaction temperature will be a better control variable than the monomer addition rate. In this case, the copolymerization reactor is operated in a batch mode. Thus, the simulation of "fictitious" feedback control system of batch non-isothermal copolymerization reactor was performed with reactor temperature (T) as a control variable. The resulting temperature trajectory ($u_c(t)$) was conditioned and implemented in open loop real reactor system. The open loop reactor simulation results are shown in Figure 8. Again, excellent molecular weight control is achieved. Note that desired reactor temperature profile consists of high initial temperature policy followed by gradually decreasing temperature policy. Under this control scheme, quite uniform MWD is obtained but considerable drift in copolymer composition occurs. Therefore, it is strongly required to implement both copolymer composition controller and molecular weight controller.

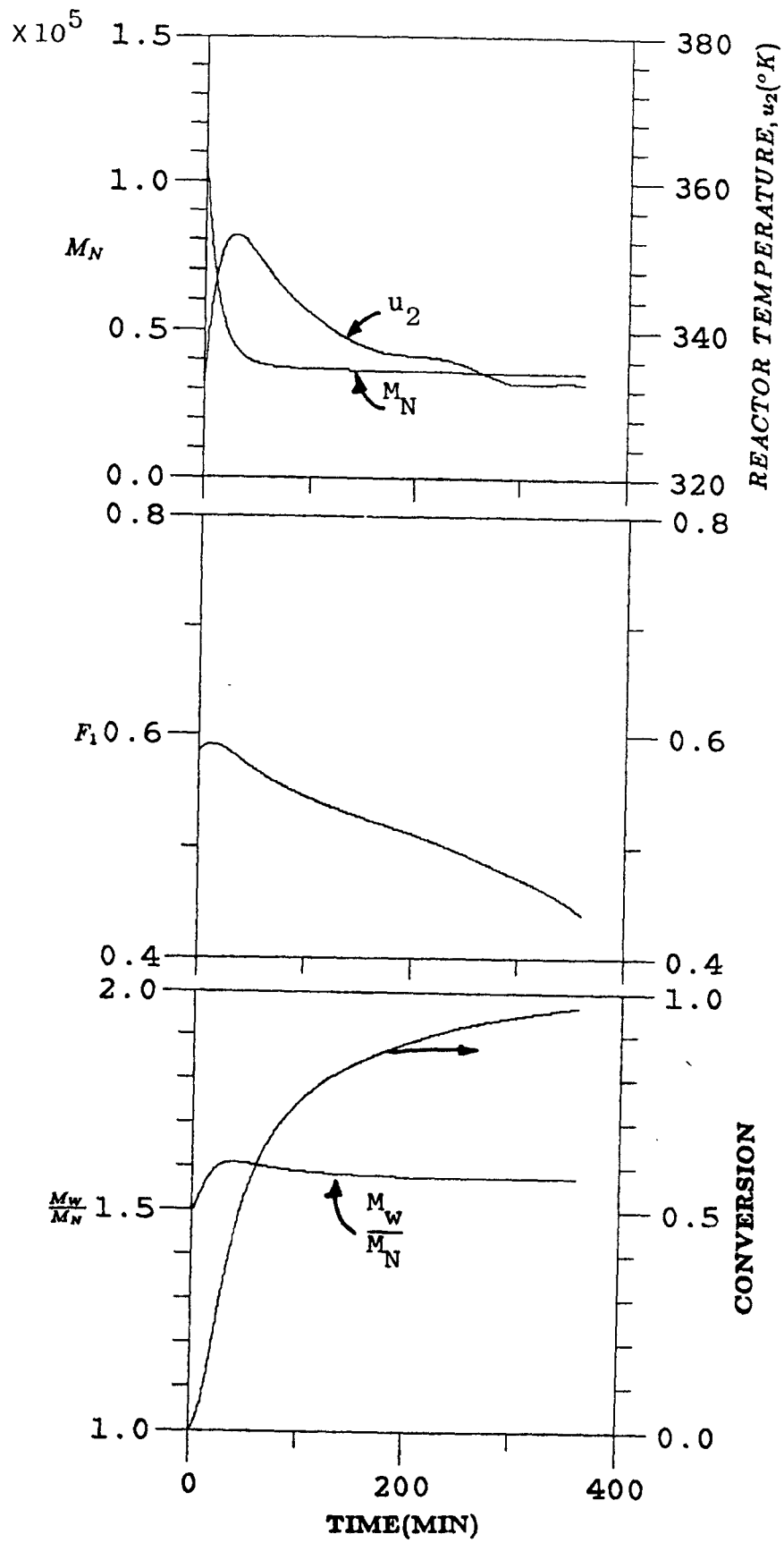


Figure 8: Open loop copolymer molecular weight control in a batch reactor ($I_s = 0.05$, mol/lit, $T_f = 50^\circ C$, $f_s = 0.5$, $\phi_s = 1.0$).

Control of Copolymer Composition and Molecular Weight

The open loop control programs for both copolymer composition control and molecular weight control have been determined by the proposed Inverse Feedback Control technique. PI controllers were used in both copolymer composition control loop and molecular weight control loop of "fictitious" multivariable feedback control system. Figure 9 shows the output responses of F_1 and M_N of the open loop semi-batch nonisothermal copolymerization process without "fictitious" on-line sensors. Note that excellent control of composition and molecular weight is achieved. Slowly changing monomer addition rate and temperature set point profiles shown in Figure 9 can be readily implemented into modern high speed process control computers. The effect of small deviation in molecular weight observed during the short initial reaction period may not be significant because overall conversion is quite low during this period. Also note that MWD is surprisingly uniform throughout the entire polymerization period. Figure 10 illustrates how the profiles of "fictitious" closed loop control signals (U_{c1} and U_{c2}) are conditioned according to the procedure described earlier. For smoothing, recursive first order lag filter was used. Also shown in Figure 10 (insets) are the output responses of molecular weight and composition parameters obtained by the "fictitious" feedback control system. When compared with M_N profiles in Figure 10, open loop molecular weight response in Figure 9 shows slight deterioration due to the adjustment of original "feedback" control signal.

