Control of Batch Particulate Processes

Research Proposal

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1 Introduction

1.1 Problem Statement

The scope of this research proposal is towards the model-based control of batch particulate processes (BPPs). A primary control objective in BPPs is to reach a desired distribution of product properties by the end of the batch. A new research topic in the field of BPP control, specifically full distribution control, is proposed in this report. The proposal focuses on the development of an end-point based non-linear predictive control scheme designed to meet the aforementioned primary control objective. Complementary issues such as non-linear model order reduction and dynamic optimization are also addressed.

1.2 Background

Particulate processes (dispersed-phase processes) are characterized by material domains that are comprised of a continuous phase and a dispersed phase consisting of particles. Particulate processes are essential for the manufacture of numerous high-value industrial products. The batch/fedbatch mode of operation is particularly well-suited for manufacturing such high-value products as batch recipes can be changed as necessary from batch to batch. Prototypical examples of BPPs include crystallization, polymerization (emulsion and suspension), and microbial fermentation.

Physio-chemical and mechanical properties of the end product in BPPs are dependent on the particle size distribution (PSD) of the product at batch termination. For instance, the PSD of polymer particles in a semi-batch emulsion reactor influences the polymer's rheological properties (i.e. viscosity)[1], drying characteristics, adhesion, and film-forming properties (Immanuel et al. [2008]). Therefore, PSD control is a means of inferential control for several key product properties. Furthermore, in many situations, the entire shape of the end-point PSD is influential in determining the product characteristics, signifying control based on simply averages or moments of the distribution is not appropriate. Accordingly, in many BPPs, a typical end-point objective is to achieve a target PSD at batch termination. Frequently, the final solids content (i.e. final particle concentration) is also prescribed (Immanuel et al. [2008]). Note that while the PSD is an important *morphological* property, several *molecular* properties of the end-product in BPPs are also given by distributions of other characteristics of the particles. For polymerization processes, a notable example is the molecular weight distribution (MWD). Consequently, end-point targets for these distributions may be specified as well in the control objectives. The proposal will focus specifically on PSDs as they are unique to BPPs. Moreover, the general framework for modeling other distributions is the same.

Due to the inherently complex dynamics that govern particulate systems along with the

[1]The detailed relationship between the PSD and rheology is available in Immanuel et al. [2008] and the references therein.

presence of input/state constraints in chemical processes, model predictive control (MPC) represents the natural framework for full PSD control. MPC schemes can simultaneously take into account optimality, constraints, measurement feedback, and the complex dynamics which govern the evolution of the PSD; however, in the context of full PSD control, a predictive control strategy poses numerous unique control challenges. The majority of these challenges stem from the distributed parameter nature of population balance equations (PBEs), which are used to model the PSD in particulate processes. The computationally prohibitive nature of finding solutions to PBEs steered the early research on full PSD control towards the off-line computation of optimal input trajectories that could drive a system to a desired PSD (i.e. open-loop control). Naturally, the presence of disturbances and plant-model mismatches invalidate the desired characteristics of the computed input trajectories in a real plant setting. Another area of early research on PSD control was on the design of model-based non-linear geometric controllers, which did not take into account optimality nor constraints. Furthermore, these controllers were designed using reduced order PBEs and aimed at regulating moments of the PSD as opposed to the full PSD.

Recently, motivated by the above considerations, vigorous research attention has been directed towards MPC for regulation of the full PSD. This has been facilitated by developments in the numerical methods for solving PBEs and dynamic optimization (DO) problems, model order reduction (MOR) techniques, state estimation methods, and computer and measurement technologies. The purpose of this proposal is to present a research topic towards MPC of the full PSD in BPPs.

1.3 Organization of Proposal

A review of the literature related to the model based control of BPPs is presented in the next section. Then, in Section 3, the research proposal is presented.

2 Literature Review

The literature review will be broken down into four sections: population balances (Section 2.1), numerical solution methods for population balance equations (Section 2.2), model order reduction techniques (Section 2.3), and model-based control of BPPs (Section 2.4). The review will frequently refer to the two prototypical examples in chemical engineering literature of BPPs for which PSD is a key control variable, emulsion polymerization and crystallization.

2.1 Population Balances

Population balance equations (PBEs) provide the natural framework for mathematically modeling PSDs. A PBE may be regarded as representing a number balance on particles of a particular state (Ramkrishna [2000]). The elements of the particle state vector, $[r\ z]$,

are: the internal states of the particle, r, and the external states, z, which designate the location of the particle in the physical space (i.e. $z \in \mathbf{R}^3$). The internal states provide a quantitative characterization of the particle's distinguishing characteristics. The choice of the internal state variables depends on the particular application. Naturally, for modeling the PSD, size is taken as the internal co-ordinate[2]. Assuming spatial homogeneity (say from a well-mixed system) and applying a dynamic balance on the number of particles with state r, the general non-linear partial integro-differential equation shown in Equation 1 is obtained. This equation describes the rate of change of the PSD, $n(r,t)$.

$$\frac{\partial n(r,t)}{\partial t} + \frac{\partial [G(r,x,t)n]}{\partial r} = w(n,r,x) \tag{1}$$

where x is a vector of continuous phase state variables (such as temperature, concentration, etc.). The function, $G(r,x,t) \equiv \frac{dr}{dt}$, is the growth rate of particles of size r, and the function, $w(n,r,x)$, refers to the net rate of introduction of new particles of size r into the system through aggregation (coagulation), breakage, and/or nucleation[3]. The PBE in Equation 1 must be supplemented with initial and boundary conditions for $n(r,t)$. The boundary condition, $n(0,t)$, is ordinarily related to the nucleation rate. To complete the model of the BPP, Equation 1 must be coupled with dynamic models (i.e. mass and energy balances) for the continuous phase state variables. These models can be given by the following general non-linear integro-differential equation system:

$$\dot{x} = f(x,u) + \int_0^{r_{max}} a(n,r,x)\,\mathrm{d}r \tag{2}$$

where $f(x,u)$ and $a(n,r,x)$ are non-linear vector functions, u denotes the input vector, and r_{max} is the maximum size of the particles. The integral term, $\int_0^{r_{max}} a(n,r,x)\,\mathrm{d}r$, accounts for the mass and heat transfer from the continuous phase to all particles in the population. The coupled system, Equations 1–2, provide the complete description of the PSD in a BPP.

2.2 Numerical Solution Methods for Population Balances

The coupled nature of the complete BPP model is not amenable to analytical solutions for the PSD. Extensive research effort has been invested in efficient numerical methods to solve the PBE in Equation 1. A brief overview of the most widely applied numerical methods is presented in this section. A comprehensive review of all the numerical methods can be found in Kiparissides [2006] and Ramkrishna [1985]. The numerical methods reviewed in this report are categorized into finite difference methods, the method of weighted residuals, and bin-based methods.

[2]The PBE for modeling other distributions, such as the MWD, follows the general framework as for the PSD but may have different internal coordinates.

[3]Breakage and aggregation phenomena give rise to integral terms in Equation 1.

2.2.1 Finite Difference Methods

The least sophisticated way to numerically solve Eq. 1 for $n(r,t)$ is through finite difference methods (FDMs). In FDMs, appropriate ranges for the particle size and time domains are chosen, and each domain is discretized by a finite number of points or nodes (Chapra and Canale [1998]). Next, finite difference approximations to the partial derivatives in Eq. 1 are made at each of the spatial and temporal discretization points, resulting in an entirely algebraic set of equations. The accuracy of the solution is dependent on the number of nodes (interval spacing) and the finite difference equations used for approximating the derivatives. Second-order approximations have been used with success for simulation purposes in literature (i.e. See Shi et al. [2005]). The number of nodes for an accurate solution can be found when the solution becomes nearly independent to the number of nodes (Shi et al. [2005], Chiu and Christofides [1999]). Another solution technique that can be classified as a FDM involves the discretization of only the particle size domain, replacing the spatial derivatives with finite difference approximations at each node, and solving the resulting ODEs at each node in the time domain.

