# Dynamic Optimization and Non-linear Model Predictive Control to Achieve Targeted Particle Morphologies

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An event-driven approach based on dynamic optimization and nonlinear model predictive control (NMPC) is investigated together with inline Raman spectroscopy for process monitoring and control. The benefits and challenges in polymerization and morphology monitoring are presented, and an overview of the used mechanistic models and the details of the dynamic optimization and NMPC approach to achieve the relevant process objectives are provided. Finally, the implementation of the approach is discussed, and results from experiments in lab and pilot-plant reactors are presented.

**Keywords:** Dynamic optimization, Emulsion polymerization, Nonlinear model predictive control, Particle morphology, Pilot-plant reactor test, Process monitoring

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## 1 Introduction

Polymers are ubiquitous in structural materials and consumer products applications not only due to the ready availability of feedstocks, but also because of their exceptional performance characteristics, such as mechanical properties or life span. The availability of polymeric materials with unique performance characteristics result in many new types of products, based on new types of useful functions.

With the acceleration of technological progress, the shelf-life of innovations is dropping. At the same time, there is a growing demand for tailored products. Due to the diverse applicability of polymers, the dimensionality of the optimization space is very high for new materials and processes. Thus, an approach to develop next generation products and processes purely driven by trial and error can hardly deal with the increasingly shorter innovation cycles [1]. In addition to the pressure to innovate faster, increasing competition demands operational excellence and a shift from conventional operation practices. One alternative to cope with these challenges is the use of model-based methods for optimization and control of processes. As the product properties strongly depend on the operating conditions, the use of

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mechanistic models in optimization and control is particularly advantageous as the underlying dynamics and interactions can be captured by the model.

In this research article, an emulsion polymerization is considered being one of the most complex polymerization processes. It is a multiphase process in which the polymerization starts in the aqueous phase and continues in the latex particles [2]. Important operational requirements are the stability of the emulsion [3] and removal of the heat from the exothermic polymerization reaction [4]. Since emulsion polymerizations are typically conducted in semi-batch mode, the process is inherently transient [5].

Standard operation in semi-batch emulsion polymerization is based on fixed and pre-defined operating conditions. This does not allow reaction to process changes due to disturbances or operation requirements. An alternative event-driven approach for improved process monitoring and control is investigated in this work. It combines novel sensor techniques, process models and (real-time) optimization strategies. These techniques further contribute to improve the efficiency of assets by lowering energy and raw material consumption and increasing the productivity while maintaining or improving product quality. Also, a lower batch-to-batch variation of the product quality can be achieved.

Three ingredients are necessary for an efficient implementation of an event-driven approach:

1) Sensors: Hard sensors provide fast and reliable data about the instantaneous process state. Techniques like online calorimetry [6] and Raman spectroscopy [7] are used to gain information about the process state.
2) Models: Availability of dynamic process models incorporating kinetics, particle morphology [8] and reactor periphery, is an essential pre-requisite to accurately quantify the process dynamics to optimize the process in real time.
3) Efficient strategies for dynamic optimization and advanced process control: Model-based control and optimization strategies and tools are needed to estimate the current process states using measurements (temperature, concentrations, flow rates) which are then used to predict, monitor and control the process ensuring the objectives (e.g., product quality and amount) are achieved, while maintaining process constraints (e.g., safety, pumps, and cooling capacity.

In this area the state of the art in emulsion polymerization is limited to the control of copolymer composition, particle size distribution, and molecular weight distributions of linear polymers mainly using calorimetric sensors [9 – 11], however, control and optimization of the particle morphology has not been addressed.

This research article illustrates recent achievements for the three ingredients for advanced process control. In Sect. 2, the use of Raman spectroscopy to monitor monomer concentrations and transmission electron microscopy (TEM) for direct measurement of particle morphology are discussed. Sect. 3 contains an overview of the models used for the dynamic optimization and nonlinear model predictive control (NMPC). In Sect. 4 the dynamic optimization and the NMPC scheme is presented. Finally, in Sect. 5, the results of the implementation of the dynamic optimization and NMPC to produce a polymer with a desired particle morphology in both a lab and pilot plant reactor are presented.

## 2 Sensor Development

This section specifically focuses on the application of Raman spectroscopy for monitoring monomer concentrations and overall conversion and on the feasibility of using in situ liquid transmission electron microscopy for monitoring the morphology of polymer particles.

### 2.1 Raman Sensor

Process Raman spectroscopy is by now a mature process analytical technology (PAT) with multiple industrial applications, in particular in quality control and real-time process monitoring from petrochemical [12] to food industry [13], to manufacture of bio-pharmaceuticals [14]. To date, there is also a significant body of literature on the application of Raman spectroscopy for monitoring monomer composition in different polymerization systems. A recent review provides several examples of industrial applications of optical spectroscopy methods, including Raman scattering, in the polymerization processes [15]. State of the art of monitoring monomer concentrations in emulsion polymerization was tested: good individual monomer concentration models were developed for up to two monomers, as both monomers were good scatterers and sufficiently accurate chemometric models could be developed. However, in a four-monomer system the monitoring of the individual monomers was too complex to deconvolute the signal due to significant spectral overlap and the presence of weak scatterers in the specific chemical system of interest; only the overall conversion could be monitored with sufficient confidence using a Raman fibre optic sensor real time data [16].

Application of a Raman in situ sensor in a specific polymerization system is, therefore, not a straightforward task and requires careful development of a calibration model for the specific polymer system. In the recent RECOBA H2020 project, identical Raman immersion probes were installed in the lab and the pilot reactors to obtain real time monomer concentrations in the demonstration polymerization case. An indirect hard modeling (IHM) [17] model was developed to monitor the concentrations in the second stage of the polymerization inline. IHM is a multivariate regression method that uses pure component spectra, that allows for nonlinear changes such as peak shifts. The five main components of the second stage polymerization were modeled with spectral models, i.e., first-stage polymer, sec-

  

ond stage polymer, water and two main monomers. These components were modeled using up to 20 peak-shaped functions. Other components were neglected due to their low concentrations and related difficulties to monitor them in an online setting. The weight fractions of the main monomers using the IHM model are predicted very well (see Sect. 5), using high-performance liquid chromatograhpy (HPLC) grab samples as reference measurement. The prediction accuracy is sufficiently high for online monitoring and control.

### 2.2 Liquid In Situ TEM for Polymer Morphology Studies

The only direct way to determine the morphology of polymer nanoparticles is to look at them. This has become feasible with the advances made recently in sampling techniques and the technology of electron microscopy. The earliest work on in situ sampling for electron microscopy is attributed to Marton who published a series of papers in 1934 – 35 on the topic; see review of the early studies on sampling techniques in [18]. The resurgence of interest in this technique in recent years is due to significant advances in nano-fabrication, which allowed to produce much thinner windows and much more reliable cells, as well as to the capabilities of the modern aberration corrected microscopes. There is an increasing number of studies reporting the use of in situ liquid transmission electron microscopy (TEM) for understanding of the mechanisms of nanoparticle formation, for imaging of cells, for understanding of catalytic and electrochemical reactions, and so on [19].

