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Iterative Batch-to-Batch Control of Particle Size Distribution in Semi-batch Emulsion Polymerisation

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Abstract

In this article, the control of particle size distribution (PSD) is discussed as a means for the inferential control of the rheology of emulsion polymers. A controllability assessment is presented through a consideration of the process mechanisms to illustrate the attainability or otherwise of bimodal PSD. The suitability of a batch-to-batch iterative feedback PSD control is demonstrated, which could act in addition to any in-batch feedback control, the latter being less feasible in certain cases, as argued in this article.

1. Introduction

In emulsion polymerisation, the polymer is produced within particles that span the sub-micron size range. The particle size distribution (PSD) is determined by three major particle-level phenomena, namely, nucleation, growth and coagulation, which are influenced by process manipulations (surfactants, monomers, initiators, etc.) [see Figure 1]. The particle-level phenomena interact with each other, and are regulated through an integrated feedback from the PSD,

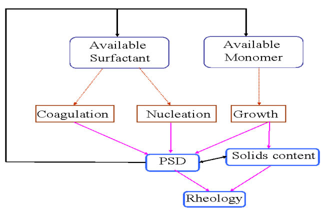

Figure 1: An integrated systems representation of the emulsion polymerisation process and the evolution of the rheology of emulsion polymer

resulting in a highly nonlinear process. The PSD plays a strong role in determining the rheology of the emulsion polymers. In particular, it is seen that the rheology is determined by three factors, namely, the polymer content (solids content) of the latex, the maximum packing factor, and the particle-particle interaction parameter [1]. The effect of PSD on the rheology can be broken down into the effects of the number of modes in the PSD, the sizes of the modes, and the relative amounts of particles in the different modes influence the maximum packing factor directly. The PSD also influences the solids content indirectly via the polymerisation mechanisms. Both of these influence the rheology.

It is evident that there is a non-unique relationship between PSD and rheology, with a considerable range of choices for the PSD that would lead to the desired rheology. Thus, in view of the interrelation between PSD and solids content, and since solids content by itself is an important controlled variable from the perspective of economy, it becomes imperative to simultaneously explicitly consider the interactions between PSD and solids content while determining the target PSD for inferential rheology control. Further, the emulsion polymerisation process places substantial restrictions on the attainable range of PSD in view of both input constraints and inherent process interactions/regulations. This will also need to be considered in determining the target PSD for rheology control. Thus, a combined process model and a rheology model will be needed in the identification of the PSD target that would lead to the desired rheology in the emulsion polymer latex. Once the target PSD that would lead to the desired rheology is identified, the rheology control can be achieved in an inferential manner through control towards this target PSD. The inferential control strategy decomposes the original complex control problem into relatively simpler sub-problems and thereby aids the identification of a desirable solution to the underlying non-convex optimisation problem, and secondly, it enables the control of variables that are not directly measurable [2].

2. Assessment of the Feedback Controllability of PSD

There are several promising methods to measure the PSD of emulsion polymers, including light scattering methods and capillary hydrodynamic fractionation [3]. In addition, by combining PSD measurements with density measurements, it is possible to obtain estimates of other key variables such as total particles and polymer solids content. Most of the methods for PSD measurement require an appreciable solids content to be reached before being able to accurately detect the particles. Secondly, all the methods of PSD measurement have a measurement delay attached to them. For example, the capillary hydrodynamic fractionator (CHDF) needs a solids content of about 3-4% to be reached for reliable PSD measurement, and in typical *ab initio* emulsion polymerisation, starting without any seed particles, it takes about 8-10 minutes to reach such a solids content value. Further, the CHDF has a

measurement delay of about 10-12 min. Thus, it takes about 20 minutes into the batch before a reliable PSD measurement is available as feedback from the process. Assuming that there is only a single PSD analyser connected to the process, a second PSD measurement is not available until about 30 minutes into the batch.
Clearly, the purpose of feedback control is to implement any correction to the open-loop identified operating conditions and feed policies, should a need for such correction be deduced based on measurements. A rapid feedback in the form of frequent measurements will be crucial aid to feedback action. In the emulsion polymerisation process, as explained above, feedback is limited and delayed. The second requirement for feedback control is the existence of a correction (feedback action) to counter the errors introduced in the process, i.e., the controllability. The emulsion polymerisation process has limitations in this regard as well, particularly with PSD as the controlled variable.