Although simple to implement, FDMs are subject to spurious oscillations and numerical dispersions in the solution due to truncation errors introduced from approximating derivatives with finite difference equations (Immanuel and Doyle III [2003a], Crowley et al. [1999]). This can be circumvented by refining the discretization grid at the expense of high computation times. The discretization interval lengths must also chosen carefully in FDMs so as to avoid numerical instability (i.e. error amplification) (Chapra and Canale [1998]).

2.2.2 Method of Weighted Residuals

To avoid the numerical oscillations and dispersion associated with FDMs, the method of weighted residuals (MWR) has been popular for solving PBEs. The central idea in the MWR is to approximate the population density function over the entire domain of interest or more commonly over finite elements (FEs) in the domain by a finite series of N orthogonal basis functions, $\phi_v(r)$, with time varying coefficients, $a_v(t)$, as shown in Equation 3 (Ramkrishna [1985]).

$$n(r,t) = \sum_{v=1}^{N} a_v(t)\phi_v(r) \tag{3}$$

Note that the convergence to the true solution can be guaranteed as $N \to \infty$ as shown in Chiu and Christofides [1999]. The approximating series in Equation 3 can be substituted into the PBE (Equation 1), yielding a residual. This residual expression is then multiplied by N individual weighting functions, ψ_v, and each of the residual expressions are integrated from 0 to r_{max}. The end result of this series of steps is a system of N ODEs for the time varying coefficients. Depending on the choice of the weighting and basis functions, the MWR may be classified as being either:

1. *Galerkin's Method* (Ramkrishna [1985, 2000]): The weighting and basis functions are chosen to be identical.

2. *Orthogonal Collocation* (Ramkrishna [1985, 2000]): The basis functions are chosen to be low-order Lagrange interpolating polynomials while weighting functions are chosen to be Dirac delta functions centered around collocation points.

3. *Method of Moments* (Hulburt and Katz [1964]): **Case I:** The basis functions are chosen to be Laguerre polynomials while the weighting functions are chosen to be $\psi_v(r) = r^v$. Hulburt and Katz [1964] show the choice of Laguerre polynomials as the basis functions can be exploited to express the time varying coefficients in terms of moments of the PSD. Thus, ODEs are yielded for the moments of the PSD. Note that the v-th moment of a PSD, μ_v, is defined as:

$$\mu_v = \int_0^\infty r^v n(r,t)\, \mathrm{d}r \quad v = 0, 1, \ldots \infty \tag{4}$$

   Case II: Under certain circumstances, ODEs for the moments can be directly generated from the PBE. If the growth rate is linear with respect to r and/or if the aggregation rate (if present) is a constant, a set of ODEs can be obtained for a finite number of the PSD moments (Hulburt and Katz [1964], Ramkrishna [2000]). These conditions are known as the closure conditions. If closure conditions are satisfied, the PBE can be multiplied by r^v and integrated from 0 to ∞ to yield ODEs for a finite number of moments.

Galerkin's method is the most suitable choice when the *global* solution can be approximated by a relatively small number of basis functions (Ramkrishna [1985]). Typically, approximating $n(r,t)$ over finite elements is preferred to avoid the use of high order basis functions (Immanuel and Doyle III [2003a]). When finite elements have been used, orthogonal collocation has been the most widely adopted MWR in the open literature (for example, see Rawlings and Ray [1988], Crowley et al. [2000])

Although moments of a PSD have physical significance (for example, the 0-th moment represents the number of particles), the construction of the full PSD from a finite number of its moments is faced with numerical instability as reported in Immanuel and Doyle III [2003a]. The authors point out the PSD should be relatively smooth and ideally unimodal for reconstruction from its moments. Otherwise, the number of moments for an accurate reconstruction can be high. For instance, a maximum entropy approach was shown in Dokucu et al. [2008b] to be effective for re-constructing a bimodal PSD from its moments, but a high number of moments (10) was necessary.

2.2.3 Bin Based Methods[4]

Bin based methods are regarded as efficient alternatives to the methods described in Section 2.2.2. In bin-based methods, the contiguous size domain is partitioned into various size ranges or *bins*, which interact collectively with particles in other bins (Kumar and Ramkrishna [1997]). Each bin represents a class of particles defined by the size interval of the bin, $[r_{b_{j-1}}\ r_{b_j}]$, where j is an integer indexing the bins and r_b denotes the upper boundary of bin j. Additionally, each bin is given a representative size (i.e. the midpoint) for use in evaluation of the size dependent parameters. The population of the j-th bin can be defined as $F_j = \int_{r_{b_{j-1}}}^{r_{b_j}} n(r,t)\ \mathrm{d}r$ and bin population balances can be formulated for each bin (Ramkrishna [2000]). In general, the balances must be written such that moments of the number density function (i.e. the total mass and number of particles) are preserved.

Three bin-based methods, which Kiparissides [2006] reports to be more efficient than other candidates in his review paper, are given by Hounslow et al. [1988], Litster et al. [1995], and Kumar and Ramkrishna [1997]. The bin-based methods described in Hounslow et al. [1988] and Litster et al. [1995] differ from that in Kumar and Ramkrishna [1997] in the way balances for each bin preserve the moments. The three techniques additionally differ in their discretization techniques. In Kumar and Ramkrishna [1997], any user-defined discretization of the size domain can be employed while in Hounslow et al. [1988] and Litster et al. [1995], the grid must be geometric. A geometric grid can result in a coarse discretization, which, in turn, can lead to inaccurate solutions.

A highly efficient, two-tier bin-based method that has been adopted in several recent BPP control studies is presented by Immanuel and Doyle III [2003a]. In this approach, the authors first derive the population balance for bin j as:

$$\frac{dF_j}{dt} + F_j \frac{dr}{dt}\Big|_{r_{b_j}} - F_{j-1}\frac{dr}{dt}\Big|_{r_{b_{j-1}}} = \delta_1 \mathfrak{R}_{nuec}(r,t) + \int_{r_{j-1}}^{r_j} \mathfrak{R}_{coag}(r,t)\ \mathrm{d}r \tag{5}$$

where $\mathfrak{R}_{nuec}(r,t)$ is the nucleation rate and δ_1 is the Kronecker's delta function (i.e. $\delta_q = 1$ if $q = 1$ and $\delta_q = 0$ if $q \neq 1$). The term $\int_{r_{j-1}}^{r_j} \mathfrak{R}_{coag}(r,t)\ \mathrm{d}r$ must be given special consideration as it is typically expressed as a double integral. This can be simplified to a pure algebraic term involving only populations from other bins using the methods shown by Immanuel and Doyle III [2003a] or Kumar and Ramkrishna [1997]. The solution procedure in Immanuel and Doyle III [2003a] is decomposed into a two-tier hierarchical approach. In the first tier, at a given initial time, the PSD is held constant and rates of growth, nucleation, and aggregation are calculated at several sub-intervals within a given time interval. In the second tier, populations in all the bins are updated according to the calculated rates, and the time step is finally advanced. Immanuel and Doyle III [2003a] report the solutions with this method show good agreement with orthogonal collocation at a fraction of the computational cost. Furthermore, the solution times have been on the order of seconds, suggesting this two-tier solution method has utility in a MPC framework. By comparison, Crowley et al. [1999] report

[4]Bin-based methods that consider simultaneous nucleation, growth, and aggregation terms are considered as these phenomena are all relevant to the present discussion.

solution times on the order of minutes when using the bin based solution method proposed by Kumar and Ramkrishna [1997] for a similar polymerization model used in Immanuel and Doyle III [2003a].