At present it is not yet feasible to use in situ liquid TEM as a process sensor, but it was of interest to the project whether images of sufficient contrast could be obtained to justify further work in this direction. A series of polymerization reactions were performed with samples collected at specific times and stored for off-line analysis. Analysis was done as soon as practically possible at the end of the experiment to avoid issues with sample stability.

For TEM sampling a Protochips Poseidon Select liquid cell was used, with imaging done using FEI Tecnai G2 TEM instrument, with a 25 fps resolution camera at an acceleration voltage of 200 kV in bright field mode. Time-course aliquots of polymerization reaction mixture as in the DLS measurements were imaged. A dilution of the sample to 0.1 % solids content with de-ionised water was sonicated for 0.5 min before imaging. A droplet of latex solution in deionized water was placed onto a bottom chip. This chip was then secured inside the sample holder and was followed by the attachment of the top chip. After the two chips were aligned within the holder, the assembly was sealed. In some cases the bottom chip was coated with a 0.001 – 0.01 % aqueous solution of poly-L-lysine (Mw 70k – 150k, Sigma-Aldrich) to improve adhesion of the latex to the silicon nitride surface. A small drop of poly-L-lysine was allowed to coat the silicon nitride surface for 15 min before removing excess liquid and depositing an aliquot of latex particle suspension.

A typical result is shown in Fig. 1. It is noted that the particle size observed in TEM is usually lower than that recorded by DLS. It should be noted that the two measurement techniques are recording somewhat different things: a mean diameter of a population of particles corresponding to a sphere of rotation, in the case of DLS, and size of a specific particle or of a small number of particles, which can be measured differently if particles are non-spherical. It is believed that the size observed in the liquid TEM is closer to the actual size of the particles.

The images collected in static in situ cell were obtained without any staining and exhibit a relatively low contrast, however, some features of particles' morphology can be identified visually, see Fig. 1. To develop this imaging technique into an online application, there will be a requirement to automate image analysis. This will require images with more contrast than what has been achieved till now.

As instrumentation and sampling technique of liquid in situ TEM continue to develop, there will be a possibility to exploit this technique in applications requiring close to real-time information on particle size and morphology, especially if information on the dynamic evolution of morphology and on interactions of particles under experimental conditions will be required.

## 3 Model Development

In this section, the different models used in this work – namely a model for the kinetics associated with monomer consumption and one for the morphology development – are briefly presented.

### 3.1 Kinetic Model

Here, two models describing polymerization kinetics and morphology evolution are presented. Last part of this section captures analysis of the models and its usability for online control.

Zubov et al. [20] presented on-line control and optimization of semi-batch emulsion copolymerization at a pilot plant. The optimization goal was controlling the batch temperature and feed rates to minimize batch time while preserving product quality or obtaining the desired shift in product quality. As a measure of the product quality, authors chose number-average molecular weight. Here, two extensions for the product quality tracking are proposed. Both extensions use kinetic model based on ordinary differential equations (ODEs) presented in [20].

First extension tracks morphology of polymer particles. The model equations, morphology optimization and control are described in Sect. 3.2. The morphology equations are implemented separately to the kinetics, so the particle

  

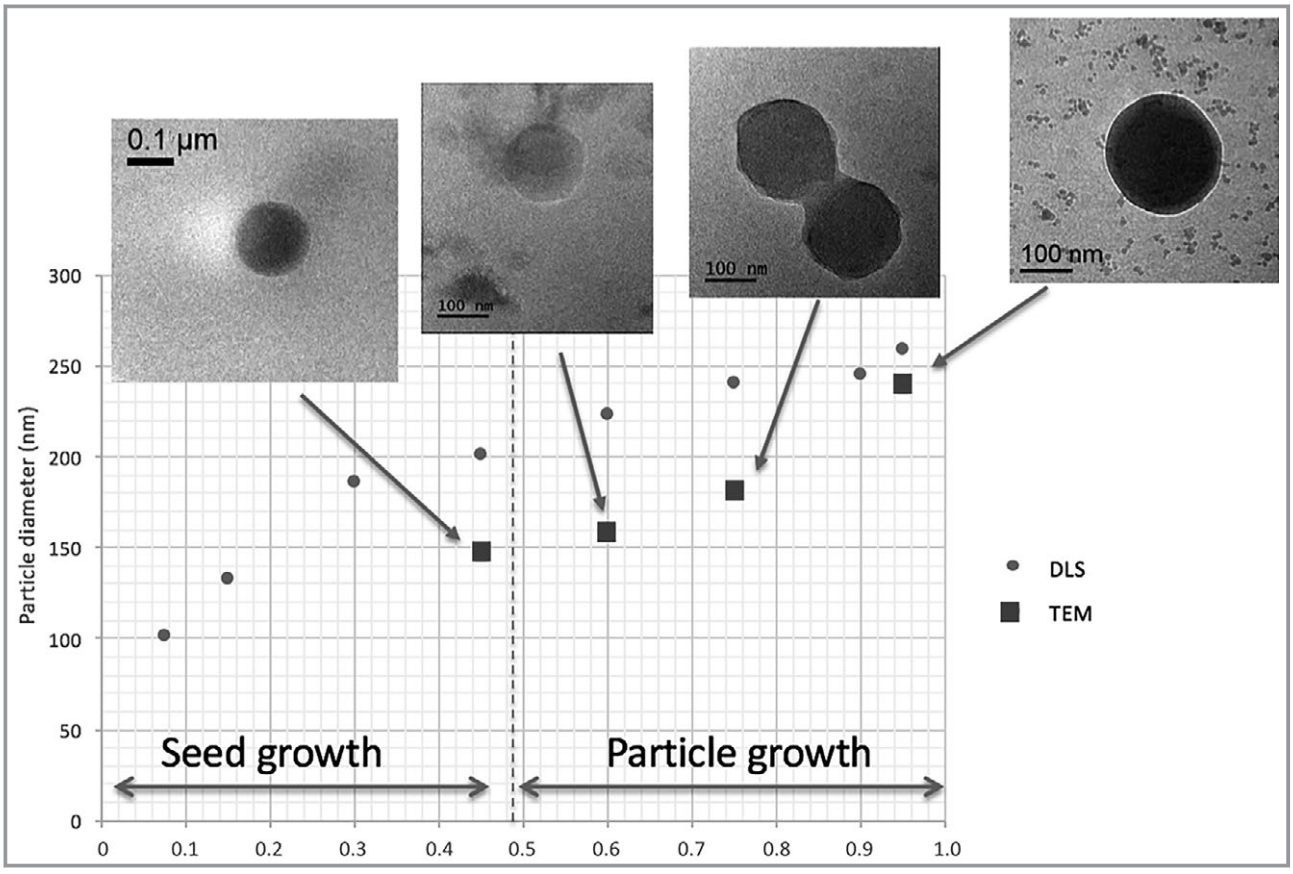


**Figure 1.** Exemplary progress in polymerization monitored by off-line dynamic light scattering and static liquid TEM.

morphology tracking is an optional extension to the polymerization control.

