Multi-Rate Model Predictive Control of Particle Size Distribution in an Emulsion Copolymerization Reactor

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1. INTRODUCTION

Emulsion polymerization is an important process for the polymer industry that has significant advantages over bulk and solution polymerization processes. These advantages result mostly from the multiphase and compartmentalized nature of the emulsion polymerization, delivering a high versatility to product qualities. Despite these advantages, the control of emulsion polymerization reactors is very challenging due to the complexity of emulsion process and the lack of adequate on-line measurement instrumentation [1]. The control of particle size distribution (PSD), which is critical for desired end-product performance, suffers from both of these aspects. The resulting control problem is high-dimensional due to the complexity of the process and lacks fast high-resolution size distribution measurements. Although the control of the emulsion polymerization systems is a well studied topic typically involving the regulation of one or more of the lumped properties (*e.g.* moments of the distributions), the control of the distribution shape is largely an open problem. The indirect control of PSD through moments reduces the high dimensionality of the problem but regulation to complex distributions is not always possible. However, the end-use properties (mechanical, rheological, optical) of the polymer products may depend on obtaining the full distributions, particularly when the target distribution is complex and/or multimodal. For example, bimodal distributions in paint applications may be desirable for rheopectic behavior [2].

Previous simulation and experimental studies by Doyle and coworkers [3], [4] on feedback control of the full PSD revealed that the delayed infrequent measurement of PSD, the primary controlled variable, deteriorates the quality of in-batch feedback control. Availability of frequent and reliable on-line density measurements for emulsion polymerization systems motivates the use of multi-rate estimation schemes as a remedy for the infrequent PSD measurements. In this study, a multi-rate estimation scheme that utilizes the density measurements with high sampling rates is used with a model predictive controller (MPC) to control the final PSD in an emulsion vinyl acetate/butyl acrylate (VAc/BuA) copolymerization system. Principal component analysis (PCA) based model order reduction is employed to remove the high dimensionality of the control problem.

2. SEMIBATCH VAc/BuA EMULSION COPOLYMERIZATION SYSTEM

In this study, a population balance equation (PBE) model, developed by Immanuel *et al.* [5] describing the evolution of the particle size distribution in a semibatch VAc/BuA copolymerization reactor is used as the plant. The model that is summarized in Table 1 consists of a PBE coupled with necessary mass balances. The model considers size dependent growth, partitioning of the nonionic surfactants, and the average number of radicals per particle calculations via first principles. A computationally efficient numerical method that

exploits the different inherent time scales present in emulsion polymerization is used to solve the particle PBE, where the PSD is discretized into 250 finite elements of 2 nm width [6].