2.1. A Potentially Feedback Controllable PSD Class

Figure 2a&b presents an illustrative PSD case where a feedback correction might be possible. The PSD control problem in emulsion polymerisation breaks down into a control of the number of modes, the size of the different modes, and their relative magnitudes. Figure 2a is a target bimodal PSD with clearly separated modes, and Figure 2b presents the profile of total particles that, based on a population balance model in open-loop, is expected to lead to that target

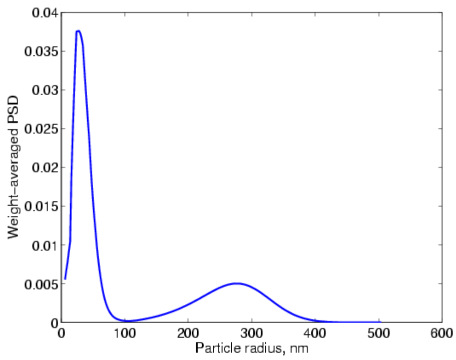

Figure 2a: Bimodal PSD with clearly separated modes

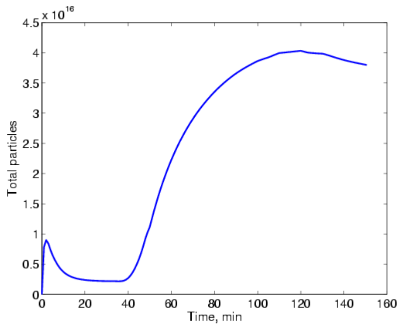

Figure 2b: Profiles of total particles indicating a second nucleation after 40 minutes

PSD. As seen in Figure 2b, the nucleation of the second mode commences at about 40 minutes into the batch, by which time, 2-3 PSD measurements have been obtained from the process as feedback. Using these measurements in combination with density measurements, it is possible to calculate the number of particles nucleated during the first nucleation event (in the first mode). Based on any corrections deduced as necessary for the relative number of particles in the two modes and in the size of the modes, one could devise a feedback action that will correct the growth rates (to correct the size of the odes) and the number of particles nucleated in the upcoming second nucleation event (to preserve the relative amount of particles in the two modes). This feedback action should also ensure that the effect on solids content is not adverse, which is crucial both for rheology control (Figure 1) and from an economics point-of-view. So, the feedback control calculation will be based on three objectives: correction of the size of the modes; preservation of the relative amount of particles in the various modes; conservation of the solids content value.

*2.2. A In-batch Feedback **Un**controllable PSD Class*

As a second example, consider the PSD target shown by dashed lines in Figure 3. In this case, the modes merge into each other unlike in the previous target class, and all the nucleation events have either been completed or have begun before the first sample at 10 min. Thus, a control of the second nucleation to annul the errors in the first nucleation and/or the error in the initial growth rates is less likely in this case. The only resort for this case is to employ a batch-to-batch feedback correction.

3. Batch-to-batch Feedback PSD Control

The methodology of batch-to-batch control has been presented in previous studies, for general batch processes as well as for distribution control problems including those in emulsion polymerisation [4-7]. This is as follows:
(i) The entire first batch is run with the pre-determined open-loop recipe, and the measurements are collected (PSD, density, etc.)
(ii) The measurements are used after the batch to identify the erroneous aspects of the model that was used to compute the recipe for the first batch, and to correct these errors
(iii) The corrected model is used to re-optimise the recipe for the next batch.

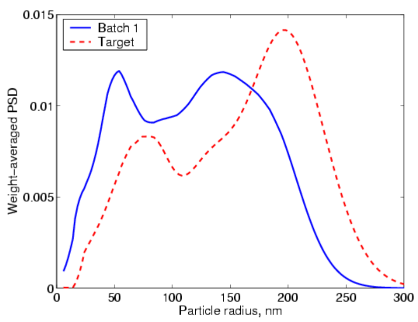

Figure 3a: Iterative PSD control - batch 1

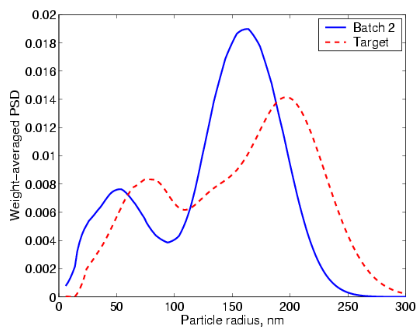

Figure 3b: Iterative PSD control – batch 2

Steps (ii) and (iii) are repeated until the errors in the control objectives are within allowable tolerances. Step 2 typically could take the form of parameter update/correction, based on model sensitivity to the parameters.