Second extension computes weight-average molecular weight (Mw), because it is closely connected to the viscosity of the polymer, which then influence the morphology generation (migration of the clusters mentioned in next section). To compute Mw, a fast Hybrid Monte Carlo (HMC) was implemented, which combines ODEs and stochastic construction of polymer chains in branched macromolecules and, thus, meets requirements on computational speed for on-line control [21]. Moreover, HMC provides good agreement with experimental data effectively (in sense of computational time), due to the separation of polymer particles [21]. However, fast HMC is burdened with random statistical error (due to Monte-Carlo generation of polymer chains), so it is not directly suitable for model-predictive control. Therefore, a surrogate learning function was implemented, which is based on a Kriging's model [22] predicting Mw based on HMC predictions at different process conditions. The controllable process conditions – a sampling plan – can be arbitrarily extended according to the current situation in the reactor and then the surrogate function can be adjusted (Fig. 2). The combination of HMC and surrogate function is an innovative approach and it was not yet tested in real conditions; it is here presented just as a concept.

### 3.2 Morphology Model

Since the final goal is the optimization and control of a semi-batch emulsion polymerization process to produce polymers with a desired particle morphology, a fast mathematical model was developed for 2-phase polymer-polymer latex particles. The model is explained in detail in a previous publication [23]. Here, only a brief summary will be presented.

The model was developed for seeded emulsion polymerization of two phase polymer – polymer system but can be easily modified and used for organic/inorganic systems [24].

In seeded semi-batch emulsion polymerization, particle morphology is a result of the incompatibility between two polymer phases (the polymer matrix phase and the polymer in the second stage of polymerization). The equilibrium morphology will be the morphology with minimum interfacial energy which can be core-shell, inverted core-shell and hemispherical for 2-phase polymer-polymer latex particles.

  

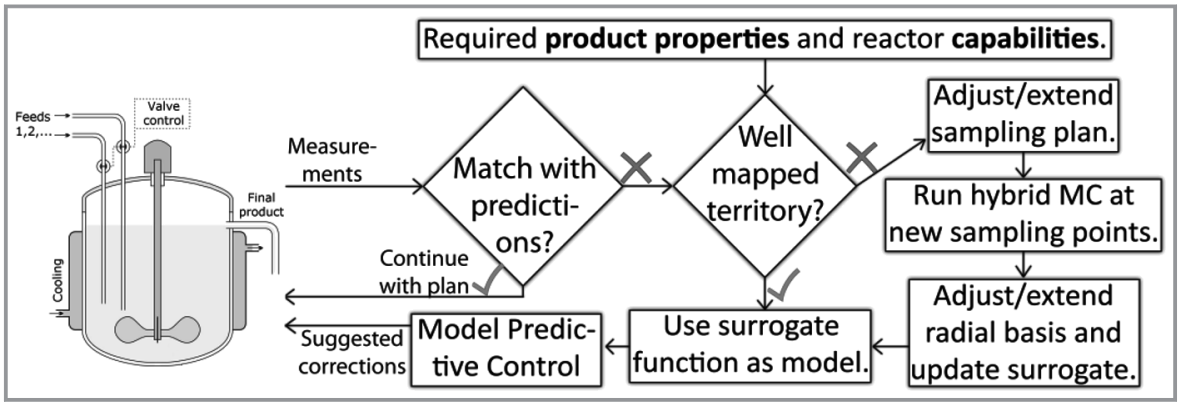


**Figure 2.** Scheme of the inclusion of the surrogate function and HMC model into control tools.

The equilibrium morphology can be predicted using the equilibrium morphological map first presented in [25]. Using this map one can predict the equilibrium morphology knowing interfacial tensions between different phases. Not always, a thermodynamically controlled morphology is reached as kinetics play an important role also on the formation of the morphology. The final morphology is the result of the balance between thermodynamics and kinetics.

In the current model, the knowledge on the thermodynamic equilibrium morphologies is used to model and predict dynamics and the evolution of the particle morphology considering all the kinetics (such as cluster nucleation and polymerization, polymer diffusion and cluster aggregation) involved in the process of the particle morphology formation. Based on the thermodynamic equilibrium morphology the particle is divided into two regions: the equilibrium position (the center of the particles for inverted core-shell morphology and the surface of the particles for hemispherical and core-shell morphologies) and non-equilibrium positions. As a result, the clusters of the second phase polymer are located either at the equilibrium positions or non-equilibrium positions. This leads to two different population of the clusters. The particle morphology can be defined as the size distributions of the clusters at these two different regions. In this approach, the computation time is relatively small (see below). In addition, the population of clusters obtained in the whole polymer particles, allows presenting the particle morphology as the cluster distribution, which gives a more realistic description of the system with almost $10^{17}$ particles $L^{-1}$ in contrast to the existing methodologies of modelling the morphology of a single particle [26, 27].

The model first predicts the equilibrium morphology of the polymer particles and based on that different population balances are computed. The population balances of the clusters in the equilibrium and non-equilibrium positions includes terms of growth of the clusters by polymerization, diffusion of polymer from matrix, aggregation of clusters and their migration to the equilibrium position. The population balances of $m(x)$ and $n(x)$ (the normalized number of the clusters in non-equilibrium and equilibrium positions with $x$ monomeric units in the polymer chains, respectively) were presented in detail in reference [23] and will not be repeated here.

The parameters of the model such as: rate coefficients of aggregation, movement to the equilibrium position, mass transfer and rate coefficient of nucleation are dynamic as the medium of the polymerization changes during the polymerization (See [23] for detailed information).

The population balances were discretized using the Kumar Ramkrishna method [28] and solved together with the kinetics of polymerization. With a given set of parameters the Matlab code runs in less than 20 seconds in Windows on a laptop with an Intel®, Core™ i7-4610M CPU @ 3 GHz.

Fig. 3 shows the model output which is presented as two distributions and how well the model predicts the effect of different parameters on the final distribution. The model was validated by fitting the evolution of particle morphology of composite particles during polymerization of methyl methacrylate on a polystyrene seed (see [23]).

Using the cluster size distribution, in a random process the representative images of the particle morphology of 1 000 000 particles, as sampling particles were defined. Fig. 4 shows the final distribution of the equilibrium and non-equilibrium clusters of case A (hemispherical) and the morphology of 10 particles randomly selected among all for a case with a set of parameters and for the case that the movement towards the equilibrium position enhanced by lowering the viscosity of the matrix (softening the system). The black circles represent the matrix and the white circles represent the second stage polymer clusters.