Table 1. PBE model for a semibatch emulsion copolymerization reactor

Mass balance equations	
Oxidizer	$d([I_w]V_{aq})/dt = -k_{d1}[I_w][Y_1^r] + v_{I_w}$
Reducer	$d([Y_2]V_{aq})/dt = -r_I k_{d1}[I_w][Y_1^r] + v_{Y_2}$
Initiator radical	$d([R_w]V_{aq})/dt = k_{d1}[I_w][Y_1^r] - V_{aq}\sum_{i=1}^{2} k_{ri}[R_w][M_i]_w - V_{aq}k_{tav}^w[R_w]\left(\sum_{l=0}^{j_{cr}-1}[P_w]^l + [R_w]\right)$
Monomers	$\frac{dM_j}{dt} = v_{M_j} - \sum_{i=1}^{2}(k_{pij}^w + k_{trij}^w)p_{wi}[P_w][M_j]_w V_{aq} - \sum_{i=1}^{2}(k_{pij} + k_{trij})p_i[M_j]_p \int_{r_{nuc}}^{r_{\max}} \bar{n}(r,t)F(r,t)dr$
Population balance equation	
Density function	$\frac{\partial}{\partial t}F(r,t) + \frac{\partial}{\partial r}\left(F(r,t)R_{growth}(r,t)\right) = R_{nuc}(r,t) + R_{coag}(r,t)$
Growth rate	$R_{growth}(r,t) = \frac{dr}{dt} = \frac{3}{4\pi r^2 \rho_p}\sum_{i=1}^{2}\sum_{j=1}^{2} k_{pij}p_i\frac{\bar{n}(r,t)}{N_A}[M_j]_p MW_j$
Nucleation rate	$R_{nuc} = R_{micellar} + R_{homo} = \sum_{l=0}^{j_{cr}-1}\sum_{i=1}^{2} e_{i,micelle}^l p_{wi}[P_w]^l C_{micelle}V_{aq} + k_{pav}^w[P_w^{j_{cr}-1}]V_{aq}$
Coagulation rate	$R_{coag}(r,t) = H(r_{uppper} - r)R_{formation}(r,t) - H(r_{cutoff} - r)R_{depletion}$
	$R_{formation}(r,t) = \frac{1}{V_{aq}}\int \beta(r',r'')F(r',t)F(r'',t)\frac{r^2}{\left(r^3-(r')^3\right)^{2/3}}dr'$
	$R_{depletion}(r,t) = \frac{1}{V_{aq}}\int_{r_{nuc}}^{r_{\max}} \beta(r,r')F(r,t)F(r',t)dr'$ where $\beta(r,r') = c_1 4\pi D_0(r+r')/W_s$
Output functions	
Weight averaged PSD	$W(r_i,t) = r_i^3 F_i \Big/ \sum_i r_i^3 F_i$ where $F_i = \int_{r_{b,i-1}}^{r_{b,i}} F(r,t)dr$
Total particle number	$N_p = N_A \sum_i F_i$ Solid content $S_c = N_A\left(\sum_i 4\pi r_i^3 \rho_p F_i/3\right)/m_T$

The actual experimental system under consideration is presented in Figure 1. The system consists of a 3L glass reactor, where the monomers, oxidizer, reducer, and the surfactant are delivered by remote setpoint metering pumps controlled by a Honeywell Plantscape digital control system. The PSD measurements can be obtained every 12 minutes by a capillary hydrodynamic fractionator, and the density measurements are available every minute from the on-line densitometer.

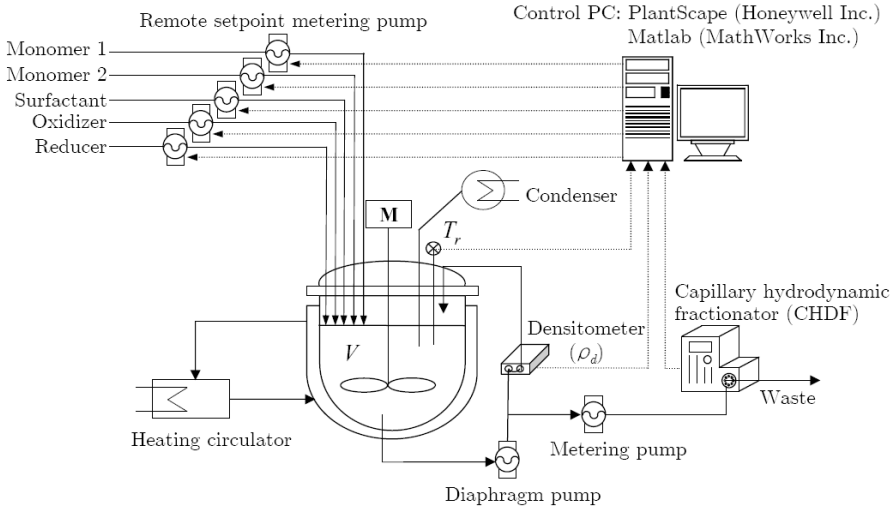

Figure 1. Schematic diagram of the experimental system for the VAc/BuA emulsion copolymerization system