2.3 Model Order Reduction

The recurring theme among the numerical methods presented in Section 2.2 is the transformation of the semi-infinite dimensional partial differential equation (PDE) in Equation 1 to a finite dimensional ODE system; therefore, the methods in Section 2.2 may be viewed as model order reduction (MOR) techniques. However, the resulting dynamic system may be of a high order and/or ill-conditioned. Due to these possibilities, stiff integrators are necessary and computation times may be too long to permit finding PBE solutions in real-time, necessitating the need for further MOR. In this section, the MOR techniques that have been applied to the high order ODE system are reviewed.

Given a high dimensional system, the purpose in MOR is to derive a low order model that accurately captures the dominant system dynamics while preserving input-out behavior. This can be done through a projection of the full order states onto a state space of reduced order. If the reduced space basis is chosen appropriately, the relevant system dynamics can be expressed with a reduced number of states. The literature database regarding general MOR techniques, specially methods for linear systems, is vast and a complete review of this topic is not given here. Instead, non-linear MOR techniques that have been applied with success in the context of BPPs and PBEs are considered.

Principal component analysis (PCA) has been applied by Park and Doyle III [2004] and Dokucu et al. [2008b,a] to reduce the high-order dynamic model resulting from the two-tier solution approach described in Section 2.2.3. The states of the high order model that are projected onto the reduced space in these cases were the integral amounts of the PSD for each bin (i.e. the bin populations). To use PCA as a MOR technique in the context of batch processes, a time-varying data set covering the reachable region of the system is first generated using the high order non-linear model. Then, at every sample instance of the batch, the principal components (PCs) of the data, which form the basis of the reduced space, and orthonormal transformation matrices, which are used to project the data onto the reduced space, are calculated. In Park and Doyle III [2004] and Dokucu et al. [2008b,a], the transformation matrices are used to reduce the order of a linear time varying model of the batch, which is obtained by linearizing the non-linear model along the nominal trajectory. The number of states in these studies were reduced to less than 10% of the original order.

Chiu and Christofides [1999] describe an approximate inertial manifold (AIM) approach to reduce the order of the ODE system resulting from applying the MWR. Recall in the MWR, the PSD is approximated as a series of orthogonal basis functions with time varying coefficient. This was an underlying assumption of the results in Chiu and Christofides [1999], which becomes restrictive if the MWR is not used to solve the PBE. Chiu and Christofides [1999] explain if trajectories of a high order system evolve exactly on manifold that is in a subset of original state space, then the system can be exactly described by a lower order

system. In the AIM approach, an approximation to such an inertial manifold is sought on which the dominant dynamics of the system evolve. However, it is first necessary to rigorously establish the existence of such a manifold, but this problem remains unresolved. Nevertheless, to check for the likely existence of this manifold, Chiu and Christofides [1999] compared trajectories of the first few moments of the PSD as predicted by a reduced order moments model with trajectories predicted by the full distributed parameter model. Because these trajectories were observed to be nearly superimposed, Chiu and Christofides [1999] concluded the dynamics of the system were governed by a few degrees of freedom (i.e. low dimensional), allowing derivation of a low order ODE system.

2.4 Model-Based Control of Particulate Processes

In this section, the research contributions towards the model-based control of BPPs are reviewed. First, control-theoretic properties, such as controllability, reachability, and observability, of particulate systems are reviewed.

2.4.1 Controllability, Reachability, and Observability

The models for BPPs preclude rigorous controllability and observability analysis because of the distributed parameter nature of the PBE and the coupling of the PBE with continuous phase ODEs. Unsurprisingly, controllability and observability results have been mainly case specific with some researchers adopting simplified models or programming-based methods to arrive at general conclusions. A few key general results are provided in this section.

Controllability and Reachability
The seminal studies on controllability analysis of particulate processes were by Semino and Ray [1995a,b]. By simplifying the PBE, they provided key insights into the controllability of continuous emulsion polymerization and crystallization processes through manipulation of feed concentrations. For instance, controllability was strongly influenced by constraints on the manipulated inputs and by the specific controller structure (i.e. PI or PID). Furthermore, with regard to emulsion polymerization, in Semino and Ray [1995b], two manipulated inputs with opposing effects (i.e. inhibitor and initiator feed concentrations) were found to expand the regions of controllability in the constrained case. Semino et. al also identified combinations of controlled and manipulated variables for approximate/weak controllability[5] in polymerization and crystallization processes. These results guided variable selections for several future control studies.

The assumptions used by Semino and Ray [1995a,b], namely particle age being the lone internal co-ordinate, limit the extent to which the results can be applied to realistic BPPs. Wang and Doyle III [2004] propose a systematic programming-based approach to define reachability of a BPP to a target PSD. Specifically, ϵ-reachability to a desired PSD is defined through

[5]Weak/approximate controllability requires that the initial state be taken to within an arbitrarily small neighborhood of the final state by the final time

a DO problem as follows. If there exists a feasible input trajectory such that the desired PSD can be reached within some defined tolerance, ϵ, by batch termination, the desired PSD is considered ϵ-reachable. In their framework, Wang and Doyle III [2004] demonstrate a means for characterizing a target PSD, n^{target}, in a finite dimensional parametric space by approximating it as a series of Gaussian distributions depending on the number of modes. For example, for a unimodal PSD, $n^{target} = k_1 \exp(-\frac{(r-a_1)^2}{\sigma_1})$ where k_1, σ_1, and a_1 are the amplitude, variance, and mean of the distribution. Having established this characterization, Wang and Doyle III [2004] define the reachable region as consisting of the variances, means, and amplitudes that define the distributions which can be reached by admissible control action by batch termination. The sensitivity of reachability regions to the control vector parametrization, batch duration, and parametric errors were also explored by Wang and Doyle III [2004]. The sensitivity to the batch duration was found to be play an important role in determining the appropriate time to the make the correction in a mid-course correction control strategy for end-point PSD control, which is described later. Two additional general conclusions reached by Wang and Doyle III [2004] were that the growth rate and batch times are the limiting factors for reachability for unimodal PSDs and the reachable region may be robust against uncertainties in specific parameters.

Immanuel et al. [2008] performed a feedback controllability assessment of bimodal PSDs in emulsion polymerization using logical arguments and illustrated the limitations of a control system with PSD as a controlled variable. Assuming the PSD measurements can be made periodically but with delay, Immanuel et al. [2008] identify classes of PSDs and circumstances which are likely to lead to feedback uncontrollable situations. Although no quantitative guidelines are derived, it is argued that PSDs with overlapping modes or systems in which there is not sufficient time for a PSD measurement between nucleation events (i.e. all nucleation events occur before the first PSD measurement becomes available) are likely to be uncontrollable situations. For these in-batch uncontrollable cases, Doyle et al. [2003] and Immanuel et al. [2008] stressed the need for batch-to-batch iterative PSD control (recipe optimization/adjustments between batches) using detailed models of the process. The general methodology behind batch-to-batch control is given in Immanuel et al. [2008] with details in the references therein. Of course relying solely on a batch-to-batch control strategy will not compensate for new disturbances that are affecting the batch being run currently. Additionally, due to the possibility of carryover of particles and other impurities from batch-to-batch, within-batch control remains necessary.