In addition to the prediction of the final morphology, the model predicts the evolution of the particle morphology during the reaction. Fig. 5 shows the particle morphology evolution during the reaction by showing the distribution of the clusters at different conversions and their corresponding representative particle morphologies in 2D, 3D and TEM-like images in which the contrast is a function of the fraction of different phases at each point.

  

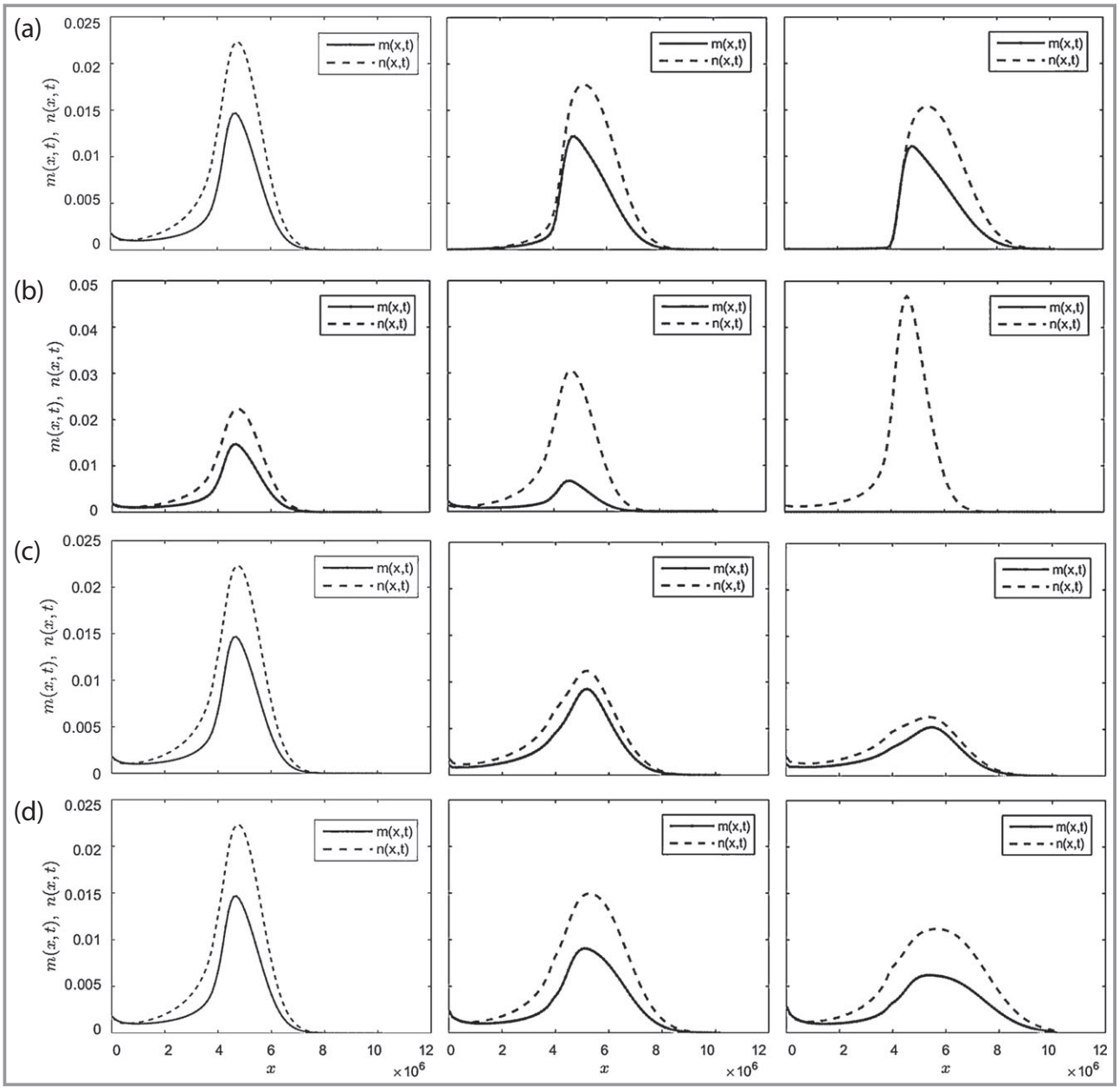


**Figure 3.** Final distribution of equilibrium and non-equilibrium clusters. (a) increasing the aggregation rate; (b) increasing the cluster migration rate; (c) decreasing the cluster nucleation rate; (d) increasing the diffusion rate; always from left to right.

## 4 Dynamic Optimization and Nonlinear Model Predictive Control

In this section, the methods and tools used for dynamic optimization and nonlinear model predictive control (NMPC) are summarized.

The batch time is minimized to produce polymer particles with a desired morphology using the combined model for both polymerization and particle morphology kinetics. Constraints related to safety, such as temperature or amount of unreacted monomer are included in the problem formulation. The recipe determined in the offline optimization step is subsequently used for tracking control in the NMPC.

The standard operating procedure is an isothermal semi-batch process with constant monomer feeding. The main feeding period is followed by a post-polymerization phase. Only the monomer feeding profile is optimized. The reactor temperature and initiator flow rate are unchanged as in the standard recipe. To reach the target morphology at the end of the feeding period an endpoint constraint is imposed. This constraint is the absolute least squares error between

  