3. MULTI-RATE MPC

The multi-rate MPC controller is developed as a combination of the extended quadratic dynamic matrix (QDMC) controller of [7] and the linear multi-rate MPC controller of [8]. The PBE model can be represented as a system of continuous-time nonlinear differential equations:

$$\begin{aligned} \dot{x} &= f(x,u,d) \\ y &= g(x) \end{aligned} \tag{1}$$

where x represents the system states, u represents manipulated variables and d represents unmeasured disturbances. The discrete version of the model can be expressed as

$$\begin{aligned} x_k &= F_{T_s}(x_{k-1},u_{k-1},d_{k-1}) \\ y_k &= g(x_k) \end{aligned} \tag{2}$$

where $F_{T_s}(x_{k-1},u_{k-1},d_{k-1})$ denotes the terminal vector obtained by integrating the continuous system equations one sampling time, T_s, from the initial state vector, x_{k-1}, holding the manipulated variable, u_{k-1}, and the unmeasured disturbances d_{k-1}, constant. After augmenting the model states with the disturbance model states, secondary and primary plant outputs, and primary measurement delay the state evolution equation is obtained as

$$\underbrace{\begin{bmatrix} x_k \\ x_k^w \\ y_k^c \\ y_k^{c_1} \\ y_k^{c_2} \\ \vdots \\ y_k^{c_\theta} \\ y_k^s \end{bmatrix}}_{\bar{X}_k} = \underbrace{\begin{bmatrix} F_{T_s}(x_{k-1},u_{k-1},C_w x_{k-1}^w) \\ A_w x_{k-1}^w \\ g^c(F_{T_s}(x_{k-1},u_{k-1},C_w x_{k-1}^w)) \\ y_{k-1}^c \\ y_{k-1}^{c_1} \\ \vdots \\ y_{k-1}^{c_{\theta-1}} \\ g^s(F_{T_s}(x_{k-1},u_{k-1},C_w x_{k-1}^w)) \end{bmatrix}}_{\Phi(x_{k-1},u_{k-1},x_{k-1}^w)} + \underbrace{\begin{bmatrix} 0 \\ B^w \\ 0 \\ 0 \\ 0 \\ \vdots \\ 0 \\ 0 \end{bmatrix}}_{\Gamma^w} w_{k-1}$$

$$\hat{Y}_k = \underbrace{\begin{bmatrix} 0 & 0 & 0 & 0 & 0 & \dots & H_c & o \\ 0 & 0 & 0 & 0 & 0 & \dots & 0 & H_s \end{bmatrix}}_{\Xi_k} \bar{X}_k \tag{3}$$

First, model prediction is performed by integrating the nonlinear model one time step with the information at time step $k-1$ and then the state vector is corrected with the current measurement vector

$$\bar{X}_{k|k-1} = \Phi(x_{k-1}, u_{k-1}, x^w_{k-1})$$
$$\bar{X}_{k|k} = \bar{X}_{k|k-1} + K_{f,k}\left(\hat{Y}_k - \Xi_k \bar{X}_{k|k-1}\right) \tag{4}$$

The output measurement vector, $\hat{Y}_k$, consists of the available noisy measurements of the primary and secondary outputs. The estimates of the secondary and delayed primary outputs are obtained form the state vector, $\bar{X}_{k|k}$, with the time-varying Ξ_k matrix such that

$$\bar{Y}_{k|k} = \Xi_k \bar{X}_{k|k} = \begin{cases} \begin{bmatrix} y^c_{k|k} \\ y^s_{k|k} \end{bmatrix} & \text{when primary and secondary measurements are available} \\ y^s_{k|k} & \text{when secondary measurements are available} \end{cases} \tag{5}$$