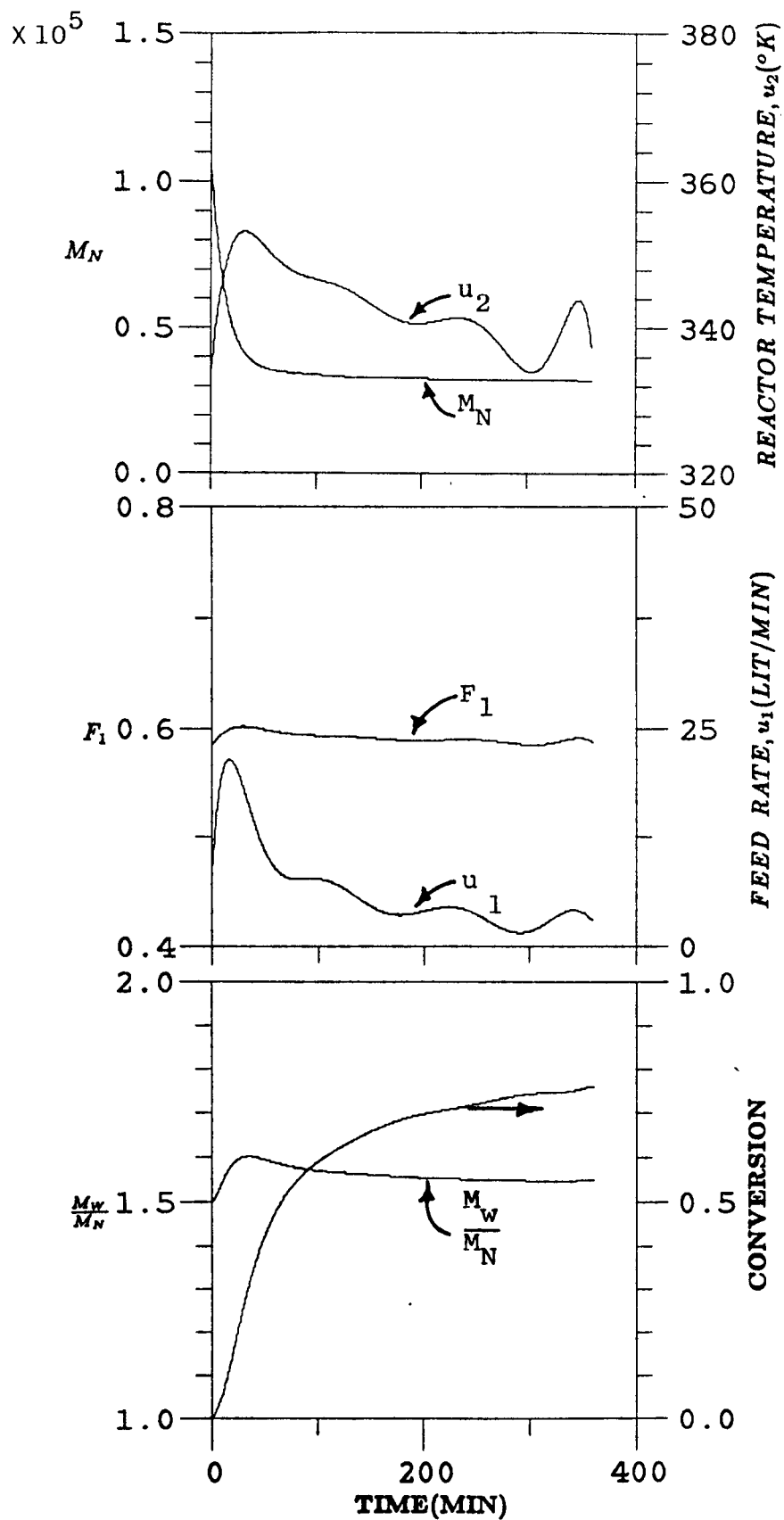


Figure 9: Open loop copolymer composition and molecular weight control in a semibatch copolymerization reactor ($I_s = 0.05$, mol/lit, $T_f = 50^{\circ}$ C, $f_s = 0.5$, $\phi_s = 1.0$).