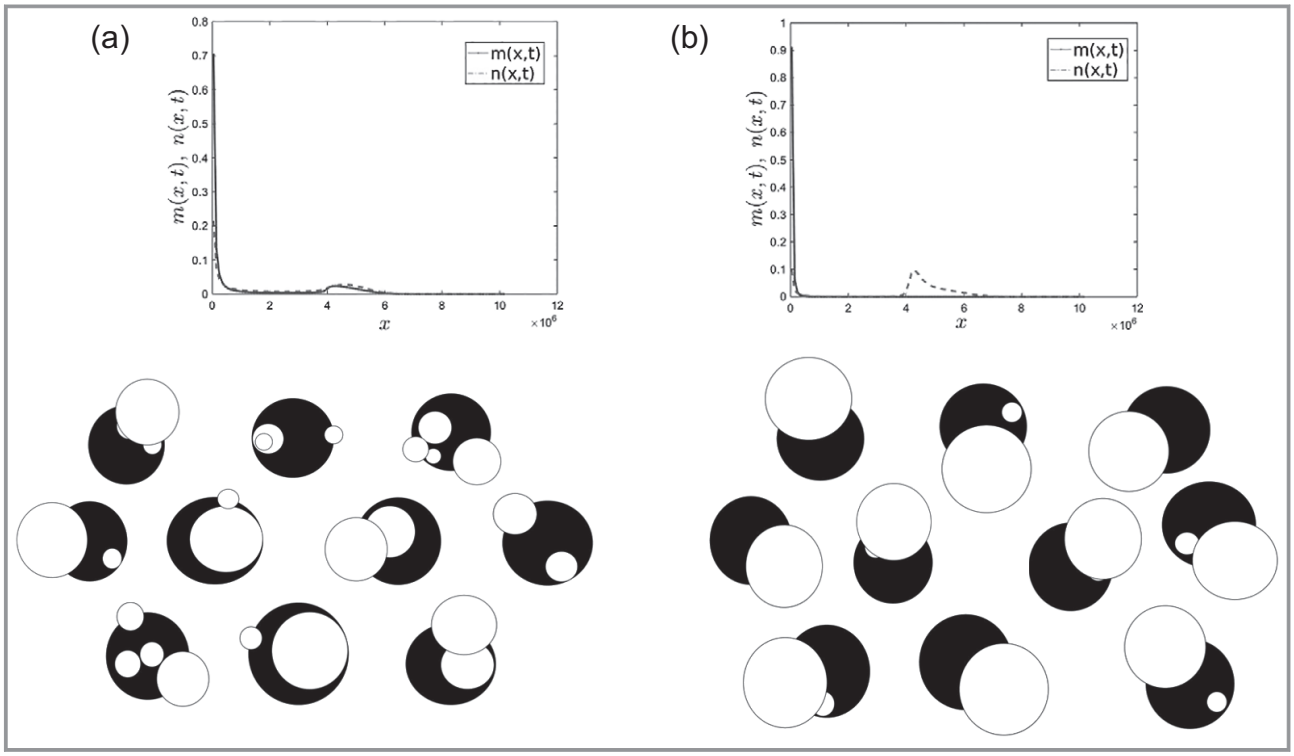


**Figure 4.** Final distribution of the equilibrium and non-equilibrium clusters and representative morphologies of 10 randomly selected particles among 1 000 000 sampling particles, (a) hemispherical equilibrium case A harder system, (b) hemispherical equilibrium case A, softer system.

the predicted and the desired cluster distributions. Additionally, the amount of monomer fed must match the recipe value and the conversion at the end must be at least as high as in the standard operation at the end of the feeding period.

Fig. 6 shows the results of the dynamic optimization. The batch time is decreased by 6 % while the desired morphology is achieved and all constraints are satisfied. It can be seen that the monomer is fed much faster in the beginning than in the standard recipe. Consequently, the monomer concentration is higher leading to a higher polymerization rate allowing for the decrease in batch time. The optimized state trajectories for conversion and temperature are used as reference for the controller.

An NMPC controller is developed for the emulsion polymerization system. The controller is set up to track the optimized concentration profiles resulting from the offline optimizer, and at the same time keep the reactor temperature at setpoint. The controller manipulates the feed rates and the jacket inlet temperature.

The NMPC controller consists of three parts: A specific model and application component, tailored to this particular emulsion polymerization system. It contains a kinetic model of the emulsion polymerization reactions, a detailed model of the cooling system and a model of the feed system. Secondly, it contains an estimator, in this case an extended Kalman filter [30], performing online state and parameter estimation using available measurements. The third component is the NMPC control algorithm, previously described in [20, 31]. Observability is an important model property which determines the reliability of the estimated states. The states in the model of the polymerization process can be subdivided into subsystem I comprising the states corresponding to the mass and energy and subsystem II consisting the states associated with molecular properties, e.g., the distribution of the molecular weights and the polymer morphology. The states in subsystem II are not observable with measurements of temperature and conversion only [32]. In the control scheme considered here, temperature and spectroscopic measurements are used to calculate the amount of free monomer. Both the measurements are, therefore, of variables in subsystem I. Hence, in the state estimation scheme implemented, only the states corresponding to the mass and energy balance are estimated using the available online measurements, the other states are only predicted.

An NMPC requires accurate, yet computationally efficient models. Such a model accounting for kinetics [33] and morphology [23] is derived and implemented in the C language in a template of the Cybernetica CENIT control software. Measures have been taken to make it numerically efficient, and the model has been trimmed in terms of state-space complexity for performance and robustness. As a result, the model consists of ordinary differential equations where all state equations and output equations are nonlin-

  

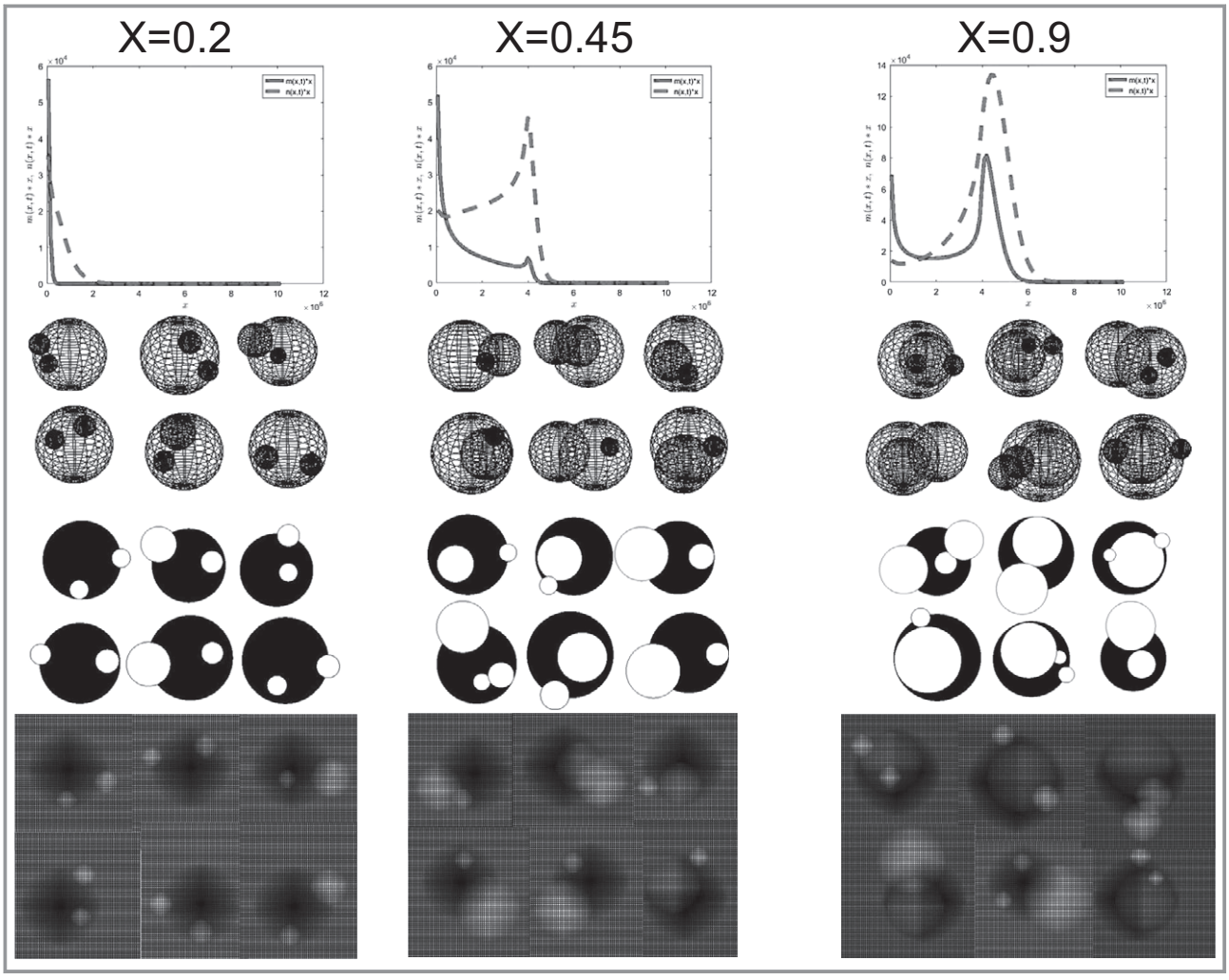


**Figure 5.** Conversion evolution of the weighted distribution for equilibrium and non-equilibrium clusters and relevant 3D, 2D and TEM-like morphology images related to each distribution for 6 randomly selected particles among all (reproduced from [29]).