The time-varying Kalman filter gain, $K_{f,k}$, is computed by

$$K_{f,k} = \tilde{\Sigma}_k \Xi_k^T \left\{\Xi_k \tilde{\Sigma}_k \Xi_k^T + R_k\right\}^{-1}$$
$$\Sigma_k = \Phi\Sigma_{k-1}\Phi^T - \Phi\Sigma_{k-1}\Xi_{k-1}^T \left\{\Xi_{k-1}\Sigma_{k-1}\Xi_{k-1}^T + R_{k-1}\right\}^{-1} \Xi_{k-1}\Sigma_{k-1}\Phi^T + \Gamma_w Q \Gamma_w^T \tag{6}$$

where Q is the covariance matrix of the expected disturbances, and R_k is the covariance matrix of the expected measurement noise on the primary and secondary measurements. In order to obtain the steady state periodic solutions, $\tilde{\Sigma}_k$, to the above Riccati equations the multi-rate system can first be converted to a single rate system at the slower sampling time of the primary measurements. The resulting Riccati equations can be solved for the single-rate state covariance matrix, $\tilde{\Sigma}^{SR}_k$, from which the periodic solution for the multi-rate system can be obtained. The multistep-ahead MPC prediction equation is formulated such that the contribution of the future unknown input changes on the controlled outputs is linear. The prediction equation can be expressed as

$$Y^c_{k+1|k} = S^x + S^u \Delta U_k \tag{7}$$

where

$$Y^c_{k+1|k} = \begin{bmatrix} \bar{Y}_{k+1|k} \\ \bar{Y}_{k+2|k} \\ \vdots \\ \bar{Y}_{k+p|k} \end{bmatrix} \quad \Delta U_k = \begin{bmatrix} \Delta u_k \\ \Delta U_{k+1} \\ \vdots \\ \Delta U_{k+q-1} \end{bmatrix} \quad S^u = \begin{bmatrix} C^c_k B^u_k & 0 & \cdots & 0 \\ C^c_k (I + A_k) B^u_k & C^c_k B^u_k & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ C^c_k \sum_{i=0}^{p-1} A^i_k B^u_k & C^c_k \sum_{i=0}^{p-2} A^i_k B^u_k & \cdots & C^c_k \sum_{i=0}^{p-q} A^i_k B^u_k \end{bmatrix}$$
$$S^x = \begin{bmatrix} g\left(F_{T_s}(x_{k|k}, u_{k-1}, C^w x^w_{k|k})\right) - y_0 \\ g\left(F_{2,T_s}(x_{k|k}, u_{k-1}, C^w x^w_{k|k})\right) - y_0 \\ \vdots \\ g\left(F_{p,T_s}(x_{k|k}, u_{k-1}, C^w x^w_{k|k})\right) - y_0 \end{bmatrix} \tag{8}$$

The computation of the optimal control input is established by formulating and solving a quadratic program,

$$\begin{aligned} &\min_{U_k} \left\| \Lambda^y (Y_{k+1|k} - R_{k+1|k}) \right\|_2^2 + \left\| \Lambda^u \Delta U_k \right\|_2^2 \\ &s.t. \qquad U_L \le U_k \le U_U \\ &\qquad\quad \left| \Delta U_k \right| \le \Delta U_{max} \end{aligned} \tag{9}$$

The discrete system matrices at time, k, are obtained by linearization around a nominal point and using zero order hold. As this linearization yields high order model (258 states, 250 outputs) that is ill-conditioned, model order is reduction is accomplished using PCA. The reduced states x are obtained from the normalized original states, x', by an orthonormal linear transformation matrix, $\mathbf{P}$,

$$x = \mathbf{P}x' \tag{10}$$

with this transformation the reduced order system matrices are,

$$A_k = \mathbf{P}^T A_k' \mathbf{P} \quad B_k^u = \mathbf{P}^T B_k'^u \quad C_k = C_k' \mathbf{P} \tag{11}$$

Further details about the application of PCA to the current system can be found in [4].

4. RESULTS AND DISCUSSION

The overall strategy of the MPC controller employed in this study is summarized in Figure 2. The MPC controller regulates the reduced order weight averaged PSD (wPSD) and the state feedback to the controller is established by the multi-rate estimator acting based upon the discrepancy between the measurements and the estimations of these measurements. The measurements of the plant include the delayed reduced primary outputs (wPSD) every 12 minutes and the secondary measurement (density from which the solids content is inferred) every minute.