Observability

State estimators play a key role in any MPC scheme because they establish an information feedback from the process to the model to account for modeling errors. Advancements in measurement technologies have lead to useful sensors for key process variables in BPPs, which, in turn, have made state estimation and online feedback control of the full PSD more feasible goals. Rawlings et al. [1993] review the technology for direct PSD measurement. Among the available technologies, capillary hydrodynamic fractionation (CHDF) has become the most viable PSD measurement technique and been employed in several experimental studies in literature. However, CHDF measurements are subject to $12 - 20$ minute delays and require an appreciable solids content before being able to accurately detect the

particles (Immanuel et al. [2008]). Another key process variable, specifically in emulsion polymerization processes, is the density of the reaction mixture for which densitometers may be used for measurement. In contrast to CHDF measurements, densitometers do not have any appreciable measurement delay. The total solids content, which is sometimes specified as a controlled variable in BPPs, can be inferred from density measurements and feed rates using a soft sensor approach (Dokucu et al. [2008a,b], Immanuel and Doyle III [2003b], Immanuel et al. [2003]).

A host of studies have illustrated the successful application of EKFs to batch processes (For example, see Crowley and Choi [1998], Dimitratos et al. [1989], Kozub and Macgregor [1992], De Valliere and Bonvin [1989], Nagy and Braatz [2003]). Naturally, the observability of the states in these studies depend on the particular system under investigation, but in all cases (even in the case of MWD control in Crowley and Choi [1998]), the system under investigation was of a relatively low order in comparison to a model describing the evolution of a PSD. Moreover, the state estimation schemes were designed for single rate systems.

In response to different time scales of measurement availability, multi-rate estimation schemes using cascaded EKFs have been addressed in the control literature (Lee et al. [1992]). The advantage of these schemes is that feedback corrective action can be taken at the smaller sampling interval of the system (i.e. at the frequency of the fast measurements). For batch emulsion polymerization processes, multi-rate schemes have been used with success in an experimental setting by Dokucu et al. [2008a,b] for PSD control and Ellis et al. [1994] for MWD control . In the latter, MWD measurements were available periodically with delay while temperatures and conversion were measured with high frequency. However, the order of the ODE system considered by Ellis et al. [1994] does not compare to that of BPPs in which PSD is the main control variable. The studies by Dokucu et al. [2008a,b] are more relevant to this proposal. To facilitate the EKF design, Dokucu et al. [2008a,b] first reduced the high order non-linear ODE model (obtained from the two-tier solution approach) to a reduced order (via PCA) linear time varying model. Frequent density measurements were used to estimate the *reduced* states of the linearized system. This indicates observability of the reduced system from the density measurements. The PCA based MOR was a critical requirement for this observability as Dokucu et al. [2008a] note that the relationship between the solids content (inferred from density) and reduced states is better defined than the relationship between the solids content and the full set of states of the system.

Another approach to state estimation in the full PSD control problem has been to combine a soft-sensor approach with hardware sensors (i.e. CHFD). Alhamad et al. [2005] utilized a soft-sensor to circumvent the low frequency and delay of PSD measurements. Specifically, the relevant operating conditions were input into a detailed dynamic model which is then subsequently integrated to provide estimates of key quantities such as the PSD.

In crystallization control problems, in which reduced order moments models are sufficient for controller synthesis, extended Luenberger observers (ELOs) can be used to estimate the states of the reduced order model based on readily available process measurements, such as concentration and temperature (Shi et al. [2005]). As building an observer with guaranteed convergence properties is a difficult task for non-linear systems, Shi et al. [2005] chose the

observer gains via open-loop simulations and then compare state profiles as predicted by the model and those estimated by the observer. It was verified that the choice of observer gains resulted in state estimate convergence to the true states for the case of no mismatch and sufficiently close convergence with parametric uncertainty.

2.4.2 Control Studies of Batch Particulate Processes

In spite of the rich literature on the modeling of PSDs through PBEs and solution strategies for PBEs, there is relatively much less reported work on PSD control. First, a remark regarding the available literature on PSD control in batch crystallization processes is made. While a host of crystallization control studies with model based control designs are available (See Shi et al. [2005], Chiu and Christofides [1999] and the references therein), these works have limited utility for the research proposal in this report. This is because the controller designs in these studies are based on low order moments models[6]. This is motivated by the fact that the regulation of moments as opposed to full PSD control is the primary control objective in crystallization processes[7]. In this section, control based on moments is not considered; instead, the literature on **full PSD control** is reviewed.

Crowley et al. [2000] were among the first to address the control of the full PSD in a batch emulsion polymerization using a detailed dynamic model. Orthogonal collocation was applied as the solution method for the PBE. Crowley et al. [2000] solved the open loop optimization problem (a DO problem) to determine the optimal input/state trajectories that can drive the polymerization process to a target PSD by the end of the batch. A notable result in Crowley et al. [2000] was that a min-max norm, tied to the distribution modes, was the most appropriate metric in determining minimizing deviations from a target bimodal PSD. Basically, the maximum deviations of the two modes of the predicted PSD from the target PSD were minimized. Being only an introductory study, Crowley et al. [2000] limited the optimization to a regime of the batch time (30 minutes) during which surfactant concentration profiles had the largest effect on the PSD. Nevertheless, this was the first successful simulation attempt at matching a full PSD to a target distribution. Similar open-loop control trajectory generation are also presented by Immanuel and Doyle III [2002] and Immanuel et al. [2003].

In Park and Doyle III [2004] and Dokucu et al. [2008a], classical non-linear quadratic dynamic matrix control (QDMC)[8] is employed to track desired PSD trajectories for the duration of the batch. The model used for the prediction of the controlled output due to future unknown input changes is a reduced order, time varying linear model referenced earlier in Section 2.3. In Dokucu et al. [2008a], the output feedback control problem was considered, calling for the application of the multi-rate estimation scheme described in Section 2.4.1. Robustness

[6]Note that the PBEs in typical crystallization models satisfy the necessary closure conditions required to derive a moments model.

[7]For instance, in Shi et al. [2005], the main control objective is the maximization of the volume averaged crystal size at the final time which is a function of the fourth and third moments of the PSD.

[8]The original reference for QDMC is given in Garcia [1984] while Gattu and Zafiriou [1995] study observer based QDMC.

to several disturbances was verified through experiments. Meanwhile, Park and Doyle III [2004] considered the full state feedback problem and their robustness studies were restricted to simulations. In both works, MOR was performed using PCA prior to controller synthesis.

In an attempt to avoid the use of high order deterministic models, Park et al. [2004] used a data-based model predictive scheme for feedback control of the PSD. A low-order partial least squares model (PLS) is used for PSD predictions. Specifically, the PLS model uses the previous values of the manipulated variables and states and candidate input trajectories to predict the shape of the end-point PSD. This control scheme was able to regulate the output distribution to the target; however, the control design only utilized the infrequent PSD measurements, which reduced the effectiveness of the algorithm.

In Alhamad et al. [2005], QDMC with a step response model and soft/hardware sensor for state estimation are employed to track temporal profiles of the particle size polydispersity index (PSPI) and the number average molecular weight, which are related to the PSD and MWD, respectively. In contrast to the time varying linear model utilized by Park and Doyle III [2004] and Dokucu et al. [2008a], Alhamad et al. [2005] adjust the step response model coefficients at each sampling instance to account for non-linearities. This is done through making unit step changes in a non-linear model of the process at each sampling instance. Peterson et al. [1992] provide another way to account for process non-linearities in QDMC involving adjustments to the predictions from the step response model. Specifically, at each execution, the controlled output trajectory is predicted by a deterministic non-linear model after initialization at plant conditions and the model predictions are subsequently updated by a disturbance vector that accounts for nonlinearities.

Dokucu et al. [2008b] regulated the end-point PSD in an experimental polymerization process by regulation of the number of particles and total solids content via two simple PID controllers (a 2 x 2 system). Although this control scheme was satisfactory for its simplicity, there were noticeable off-sets for both the end-point PSD as well as for the two controlled variables, the number of particles and solids content. Note that the number of particles can be inferred from density and CHDF measurements as explained by Immanuel and Doyle III [2003b].