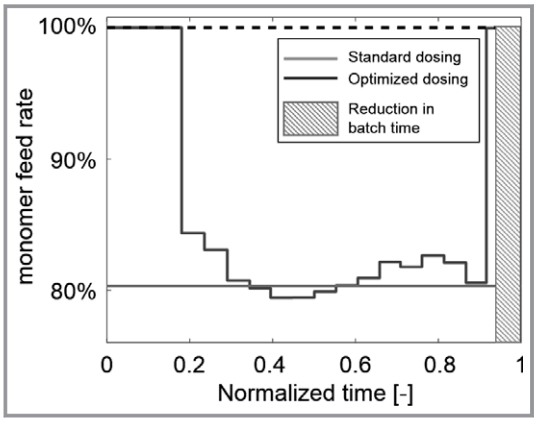


**Figure 6.** Results of dynamic offline optimization of the semi-batch polymerization process.

ear but explicit, eliminating the need for a differential-algebraic solver. The kinetic model is updated online by estimating propagation factor of monomer from inline Raman measurement, while the jacket cooling model is updated from measured reactor and cooling water temperatures.

## 5 Implementation and Demonstration

Some of the technologies developed in the project, were selected for implementation and for online demonstrations. The demonstrations were carried out in a series of experiments, first on lab-scale and then in a pilot plant.

The purpose of these demonstrations were twofold:

1) To demonstrate that a certain particle morphology can be produced based on pre-optimized semi-batch polymerization profiles.

  

2) To demonstrate how a NMPC system can implement pre-optimized trajectories, suppress disturbances and re-optimize when conditions change.

The lab reactor used in the demonstration, is a metallic 3-L-reactor. It is equipped with the essential sensors to enclose the energy balance around the reactor. These involve flow meters for the coolant fluid and for the reactor feeds via four weighing scales and pumps. The reactor is also equipped with temperature sensors for all the materials entering and leaving the system. The reactor is sealed in a thermobox to minimize heat loss. The primary control is programmed in Lab-manager using PID control functions.

Except for the size, the pilot plant reactor has a similar setup as the lab reactor. The reactor is a 2300-L vessel, equipped with cooling jacket and dosing pumps. All necessary sensors are installed. The DCS communicates with the MPC module through an OPC server.

The developed Raman spectroscopy sensor was used for inline measurement of monomer concentration. The Raman sensor was calibrated in lab scale and implemented in lab and pilot scale. The NMPC was implemented using Cybernetica CENIT. It deploys nonlinear state-space models derived from first principles. The process model is a key component of the control function. It has to be precise enough to estimate the necessary properties as well as fast enough to simulate many times faster than the offline model version.

As described in Sect. 4, soft sensors estimate the current state of the process based on the data provided by hard sensors. Subsequently, the model is updated and used by the control module as a starting point for prediction into the future. An optimization is carried out inside the controller to find the best way to achieve the desired objective while considering process variations, and maintaining the process within allowed limitations. The model-based predictive control (MPC) concept is shown in [20].

The implemented control structure can be explained as a three-level hierarchy:

1) On the highest level, an off-line optimization is carried out, as described in Sect. 4. Monomer feed and concentration profiles are calculated, so as to attain the desired morphology while respecting process constraints.
2) The model predictive controller tracks the optimal concentration profiles. It uses the feed rates and jacket inlet temperature as manipulated variables, and optimizes these variables so as to hold the reactor temperature at setpoint and track the pre-calculated concentration profiles with high precision.
3) At a regulatory level, the manipulated variables from the MPC are setpoints for PID controllers

### 5.1 Model Validation

First, the demonstration is conducted in the lab reactor starting with process observation. In this, the process is run conventionally with time-based recipes. The data from these experiments are used to fit the model to the process. For the parameter estimation, the kinetic parameters of the monomers, namely the rate coefficients of propagation and the reactivity ratios, are assumed to be known, whereas those associated with the radical balance are uncertain and are estimated. Fig. 7 shows that the model over-estimates the measurements marginally, but is able to capture the dynamics of the process qualitatively well. After successful qualification of the reference batch, the closed loop controller was activated. Optimization of temperature control using the cooling circuit as degree of freedom, batch time optimization by increasing the feed rates as much as possibly while respecting all process and product property constraints as well as the optimization of polymer structure has been shown in [20], and is not shown here.

### 5.2 Online Control Experiments in Lab and Pilot Scale

Fig. 8 shows the closed loop tracking of the monomers and the parallel control of the reactor temperature, which translates to the indirect control of morphology of the emulsion particles. The feed rates are controlled to achieve the desired concentration profiles in the reactor, and the cooling circuit is adapted to keep the reactor temperature constant. All variables are in a very good agreement with the set-point or given profiles to be tracked. As mentioned earlier, the tracking of pre-defined concentration and reactor temperature profiles is used to achieve the desired particle morphology.

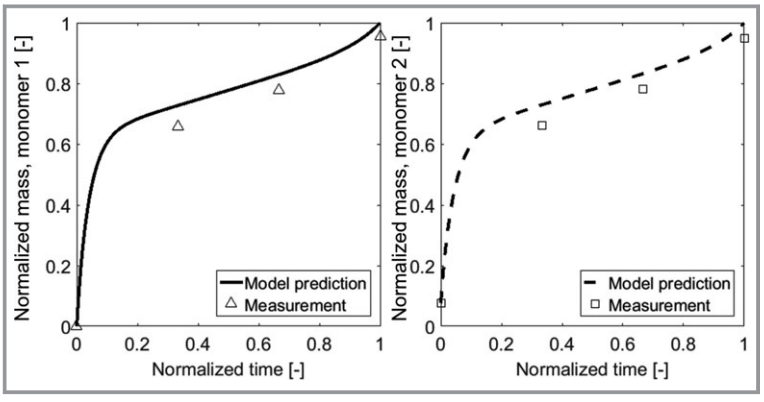


**Figure 7.** Comparison of model prediction and measurements of unreacted monomers in a semi-batch experiment.

  