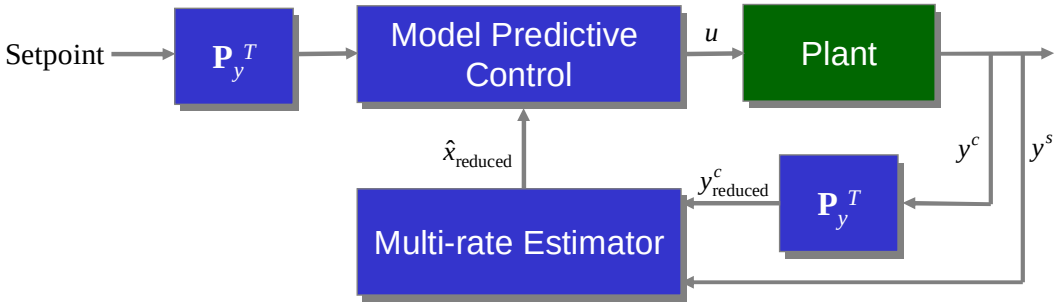

Figure 2. Overall strategy of the MPC controller

The database for the calculation of the transformation matrices were generated by introducing pseudo-random 4-level input signals with a switching probability of 7% to the plant (PBE model of the system) for 60 batches. Since the actual plant was used for the PCA rather

than experimental data, capturing the noise with a high number of principal components (PC) was not a concern and 24 PCs were chosen that explained 99.9% of the simulated database. In Figure 3, the weight averaged PSDs for the simulated database are presented and it can be seen that the majority of the endpoint distributions are bimodal resulting from a combination of homogeneous and micellar nucleation regimes.

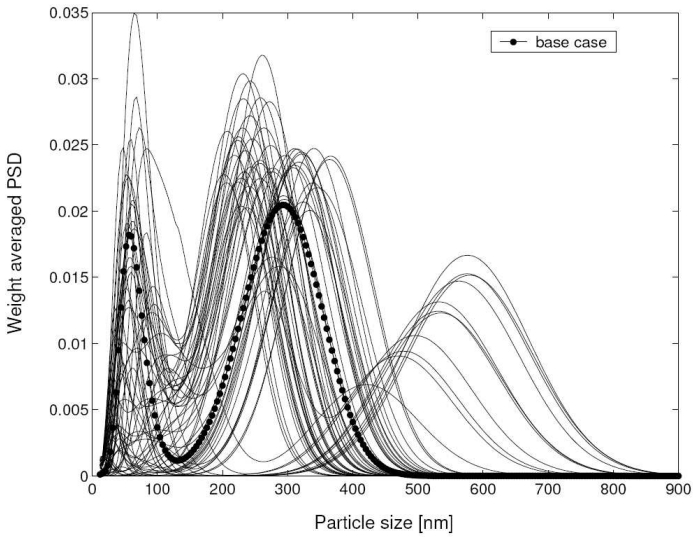

Figure 3. Weight averaged PSD at the final time generated by 4-level pseudo random input sequences for 60 batches

Although the MPC controller uses the nonlinear system equations to make the open-loop predictions, the effect of the future input changes are incorporated on the basis of a linear model for the closed-loop predictions. This approach is further motivated by the observed linear effect of the inputs on selected outputs that represent the overall behavior of the system. For example, the averaged effect of step inputs given to the nominal surfactant feed profile at different times results in linear responses for the total number of particles as shown in Figure 4.

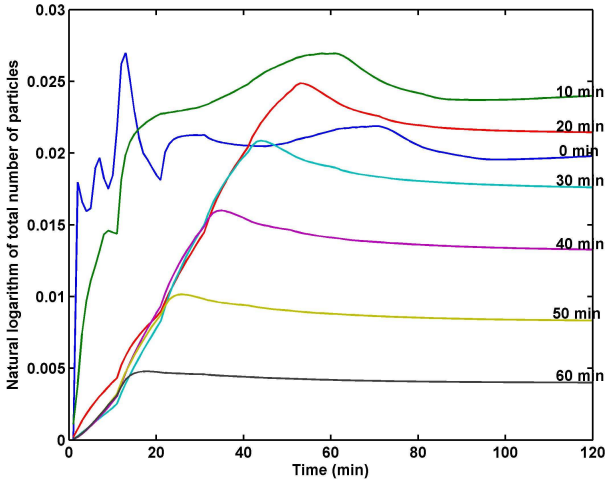

Figure 4. Reaction curves of the natural logarithm of the total number of particles to the step inputs introduced to the surfactant flowrate at different times throughout the batch.