A mid-course correction strategy aimed at driving a batch emulsion polymerization to a desired end-point has been shown to be effective by Flores-Cerrillo and MacGregor [2002] and Yabuki and MacGregor [1997][9]. Mid-course corrections are particularly effective when quality samples are taken and analyzed only taken at a few discrete points during the batch, which is often the case in industry. The main idea behind these mid-course corrections is to utilize measurements taken throughout the batch and those obtained from one or more samples at specified times in the batch to predict the final properties using a deterministic or empirical regression model. If the prediction falls outside of pre-defined no-control zone, then a correction is made to the nominal operating policy according to the underlying phenomena in the system. Flores-Cerrillo and MacGregor [2002] combined a mid-course correction strategy with batch-to-batch adaptation to improve the empirical models used for end-point

[9]In Yabuki and MacGregor [1997], the average number molecular weight was controlled not the full PSD, but the methodology in both studies were similar.

predictions.

In Dokucu et al. [2008b], the size of the full PSD control problem was reduced by only considering regulation of the moments of the PSD. Dokucu et al. [2008b] first determined a suitable number of moments required to accurately reconstruct the desired end point PSD in an emulsion polymerization. Next, these moments are calculated off-line of the nominal PSD trajectory at each sampling instance. The moment trajectories (along with desired profiles for the solids content and total number of particles) were then tracked using QDMC. In the presence of disturbances, moment trajectories were shown to be well regulated, but off-sets remained for the end-point PSD and total solids content. Among the three control strategies studied by Dokucu et al. [2008b], the 2 x 2 PID structure for tracking solids concentration and total particles, moments regulation using QDMC, and PSD regulation using QDMC, PSD regulation exhibited the greatest robustness to disturbances (i.e. least offset in the final PSD in the presence of disturbances).

3 Research Proposal

The research proposal is presented in this section. First, in Section 3.1, the motivation behind the proposed topics is given followed by the main research objectives. The scope of the proposal is then defined in Section 3.2. In Section 3.3, the methodologies for meeting the research objectives are outlined. Finally, the research plan is sketched with a particular case study in mind.

3.1 Research Opportunity and Objectives

In the past, work on full PSD model predictive control for BPPs has been limited in the open literature. In addition to an absence of measurement technologies for the PSD, the semi-infinite dimensional nature of PBEs were the main contributing factors for this trend. Recall that while the solution methods discussed in Section 2.2 transform the infinite-dimensional system into a system of ODEs, the dynamics of the system can be of high order and\or ill-conditioned, prohibiting the synthesis of low-order, practically implementable controllers. For these reasons, the majority of the early research on model-based control for BPPs focused on the regulation of moments while the control of the entire PSD remained an open problem. Note that moments regulation is a suitable control objective for numerous batch crystallization processes and in some polymerization processes[10].

The first work towards the control of the entire PSD was attempted by Crowley et al. [2000] where open loop control policies that could achieve a target PSD were computed. For this work and the majority of the subsequent work on recipe optimization, the MWR was first applied and the resulting system of ODEs was used within a dynamic optimization (DO)

[10]For example, if the main control objective in the polymerization process is simply to achieve a target MWD, a low dimensional moments models can be utilized. See Kiparissides [2006] for an example.

framework to calculate optimal control policies. Naturally, the implementation of the calculated input profiles on the plant without any in-batch feedback would not result in the desired end-point PSD due to unavoidable disturbances and model-mismatch. Flores-Cerrillo and MacGregor [2002] responded to this by developing a mid-course correction control strategy. The critiques of this control strategy are that the end-point PSD variability can still be significant even after correction, the time and nature of the correction is system specific, and major disturbances may not be correctable through one or two corrections. Most recently, with advancements in the technology for PSD measurement, the full PSD control problem has been approached as a PSD tracking problem. The disadvantages of utilizing tracking controllers is that process noise and disturbances can render the tracking problem infeasible. In other words, noise or disturbances in the system can render the desired PSD profiles (i.e. set points) un-trackable, leading to offsets for the end-point PSD.

The deficiencies of the control structures currently used for full PSD control leave some open research opportunities related to the choice of the control scheme. The objective of this proposal is to achieve tighter PSD control through the application of existing batch process control strategies. Two such strategies that could possibly lead to more robust and tighter PSD control are:

1. Removal of local tracking controllers in favor of an end-point based non-linear predictive controller that incorporates measurement feedback directly in the computation of the control action. The objective of this controller at each sampling instance would be to drive a predictive model to a desired PSD by batch termination in a DO framework. The first step of the calculated input trajectory would then be directly implemented on the process, and the DO problem can be re-solved at the next sampling instance following re-initialization at plant conditions. This describes a shrinking horizon non-linear model predictive control (NMPC) structure that has yet to be considered for full PSD control.

2. Maintaining PSD tracking controllers but periodically updating PSD set-point (SP) profiles sent to these tracking controllers. The PSD SP updates would be based on the solution of a DO problem that: (1) utilizes a non-linear model of the plant, (2) incorporates plant measurements, and (3) is formulated to drive the non-linear model of the plant to the desired end-point PSD. Through periodically taking into account plant measurements in the SP updates, tracking feasibility would be improved as errors in the model used to compute the original SP profiles get taken into account to some extent. The multi-rate nature of measurements in BPPs, specifically in emulsion polymerization processes, can be exploited in meeting this objective. For instance, the PSD SP profiles could be updated at every sampling instance primary measurements, such as the PSD itself, become available.

The two proposed control structures are illustrated in Figures 1–2.