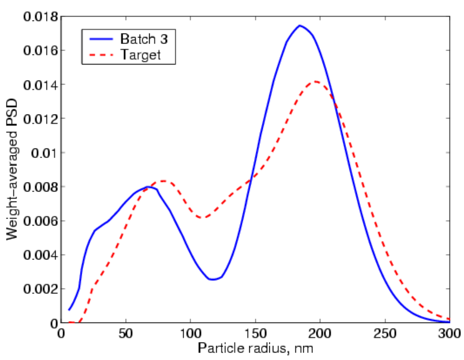

Figure 3c: Iterative PSD control - batch 3

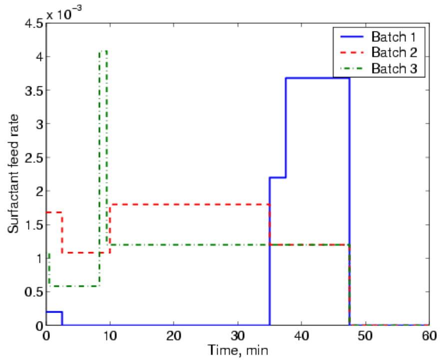

Figure 4: Variation of feed policy in the three batches

Figures 3 and 4 show the implementation of the batch-to-batch PSD control in emulsion polymerisation. The solid lines in Figures 3a-3c are the actual PSD obtained from a hypothetical process, in batches 1, 2 and 3, respectively. The dashed line in these plots is the target PSD. Figure 3a shows that there is an error both in the size of the modes and the relative number of particles in the two modes compared to the target shown by the dashed line. In Figure 3b, the error in the relative amounts of particles in the two modes is already considerably corrected, and this is further correct in Figure 8c. Figure also shows a correction of the sizes of the two modes. Note that, both in view of measurement errors, and also bearing in mind the inferential control goal (of rheology control through control of PSD), a much further refinement may not be warranted in the match of the target. Figure 4 shows the variation of the feed policies of the surfactant solution, process input, over the three batches.

4. Conclusions

The suitability of rheology control via PSD control was shown, to illustrate an inferential control strategy for end-product property control. Inferential control enables control of properties that are not measurable in real time, and in several cases cannot be characterised in short times. Secondly, the strategy decomposes the non-convex problem that underlies the controller into more manageable sub-problems. The identification of the target PSD, for rheology control for instance, necessitates an integrated consideration of the process aspects as well as the property relations. This is in view of (1) the combined influences of PSD and solids content on the rheology, (2) the interactions between PSD and solids content, the latter by itself being an important controlled variable, and (3) the process imposed limitations on the ranges of attainability of PSD. Thus, a combined process model and a property model should be used to solve this pre-control problem to identify the target PSD that would lead to the desired rheology, resulting however in a more manageable feedback control problem. The scheme is relevant for several other end-use polymer properties that are not directly measurable, providing a means for their control.

The emulsion polymerisation process has measurement limitations in that PSD measurements are sparse and appear only after considerable part of the batch has proceeded. Classes of PSD with clearly separated modes, wherein the second nucleation event occurs after a few measurements are obtained, are likely to be feedback controllable. This will be achieved through a control of the second nucleation event (and any subsequent nucleation events), and a control of the growth event. The objective will be to preserve the relative number of particles in the various modes and the size of the modes. However, the discontinuity that governs both the nucleation and growth phenomena, and the strong internal feedback from the PSD on the rate processes, makes it likely, even in these cases, that a feedback control move does not exist. A last hurdle is the need to ensure no adverse effect on solids content while matching the target PSD, again in view of the interrelation of the solids content and PSD. Thus, PSD classes with distinct modes having sufficient time between the nucleation events to obtain PSD measurements are *likely* to be feedback controllable. On the other hand, PSD classes with overlapping modes or those in which there is not enough time for PSD measurements in between nucleation events are to a major extent feedback uncontrollable.

A combined in-batch and batch-to-batch strategy may be employed when in-batch is applicable. In view of the non-convexity in the identification of the target PSD, one could actually choose a target PSD that is also likely to be in-batch feedback controllable.

Acknowledgements

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