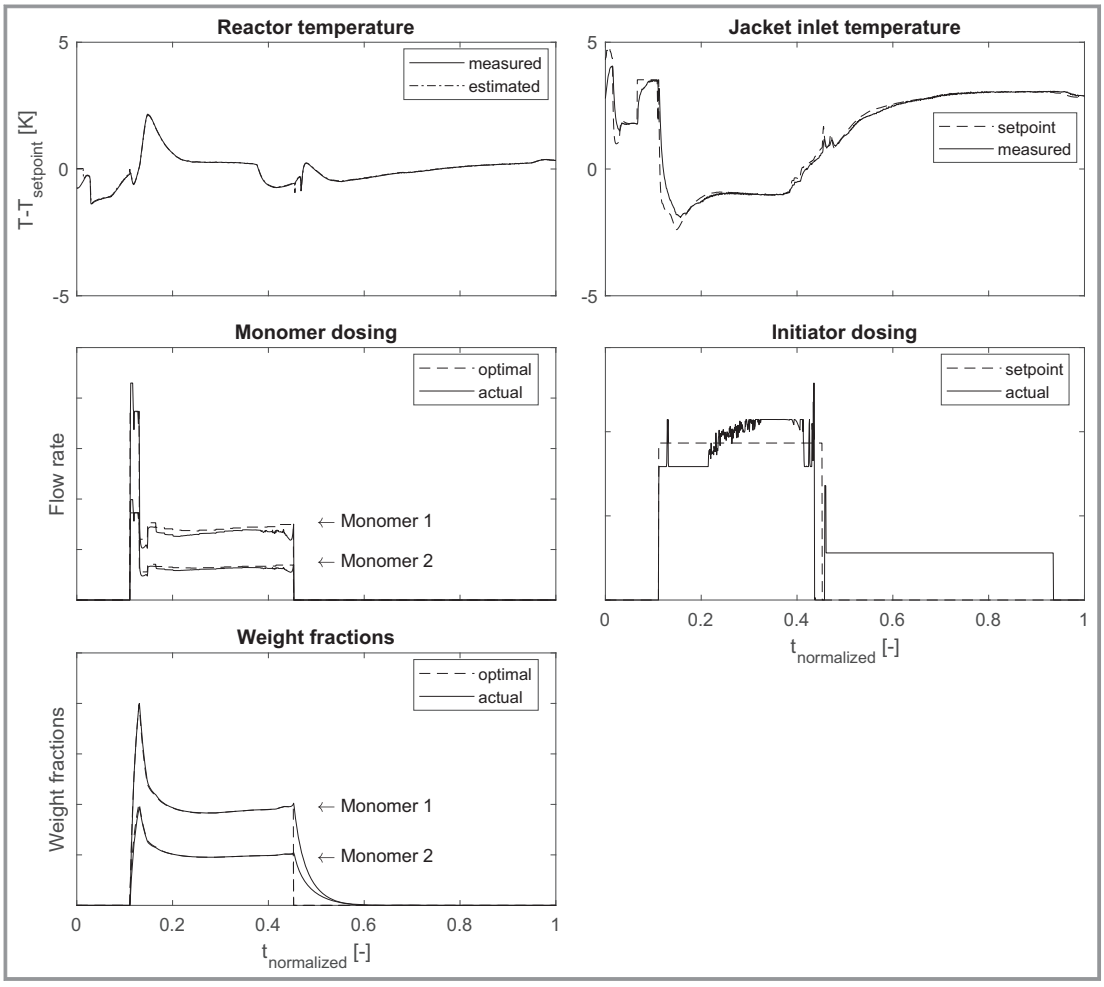


**Figure 8.** Temperature, dosing and concentration evolutions for morphology control batch in lab scale.

Fig. 9 shows the results from one of the pilot plant experiments illustrating closed loop control of emulsion polymerization, including the achievement of the desired morphology via control of the monomer trajectories and reactor temperature. In this pilot scale experiment, the dosing of the initiator was modified from the optimum profile in the beginning of the experiment so that the monomer concentrations are lower than the optimally desired concentration profiles. In this situation, the controller calculates different dosing profiles, keeping the monomer feed rate at high limit up to a point where the concentrations are at optimum. Through this, the NMPC uses the inline Raman measurements for model updates. This illustrates how to compensate for a mismatch between the model and the reality through process disturbances. Thus, the controller operates to keep the batch in-spec by following the inline measurements, which consequently result in a product with the desired quality.

The results show successful demonstration of the closed loop control for tracking concentration/weight fraction profiles of monomers, at the pilot plant reactor.

To evaluate the strength of the developed methods, the newly developed tools are compared with the conventional practice.

Traditionally, the temperature and all raw material feeds, like monomers, initiators, and additives follow a predefined, typically flat profile. The process operation is time based. Such recipes must be robust against any operation disturbances and contain large margins for variations, and an optimization cannot be done individually for each batch. As a result, the time-based recipes need to be somewhat conservative. Fig. 10 shows the resulting distance between the optimum profile and the time-based recipes.

Process disturbances like season's changes, variations in the heat transfer due to deposition of polymers at the walls, fluctuating monomer quality, the monomer concentrations in a standard procedure causes the pre-defined recipe to be non-optimal. By process monitoring with hard and soft sensors, process variations can be tracked and fluctuations observed. Optimal trajectories can be followed allowing for optimum product properties for every batch also during fluctuations. The online controller ensures a process operat-

  