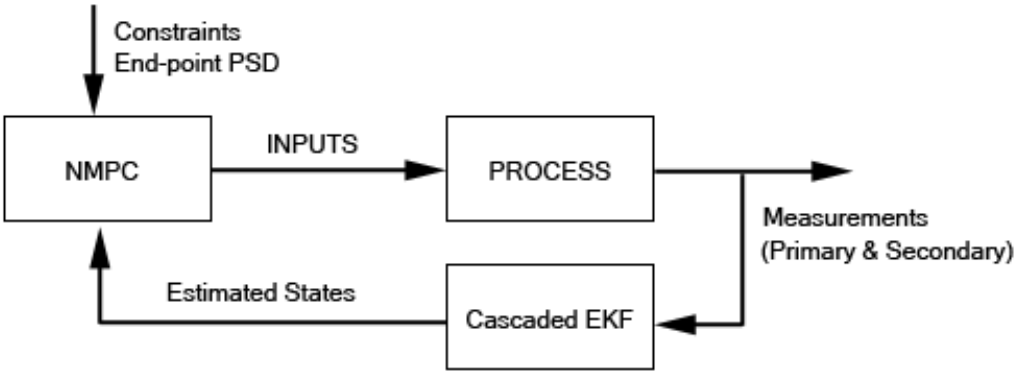

Figure 1: Overview of NMPC control structure

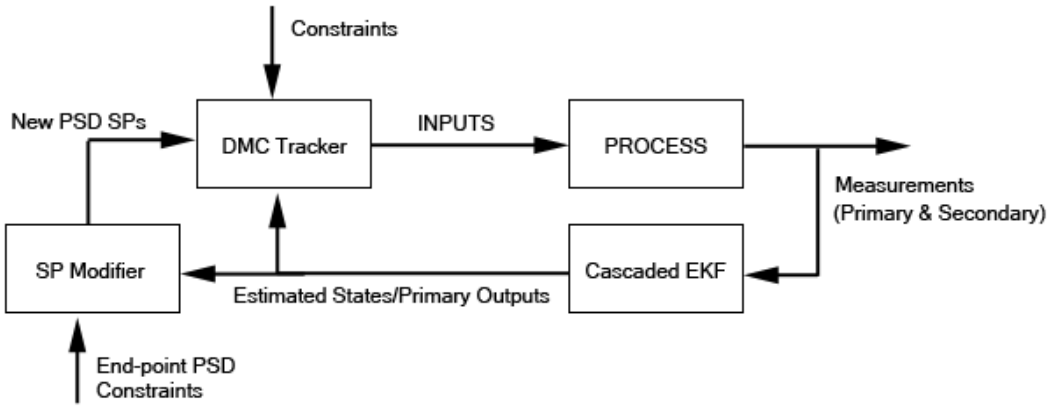

Figure 2: Overview of SP modification structure

Note that measurement incorporation in both strategies is achieved via a cascaded EKF (See Section 2.4.1) scheme in the diagram, but this is not the only state estimation strategy that may be used. The estimated states are subsequently used to initialize the DO problem in the NMPC/SP modifier blocks. The main hypothesis to be tested is if computation times required for solving the DO problem in both schemes are sufficiently low to allow for the real-time application of both two control structures. This research proposal will outline the necessary steps and methodologies for the development of the two control structures with particular attention given to reducing the computational requirements of the DO problem. First, the scope of the proposed research is defined.

3.2 Scope of the Proposed Work

This research proposal pertains to full PSD control of a generic BPP with frequent and infrequent measurements. A generalized dynamic model of a BPP was presented earlier in Equations 1 – 2. The bin-based two-tier solution approach reviewed in Section 2.2.3

is proposed as the means to transform the PBE into a system of ODEs for its reported computational efficiency. Using Equation 5, a generalized state space representation of the BPP system can be given by:

$$\dot{x}_T = f_T(x_T, u) \tag{6}$$

$$y_s = g(x_T) \tag{7}$$

$$y_p = h(x_T) \tag{8}$$

where $x_T = [x_1\ x_2\ \ldots\ x_n\ F_1\ F_2\ \ldots\ F_N]$ define the states of the system, y_s represents secondary outputs with negligible delay, and y_p represents primary outputs with a measurement delay. The primary outputs are chosen to PSD measurements (as provided by a CHDF for example) and therefore also correspond to the controlled outputs. Meanwhile, the secondary outputs correspond to the more frequent measurements, one of which must be the density, that are not controlled but used for the estimation of the primary output and states.

3.3 Methodology

The theoretical foundation required for fulfilling the desired research objectives are outlined in this section. The formulation of an efficient DO problem is central to the two proposed control structures. Accordingly, this section is primarily concerned with the theoretical basis required for such a formulation.

3.3.1 Model Order Reduction

The strong non-linearities prevalent in BPPs motivate the use of the general non-linear model (Equations 6–8) in the DO problem formulation. The main impedance to using the full non-linear model is its high-order and possible ill-conditioning, which make computation times intractable for real-time applications. Thus, non-linear MOR is a necessity for the formulation of an efficiently solvable DO.

To this end, the Galerkin projection (GP) is proposed as the MOR technique. As in other MOR techniques, the GP is based upon the idea that the dynamics of any high order, non-linear system can be represented by the dynamics in a subspace of that system. According to the results in Hahn and Edgar [2002], the dynamics of a reduced order version of the non-linear system in Equation 6 can be written as:

$$\dot{\bar{x}}(t) = P_x f_T(P_x^T \bar{x}(t)) \tag{9}$$

where $\bar{x}$ denotes the reduced state vector and P_x represents the transformation matrix that projects the dynamics of the high order system onto the reduced space. For this proposal, PCA is nominated as the way to find this projection matrix (Hahn and Edgar [2002]). Note that the population density states are only typically reduced as opposed to both population density and continuous phase states.

As mentioned in Section 2.3, training data is necessary for MOR using PCA. This can be generated as follows. First, a nominal input trajectory that steers the BPP system to the desired end-point PSD by a given final time must be identified through a DO problem. The training data can then be generated by simply adding random normal variation around the nominal input trajectories and repeatedly simulating the full order non-linear model of the system for the given final time with each simulation representing one batch. The data generated from this procedure can be arranged and unfolded according to Figure 3.

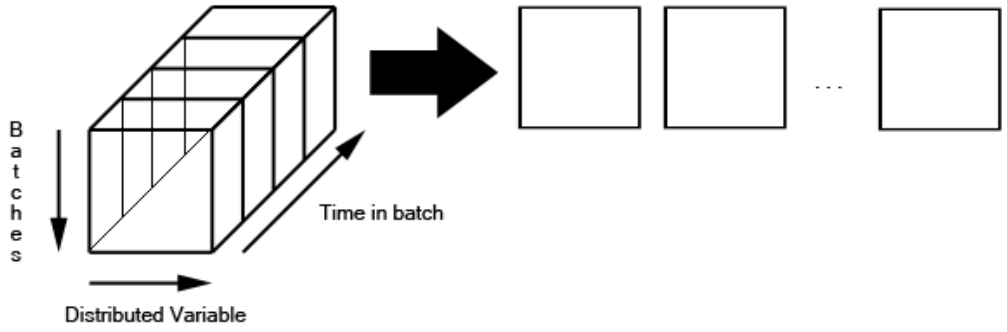

Figure 3: Batch data arrangement and unfolding scheme

The distributed variable in Figure 3 refers to the population density states, $F_1, F_2, \ldots, F_N$; thus, each individual block to the right of the arrow in Figure 3 represents the distributed variable for all batches at one specific time in the batch. PCA can then be applied to each of these data blocks using any standard algorithm available in literature (Eriksson et al. [2006]), thereby yielding a transformation matrix at discrete points during a batch. The reduced states at a specific point in time can then be expressed as:

$$\bar{x}(t) = P_x(t)x_T(t) \tag{10}$$

The controlled/primary output, the PSD, is also of a high order. In fact, the CHDF outputs a continuous PSD at each sampling instance of the primary output. For practical considerations, the primary measurements can be taken as the PSD measurement at N discrete particle sizes corresponding to N bins. As N can be a high number, transformation matrices to reduce the number of outputs, $P_y(t)$, are also required. These matrices can be readily determined from the simulation results by considering the PSD as the distributed variable in Figure 3. Thus, the reduced controlled outputs can be given by:

$$\bar{y}_p(t) = P_y(t)y_p(t) \tag{11}$$

The number of principal components (PCs) (i.e. the number of reduced states or outputs) to use in a given PCA model is an important design variable. This is further complicated by the fact that different PCA models are developed at different times during the batch and the optimum number of PCs required to capture the significant portion of the dynamics varies with time. The optimum number of PCs at a given time can be found through a cross

validation method (Eriksson et al. [2006]). The number of PCs (reduced states/outputs) chosen at each time should be verified by reconstructing the states and primary outputs using only the reduced states and transformation matrices and then comparing the results with the actual values. Similar to the case in Park and Doyle III [2004], it is suggested to first note the highest number of PCs required at any point during a batch and to keep this number of PCs throughout the batch for all the PCA models[11]. This is done to maintain consistency in the controller design at the expense of including additional states in the reduced order model at certain times.

3.3.2 State Observer

A multi-rate observer is proposed as one way handle the asynchronous measurements considered for this proposal. As discussed previously, the sampling interval and measurement delay associated with the primary measurements can be long compared to the inherent time scales of the governing phenomena of system. This necessitates the use of a multi-rate observer to estimate the (reduced) states and primary outputs of the system in between primary measurements. The observability of the (reduced) system states from density measurements alone were addressed in Section 2.4.1.

The mathematical details of the design procedure are available in Dokucu et al. [2008a] and Lee et al. [1992] and not reproduced here for brevity. A brief qualitative description of the observer is provided instead. For clarification, the two EKFs cascaded in the multi-rate observer will be denoted here as low-level and high-level.