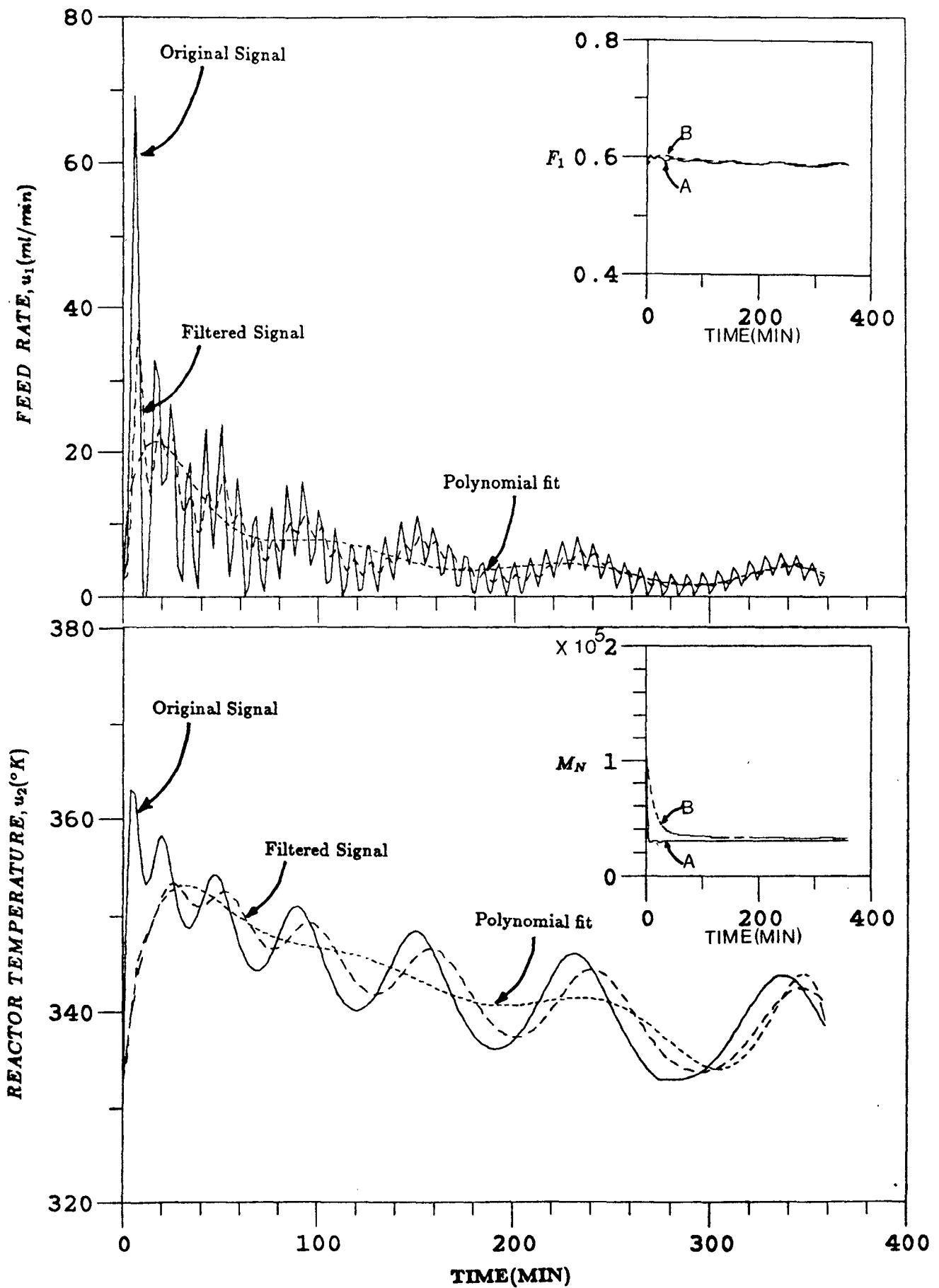


Figure 10: Conditioning of time varying profiles of manipulative variables for Figure 9: A, with original feed back signal; B, with polynomial fitted signal.

Figure 11 shows the performance of open loop control system when reactor jacket temperature (T_c) is used as a manipulative variable for molecular weight control. Both monomer addition rate (U_1) and jacket temperature (U_2) profiles are slightly more oscillatory than those shown in Figure 10. This is due to the superposition of cooling jacket dynamics onto the polymerization reactor dynamics. Here, the maximum temperature available for jacket fluid is assumed to be 100°C .