The proposed controller was tested against a disturbance in the surfactant feed concentration (the concentration of the surfactant in the plant was 30% less than that in the controller), using VAc, BuA, surfactant, and the initiator pair of reducer and oxidizer flowrates. The MPC controller had the same weights for all the manipulated variables and the weighing of the scaled control errors was twice the weighting for the scaled inputs. The prediction horizon was set to 25 and the move horizon was set to 5. The resulting wPSD and the corresponding input moves presented in Figure 5 indicate that the designed MPC controller was successful in regulating the wPSD to the target. The controller was able to move both the primary mode and the secondary mode of the distribution close to the target with respect to the uncontrolled (open loop) distribution. Although the controller does not receive any size measurements until 13 minutes into the batch (wPSD at 1 minute), the primary mode of the bimodal distribution formed by homogenous nucleation was corrected by manipulating the initiator pair and the BuA flowrate based upon the solids content measurement before 13 minutes. The corrective action taken by the controller is more apparent on the secondary mode formed by the micellar nucleation.

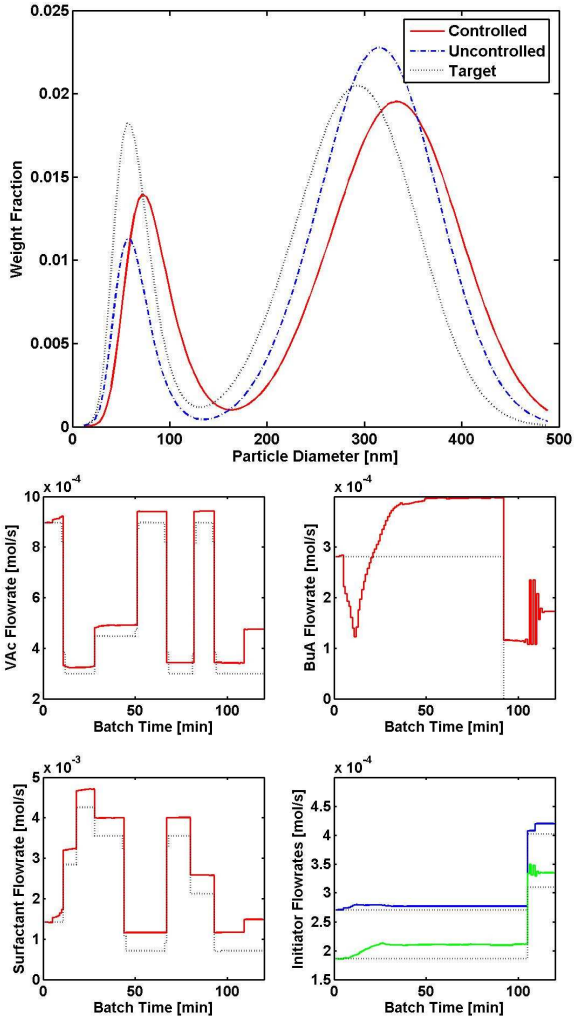

Figure 5. The performance of the controller in rejecting a -30% plant/model mismatch in the surfactant concentration and the corresponding input profiles(dotted black: nominal input trajectories, red: variation the respective input profile)

4. SUMMARY

A multi-rate MPC controller utilizing PCA-based reduced order linear models was applied to a nonlinear mechanistic PBE model of a semibatch VAc/BuA emulsion copolymerization system. The results showed that the controller was successful in regulating the system to a target PSD. The designed controller was able to act upon the frequent secondary measurements to detect the disturbances and take corrective action on the primary mode of the distribution as well as the secondary mode.

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