A single rate system (single rate system I) is first constructed for which the states include: (1) reduced system states, (2) reduced primary outputs (i.e. the PSD), (3) delayed primary outputs, and (4) secondary outputs. The non-linear model of the system, Equations 6 – 8, are used to construct this single rate system. The output of single rate system I is simply the secondary outputs. Using estimates from the previous time step, single rate system I is integrated forward one sampling instance (of the secondary measurements) to bring it to the current time step. Then, secondary measurements from the plant are compared with those from the integration, recalling the outputs of the single-rate system are the secondary outputs. Finally, the augmented state vector is corrected based on the deviation using an appropriate EKF gain. The Kalman gain at this time step is for the reduced linearized version of the non-linear single rate system.

At sampling instances when the primary measurements become available, the augmented state vector as estimated from the low-level EKF, namely the elements corresponding to the primary outputs, are updated using a high-level EKF. In the design of the high-level EKF, another single rate system (single rate system II) is considered in which the states are the primary outputs and delayed primary outputs as predicted by single rate system I while the output consists of the delayed primary output one sampling interval ago (as estimated by single rate system I). Single rate system II has a sampling interval equal to

[11]The state and primary output PCA models are analyzed separately in this context.

the primary measurement sampling interval. Based on the difference between the primary measurement (which is delayed) and the output of single rate system II[12], the primary outputs as estimated from the *low-level EKF* are corrected. In brief, the objective of the second EKF is to correct the primary outputs as estimated from single rate system I based on the delayed primary output measurements; therefore, another single rate system (single rate system II) is constructed accordingly. The EKF gain used in this high-level EKF is based on the state space representation of single-rate system II. Based on this description, the ultimate result of this multi-rate EKF scheme is an estimation of the reduced system states and primary outputs at the current sampling instance based on plant measurements. However, note that the reduced states are not corrected using primary measurements in the high-level EKF. Instead, the primary measurements are only used to correct the estimated primary outputs from the single rate system I and not the states.

One can readily construct an estimation scheme to update states based solely on primary measurements (which are delayed) using the procedure outlined by Gattu and Zafiriou [1999]. Denoting the primary sampling time unit to be k, the goal of this scheme would be to use the primary measurement obtained at sampling instance k, which corresponds to the state of the system at $k-1$ (due to the delay), to correct the previously estimated states at $k-1$ using an EKF. The linearized model of the plant around the nominal trajectory at $k-1$ can be used to compute the Kalman gain. Note that states at $k-1$ are initially predicted using the information available at $k-2$. Following the correction at $k-1$, the non-linear system of the model can be integrated one sampling instance to time k, yielding an estimate of the states at k.

3.3.3 NMPC

The control structures proposed in Section 3.1 both involve solving a DO problem in which the primary objective is the minimization of the deviation between a predicted end-point PSD and desired end-point PSD. This requires the iterative integration of a non-linear predictive model of the BPP to the final time, which is computationally expensive. The computational requirements of solving the DO problem with a reduced order non-linear model has not been examined in the open literature. In Section 3.3.1, a non-linear MOR technique involving PCA is addressed. The issues with using a reduced order non-linear model in a DO framework are discussed in this section. Next, it is demonstrated that the choice of the solution strategy used for solving the DO can be used to overcome these issues and possibly make computation times tractable for real-time applications.

The numerical solution strategies for DO problems can be broadly categorized as being either simultaneous or sequential. The sequential solution strategy is a partial discretization approach in which the control trajectories are discretized over the time domain, commonly as piecewise constants. Then, given an initial guess for the discretized input trajectory and initial conditions, the dynamic model is integrated to the final time at each iteration, the objective function and any terminal constraints are evaluated, and an optimization routine

[12]Note that these both correspond to the state of the system one sampling interval ago.

adjusts the input trajectory accordingly. Because the dynamic model must be integrated to the final time at each iteration, the computational times associated with a sequential approach is heavily influenced by the properties of the ODE integrator. In principle, one could use the reduced order non-linear model in the DO problem and solve for the input trajectory with the sequential approach. However, the iterative integration of a reduced order non-linear model can become extremely computationally expensive because the transformation matrix at every numerical integration step must be both stored and evaluated. This is especially computationally prohibitive with highly accurate ODE solvers. Nevertheless, it would be constructive to evaluate the performance of the sequential solution strategy with an ODE solver using fixed integration steps. With a fixed step integration routine, the number of transformation matrices which need to be stored and evaluated can be reduced.

With the simultaneous solution strategy, the computational effort required for iterative evaluation of the reduced order non-linear model (Equation 9) can be significantly reduced. In the simultaneous approach, the solution of the model and optimization problem are determined simultaneously. To this end, the state and inputs trajectories are first both discretized along the time domain. The inputs are again typically discretized as piecewise constants whereas the states are discretized using one of many available techniques. The discretization of the states reduces the model ODEs into a system of algebraic equations. The algebraic system is then included as equality constraints in a single non-linear program (NLP). The popular state discretization method among the process systems community has been collocation, specifically orthogonal collocation on finite elements (OCFE). The principle behind collocation is explained next in very simplistic terms to illustrate how a collocation based simultaneous solution approach can reduce computational requirements. Assuming only one finite element, each reduced state in Equation 9 is approximated with a polynomial in the time domain with unknown coefficients. For instance, with r states in the reduced order non-linear model and quadratic approximations, one obtains the approximations, $\tilde{\tilde{x}}$:

$$\begin{aligned} \tilde{\tilde{x}}_1 &= a_0 + a_1 t + a_2 t^2 \\ &\vdots \\ \tilde{\tilde{x}}_r &= r_0 + r_1 t + r_2 t^2 \end{aligned} \tag{12}$$

The approximating functions can be substituted into their corresponding ODEs (Equation 9) and evaluated at a number of collocation points, yielding residuals for each ODE. These residuals can then be set to 0 and together with initial conditions, the unknown coefficients of the approximating polynomials can be calculated. For the approximations in Equation 12, two collocation points, (t_1 and t_2), and initial conditions for each reduced state are required to be specified. The residual system for the approximating functions in Equation 12 at the two collocation points are:

$$\begin{pmatrix} a_1 + 2a_2 t_1 \\ b_1 + 2b_2 t_1 \\ \vdots \\ r_1 + 2r_2 t_1 \end{pmatrix} - P_x f_T \left(P_x^T \begin{pmatrix} a_0 + a_1(t_1) + a_2(t_1)^2 \\ b_0 + b_1(t_1) + b_2(t_1)^2 \\ \vdots \\ r_0 + r_1(t_1) + r_2(t_1)^2 \end{pmatrix} \right) = 0 \tag{13}$$

$$\begin{pmatrix} a_1 + 2a_2t_2 \\ b_1 + 2b_2t_2 \\ \vdots \\ r_1 + 2r_2t_2 \end{pmatrix} - P_x f_T \left(P_x^T \begin{pmatrix} a_0 + a_1(t_2) + a_2(t_2)^2 \\ b_0 + b_1(t_2) + b_2(t_2)^2 \\ \vdots \\ r_0 + r_1(t_2) + r_2(t_2)^2 \end{pmatrix} \right) = 0 \tag{14}$$

The residual systems can be set as equality constraints in a NLP with the coefficients being decision variables (in addition to the input profiles). While this may lead to a large NLP, efficient optimization routines exist for solving such problems. Based on these considerations, when using the simultaneous approach to solve the DO problem, the computational requirements are significantly reduced compared to the sequential approach. More specifically, excessive storage and evaluation of the transformation matrices are not necessary for the integration of the reduced order non-linear model. Instead, these matrices are only required at collocation points within each finite element, which can be determined beforehand.