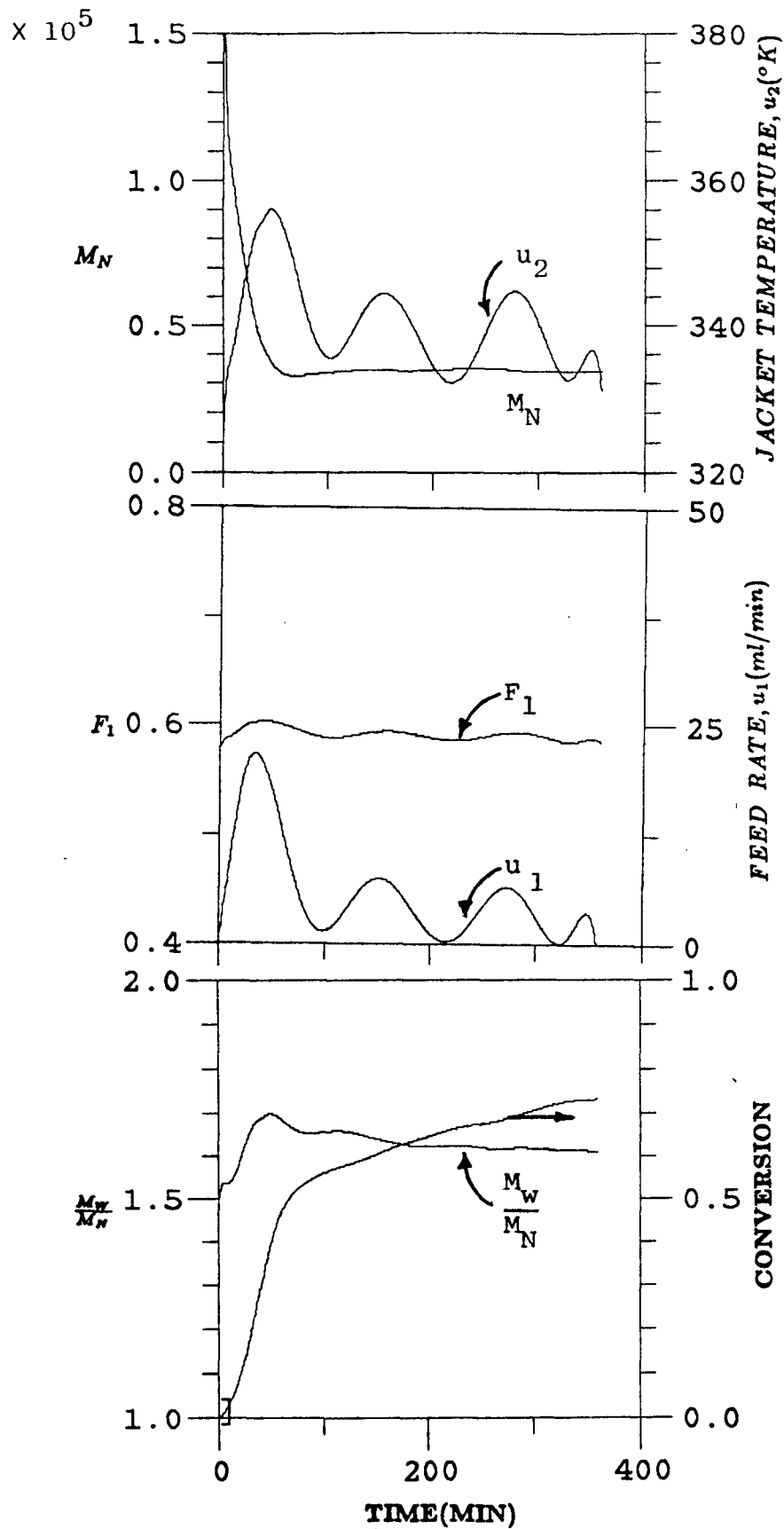


Figure 11: Open loop copolymer composition and molecular weight control in a semibatch copolymerization reactor ($I_s = 0.05$, mol/lit, $T_j = 50^{\circ}$ C, $f_s = 0.5$, $\phi_s = 1.0$).

CONCLUSION

In this paper, we have introduced a new approach for the design of optimal (in a strict sense, "suboptimal") open loop control strategy for semibatch free radical copolymerization processes in which no single control variable is sufficient to control copolymer composition and molecular weight simultaneously. In our proposed Inverse Feedback Control techniques, the open loop optimal control problem is converted to the closed loop control system design problem. For semibatch copolymerization process, optimal open loop programs of monomer addition rate and temperature variation are obtained by designing the "fictitious" feedback control system in which perfect on-line sensors are assumed to be available for copolymer composition and molecular weight measurements. The time varying trajectories of the manipulated variables obtained from such "fictitious" feedback control system are equivalent to the optimal trajectories of control variables or the time varying state set point values required in the original open loop control system.

Our simulation result indicates that excellent open loop control of copolymer composition and molecular weight is obtained with easily implementable control trajectories. Like other model-based optimal open loop control systems, the performance of the proposed open loop control scheme in real world polymerization processes will be dependent upon the accuracy of the reactor model being used. Finally, this Inverse Feedback Control technique will also be applicable to many other open loop control problems which have similar process characteristics as the

examples considered in this work.

Acknowledgement

This research was supported by the Systems Research Center at the University of Maryland, College Park.