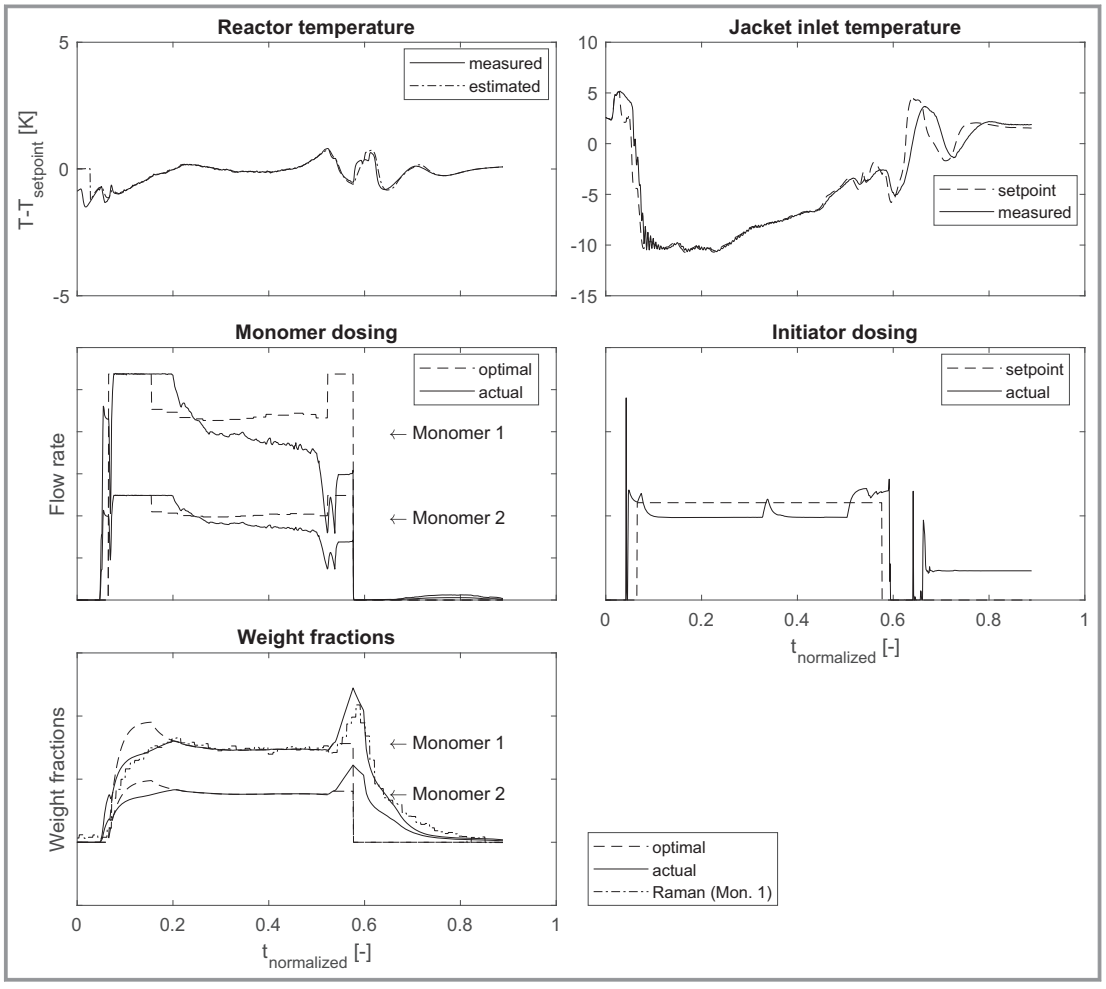


**Figure 9.** Trajectories for monomer and initiator dosing as well as monomer concentration in the demonstration at pilot scale.

ing optimally for given constraints under the actual disturbances. With pre-defined recipes, this is not always possible.

State-based recipes using the developed event-driven approach are superior in all possible conditions to improve the process operations, by increasing the asset effectiveness, reduction of effort to blend bad/good batches, use of accurate amount of educts to operate the batch, as well as the lower energy consumption by operating at optimal temperature, thus positively influencing the sustainability of production processes.

## 6 Conclusion

Due to the demand on faster innovation and the increasing competition, new approaches in R&D are necessary. The current paper illustrates digital tools and methods, i.e., data and models, which support research and development as well as the optimization of production:

- Data obtained by hard sensors provide fast information about the process and can be used to develop and calibrate models describing the polymer structure and particle size and morphology.
- These models are used as soft sensors to get accurate and real-time information about the current state of the process.
- Advanced process control methods can be used for optimization and control for each individual batch to gain an optimum processing.

It has been shown for the second stage of a semi-batch emulsion polymerization process how these ingredients can be utilized to describe and achieve optimum particle morphologies. The hard sensoring of process conditions reveal a huge amount of data during the batch time of many batches. In this work, inline Raman spectroscopy in combination with online calorimetry is discussed to monitor emulsion polymerization. Additionally, the benefits and challenges of using TEM to measure particle morphology are presented. Although promising, the technology for on-

  

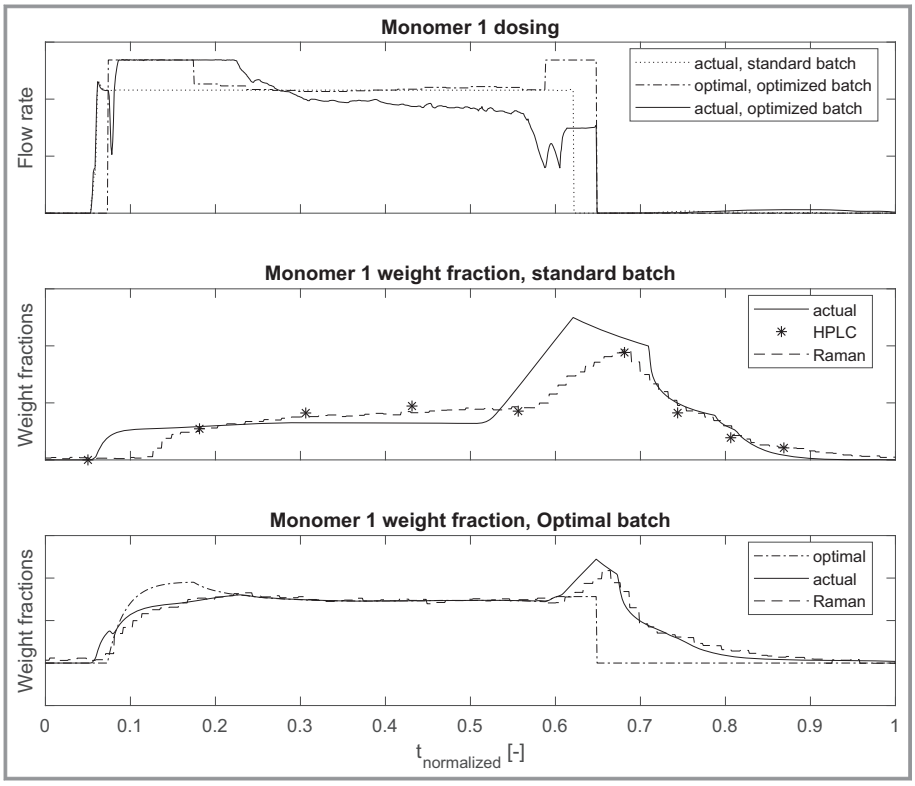


**Figure 10.** Comparison of NMPC batch with conventional practice.

line TEM needs to be further advanced before it can be used for online morphology monitoring. Together with computationally efficient and sufficiently accurate models for the kinetics and particle morphology, the process monitoring is used to determine the optimum operating conditions to produce polymer particles of the desired morphology. These tools can be used in R&D to develop new products exactly matching customer demands.

Furthermore, an online control concept is shown in lab and pilot scale by which the desired particle morphology in a semi-batch emulsion polymerization can be obtained by optimizing the process trajectories. The advantages of the NMPC compared with standard operation concepts is presented.

The technologies demonstrated here show potential for significant benefits in semi-batch plant operation and the demonstration of optimization and NMPC together with Raman spectroscopy for process monitoring in a pilot-plant is a significant step in the direction of event-triggered operation. Realising the full potential of the technologies mentioned requires, however, a big change in mind-set of the chemical industry as changing from time-based operation to event-driven operation requires re-definition of the product quality work and re-training of the plant operating personnel. This switch can be compared to changing from driver-controlled cars to self-driven cars.

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