Having established the solution strategies, a general formulation of the optimization problem is presented next. Given a desired end-point PSD, it is initially recommended to project the desired end-point PSD onto the reduced output space using $P_y(t_f)$ to reduce the size of the control problem. Denoting this projection as $\bar{y}_p^{des}(t_f)$ and assuming the reduced output space to be comprised of M principal components, the general optimization problem can be given by:

$$u(t) := arg\min \sum_{i=1}^{M} (\bar{y}_{p,i}(t_f) - \bar{y}_{p,i}^{des}(t_f))^2 \tag{15}$$

subject to:

$$\dot{\bar{x}}(t) = P_x f_T(P_x^T \bar{x}(t), u) \tag{16}$$

$$\bar{y}_p(t) = P_y h(P_x^T \bar{x}(t)) \tag{17}$$

$$q(x(t_f), y_p(t_f), u) = 0 \tag{18}$$

$$w(x(t), y_p(t), u) \leq 0 \tag{19}$$

$$u_{min} \leq u \leq u_{max} \tag{20}$$

Equation 20 represents the input constraints while the vector functions, $q(\cdot)$ and $w(\cdot)$, in Equations 18 and 19 (respectively) denote general terminal and path constraints. It is worth noting that the path and terminal constraints will typically require evaluation of the transformation matrices as these constraints will be defined in terms of the variables in the full order model space (as opposed to the reduced space).

3.3.4 Set Point Modification

The second control strategy proposed in this report involves periodically modifying PSD SP profiles sent to local tracking controllers. In this strategy, the SP profiles can be updated whenever primary measurements (with delay) become available. The updated SP profiles, which incorporate plant conditions to some degree, increases tracking feasibility. The new SP profiles are determined by solving the DO problem posed in the previous section after

initialization at the current estimated states of the plant.

The set-point updates should occur as soon as possible once primary measurements are available to maximize the duration of the feedback. As illustrated by Gattu and Zafiriou [1999], tracking controller performance can be significantly improved by making set point changes one step in the right direction instead of finding completely new set-point profiles. Thus, the full solution to the DO problem at every sampling instance is not necessary. Instead, if it is observed the objective function in the DO decreases sufficiently (as defined by a user-defined threshold) after a certain iteration while satisfying all constraints, the DO can be terminated and the SPs sent to the tracking controllers can be taken as PSD profiles at the current iteration. The control strategy for tracking the PSD is recommended to be QDMC based on its successful application for PSD tracking in Dokucu et al. [2008b] and Dokucu et al. [2008a].

3.4 Research Plan

With the methodology of the control structures established, simulation case studies should be performed to demonstrate the efficacy of the two structures and to assess if they can be implemented in real-time applications (the main hypothesis). Generally, any BPP that can be described by PBEs (for the PSD) and ODEs (for the continuous phase state variables) and for which the main control objective is to reach a desired PSD by batch termination can serve as a good candidate for the case study[13].

To this end, an emulsion polymerization process is suggested as the case study. Detailed population balance based models for various particulate polymerization processes are widely available in the literature. One can consult the review article by Kiparissides [2006] for a detailed listing. The primary control objectives in emulsion polymerization processes are to achieve a desirable end-point PSD and to reach a total final solids concentration by batch termination for the reasons given in Section 1.2. With respect to the sensors, a CHDF and densitometer can be assumed to exist for simulation purposes (See Section 2.4.1). For any case study involving an emulsion polymerization process, it is recommended to first consider a coagulation-free population balance model so as to simplify the numerical methods required to solve the PBE[14], and then increase the level of complexity by including the coagulation phenomena in the PBE if results from using coagulation free model are encouraging.

The tasks required to be completed for this proposal is provided next in a chronological order with expected times of completion for each task. Tasks denoting major milestones are also identified with stars.

1. **Task:** A literature review of PBEs and existing solution methods with focus on bin based methods. The information in Ramkrishna [2000] (Chapters 1 - 4) is a comprehensive source for population balances and the existing solution methods. The two-tier

[13]Processes for which control based on moments of a PSD is appropriate are not considered.

[14]This assumption is often made in literature for simplification purposes or under certain operating conditions (high surfactant concentration or vigorous mixing inside the reactor).

solution approach developed by Immanuel and Doyle III [2003a] specifically needs to be reviewed as this method will be used later to discretize the population balance model. (2-3 months)

2. **Task:** A literature review of emulsion polymerization and the existing models followed by selecting a specific emulsion polymerization system. Emphasis should be placed on understanding the expressions for the different terms in the population balance. The necessary model parameters, initial conditions, batch time, and constraints (input and operational) should also be established. (2-3 months)

3. **Task ★:** Development of the code for the numerical solution (the two-tier bin based method in Immanuel and Doyle III [2003a]) of the chosen emulsion polymerization process. The model should then be simulated and the resulting PSD profiles should be compared with existing results. The full order model will later serve to act as the 'plant' in closed-loop simulations. (4-5 months)

4. **Task:** Reachability analysis to determine reachable PSDs. The full order model in the previous step must be used as the predictive model in the DO problem. From the reachable PSDs, a target end-point PSD should be chosen. For this target PSD, the input, state (i.e. the population density states), and output (PSD) profiles must be saved. The influence of the control vector parametrization on the reachable region should be assessed to determine an appropriate discretization. (1-2 months)

5. **Task:** Generation of the training data set, development of PCA models for the population density states and outputs, and formulation of the reduced order linear time varying model of the process. (2-3 months)

6. **Task:** Development of the code for the two observer systems described in Section 3.3.2, computation of the necessary Kalman gains from the reduced order linearized models, and assessment of convergence properties of the observers. The latter can be done through simulations. (0-1 month)

7. **Task ★:** Development of the code for the QDMC algorithm for PSD trajectory tracking coupled with the multi-rate observer and evaluation of tracking control by first assuming a perfect model and then considering disturbances and parametric uncertainty. Any tuning should be done as necessary to achieve acceptable control performance. (2-3 months)

8. **Task ★:** Formulation of the DO problem required to be solved for the two proposed control strategies, development of the code to solve the DO problem with a sequential and simultaneous approach while incorporating MOR, and assessment of solution times and accuracy. The open loop DO problem is considered for this task. The number of finite elements in the code for the simultaneous strategy will be dictated the control vector parametrization. The accuracy of solution to the DO should be compared against the solution when using a full order model. (6-7 months)

9. **Task:** Comparison of QDMC and NMPC performance and robustness to disturbances and parametric uncertainty assuming full state feedback and then output feedback

(using the multi-rate scheme for state estimation). The main performance criteria will be the deviation of the final PSD from the target. This task will answer if replacing tracking controllers with NMPC is beneficial. (4-5 months)

10. **Task ★:** Development of the code for the SP modifier control scheme first with full state feedback and then with only output feedback and evaluation of the performance benefits (if any) of the SP modification scheme in the presence of model errors and disturbances. The results to compare against were already generated in evaluating the performance of QDMC. (2-3 months)

11. **Task:** Comparison of the robustness and performance of the NMPC scheme with the SP modifier scheme. (1-2 months)

The total duration of the project is estimated to be $26 - 37$ months.

3.5 Anticipated Significance

The main contribution of this work is the application of effective non-linear predictive control strategies to the problem of full end-point PSD control in BPPs. The proposed non-linear predictive structures have yet to be considered in the open literature mainly due to the computational burdens associated with using a high order non-linear model in a DO problem. This proposal includes possible ways to incorporate a reduced order non-linear model within a DO framework such that solution times can be tractable for real-time applications (owing to both the model reduction and formulation of the DO). By employing a non-linear process model in the control structures, tighter and robust control of the PSD is anticipated.

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