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NOTATION

A_c	Heat transfer area per reactor volume (cm^2/liter)
C_p	Specific Heat of reaction mixture ($\text{cal}/\text{g}^\circ\text{K}$)
F_i	Mole fraction of i th monomer in copolymer, $i = 1, 2$
f	Initiator decomposition efficiency
f_i	Mole fraction of i th monomer in reaction mixture; $i = 1, 2$
h_c	Overall heat transfer coefficient ($\text{cal}/\text{cm}^2\text{-sec-}^\circ\text{K}$)
I	Initiator concentration in reaction mixture (mol/liter)
I_f	Initiator concentration in feed (mol/liter)
k_d	Initiation rate constant (sec^{-1})
k_{fij}	Chain transfer rate constant; $i, j = 1, 2$ ($\text{liter}/\text{mol-sec}$)
k_{pij}	Propagation rate constant; $i, j = 1, 2$ ($\text{liter}/\text{mol-sec}$)
k_{tcij}	Combination termination rate constant; $i, j = 1, 2$ ($\text{liter}/\text{mol-sec}$)
k_{tdij}	Disproportionation termination rate constant; $i, j = 1, 2$ ($\text{liter}/\text{mol-sec}$)
M_i	i th monomer concentration in reaction mixture; $i = 1, 2$ (mol/liter)
M_{iF}	i th monomer concentration in feed; $i = 1, 2$ (mol/liter)
M_N	Total number average molecular weight
M_{NS}	Desired value (set point) of total number average molecular weight
M_W	Total weight average molecular weight
P	Total growing polymer concentration of type-1 (mol/liter)
P_i	i th moment of the total number MWD of radicals of type-1
$P_{n,m}$	Concentration of growing polymer containing n units of monomer-1 and m units of monomer-2 (mol/liter)
PD	Polydispersity
Q	Total growing polymer concentration of type-2 (mol/liter)
Q_i	i th moment of the total number MWD of radicals of type-2

$Q_{n,m}$	Concentration of growing polymer containing n units of monomer-1 and m units of monomer-2 and ending in monomer-2 (mol/liter)
R_p	Copolymerization reaction rate (mol/liter-sec)
r_{ij}	Monomer reactivity ratios
T	Reactor temperature ($^{\circ}\text{K}$)
T_c	Jacket media temperature ($^{\circ}\text{K}$)
T_f	Monomer feed temperature ($^{\circ}\text{K}$)
t	Reaction time (min)
u	Monomer feed rate (liter/min)
u_i	i th manipulated variable; $i = 1$, Feed rate; $i = 2$, Jacket (or reactor) temperature
V	Reactor volume (liter)
w_i	Molecular weight of i th monomer; $i = 1, 2$ (g/mol)

Greek Letters

ΔH_{cp}	Heat of copolymerization (kcal/mol)
λ_k^d	kth moment of the copolymer total number MWD; $k = 0, 1, 2$
ρ	Density of reaction mixture (kg/liter)
ϕ	Molar ratio of monomers in reaction mixture
ϕ_f	Monomer mole ratio in feed stream
ϕ_s	Desired value (set point) of molar ratio of monomers in reaction mixture
ϕ_t	Cross termination factor

Figure Captions

- Figure 1 Styrene-acrylonitrile copolymer composition curves for different reaction temperatures.
- Figure 2 Isothermal batch copolymerization of styrene with acrylonitrile ($I_o = 0.05$ mol/l, 70°C , $f_s = 0.5$, $\phi_s = 1.0$).
- Figure 3 Open loop single input-single output control system.
- Figure 4 Closed loop single input-single output control system.
- Figure 5 Multivariable "fictitious" feedback control system for copolymer composition and molecular weight control.
- Figure 6 Open loop copolymer composition and molecular weight control system.
- Figure 7 Open loop composition control in isothermal semibatch copolymerization reactor ($I_o = 0.05$ mol/l, 70°C , $f_s = 0.5$, $\phi_s = 1.0$).
- Figure 8 Open loop copolymer molecular weight control in a semibatch copolymerization reactor ($I_o = 0.05$ mol/l, $T_f = 50^\circ\text{C}$, $f_s = 0.5$, $\phi_s = 1.0$).
- Figure 9 Open loop copolymer composition and molecular weight control in a semibatch copolymerization reactor ($I_o = 0.05$ mol/l, $T_f = 50^\circ\text{C}$, $f_s = 0.5$, $\phi_s = 1.0$).
- Figure 10 Conditioning of time varying profiles of manipulative variables of Figure 9 (A, with original feedback signal; B, with polynomial fitted signal).
- Figure 11 Open loop copolymer composition and molecular weight control in a semibatch copolymerization reactor with jacket temperature as a manipulative variable ($I_o = 0.05$ mol/l, $T_f = 50^\circ\text{C}$, $f_s = 0.5$, $\phi_s = 1.0$).