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Dynamic Modelling and Simulation of Lactose Cooling Crystallisation: from Batch and Semi-Batch to Continuous Operations

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Abstract

The focus of this work is to revise for semi-batch and continuous operations, the model equations of a seeded batch cooling crystallisation of α-lactose monohydrate. A case study of evaporative semi-batch operation is also included for reference as models of cooling and heating are not much different. In order to achieve the highest yield, dynamic optimisations must be performed on these models to get optimal cooling, feeding and heating (for evaporative mode only) strategies. Applying these strategies in bench scale experiments shows that models of batch and semi-batch work well before the growth becomes slow. Semi-batch is slightly faster than batch and evaporative semi-batch is ten times faster than cooling but is more difficult to control. The performance of a cooling and seeding run in continuous mode is simulated. The system reaches steady state after seven residence times but the predicted particle size could not be stabilised and continued to increase up to one hundred hours. A plug flow reactor is being studied instead of a continuous stirred tank reactor to close the mass and population balances.

Keywords: continuous operation, cooling profile, crystal size distribution, evaporative, semi-batch

1. Introduction

Previous study (Vu et al., 2004) has validated the model equations of a seeded cooling batch process to produce α-lactose monohydrate crystals from highly pure syrup. Using a nonlinear least square method, the best curve fitting of experimental data to the model yielded values of k_g and n, two constants used in the growth rate equation $G = k_g T(\text{driving force})^n$. In order to achieve the highest yield and quality of the product, some process alternatives must be compared, such as cooling against heating, batch and semi-batch against continuous mode. These process alternatives are not possible with the impure feeds currently used in the dairy industry.

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In this paper the process dynamic model of a seeded batch is revised for these operations, using values of k_g and n previously found. Cooling semi-batch is the focus of the work since it is a start-up stage of both batch and continuous operations. Dynamic optimisations are performed to obtain optimal cooling temperature trajectories, which are used as set point profiles in the experiments carried out in bench-scale batch and semi-batch crystallisers. For comparison, dynamic optimisations are also conducted for an evaporative case study to obtain evaporation and feed rates. Comparison of experimental and simulation results validates the computer model, which can be used for industrial scale crystallisations.

Neither continuous nor evaporative lactose crystallisation is commonly practiced in the dairy industry or studied in the literature (Thurlby and Sitnai, 1976; Shi et al., 1990). Based on the results found for semi-batch operation, experimental design and simulation in continuous mode are investigated here to compare with batch and semi-batch. The outcome expected is an alternative process, which produces uniform distribution of fine crystals in the highest yield.

2. Problem Description And Solving Techniques

2.1 Process modelling

The dynamic model of lactose batch cooling crystallisation has been shown in previous work. Only the state equations describing the rate change (kg.hr^{-1}) in mass of water, mass of dissolved ∝- and β-lactose and mass of crystals of pure lactose must be modified for the following operations. In all cases syrup feed is pumped into the vessel at the rate F (kg.hr^{-1}) but the differences are:

- Cooling semi-batch: heat is removed;
- Evaporative semi-batch: steam is supplied at the rate E (kg.hr^{-1}) to evaporate λE kg of water per hour ($1 > \lambda > 0$).
- Continuous flow: seed particles are pumped in with the feed while product and heat are withdrawn.

Under the conditions of no nucleation, agglomeration or breakage, in a batch or semi-batch operation, the number of particles remains constant. In a continuous run, it is assumed that the number of seed particles in the feed stream and the product stream are the same but their average sizes and total masses are different. If this condition is held, at steady state the rate changes in mass of water, dissolved ∝- and β-lactose and crystals of pure lactose approach zero in a continuous operation. The modified equations for cooling and heating semi-batch from cooling batch operation are shown below. The subscript “f” denotes feed. Other state equations and definitions of M and R can be found in Vu et al. (2004). Equations (1) and (1a) define the rate change in mass of water in cooling and heating semi-batch, respectively. In these equations C with corresponding subscripts are the mass fractions of water, ∝- and β-lactose in the feed.

$$\frac{dx_w}{dt} = -0.05R + FC_{wf} \tag{1}$$

$$\frac{dx_w}{dt} = -0.05R + FC_{wf} - \lambda E \tag{1a}$$

Equations (2) and (3) show the rate change in mass of dissolved α- and β-lactose.

$$\frac{dx_\alpha}{dt} = -Mx_\alpha + \frac{Mx_\beta}{K} - 0.95R + FC_{\alpha f} \tag{2}$$

$$\frac{dx_\beta}{dt} = Mx_\alpha - M\frac{x_\beta}{K} + FC_{\beta f} \tag{3}$$

2.2 Problem formulation and solving

2.2.1 Batch and semi-batch

The same technique is applied to optimise performances of a cooling batch, cooling or evaporative semi-batch. To avoid solving the ordinary differential equations (ODEs) required for each recursion, Lagrange interpolation polynomials are used to approximate the state variables x. These approximations can be differentiated and back substituted into the ODEs, which are converted to an algebraic set. The dynamic optimisation problem now becomes a normal nonlinear programming problem and can be solved for an optimal cooling temperature, a feed rate or an evaporation rate profile by any existing optimisation technique.

The constraints commonly imposed on seeded batch crystallisation include solubility and secondary nucleation threshold (SNT) concentration. In particular these relations are used for α-lactose: $sol_{lac} = e^{2.389+0.028T}$ and $sol_{snt} = e^{2.992+0.0196T}$ (Butler, 1998). Below the solubility, crystals dissolve instead of growing. Beyond the upper bound, nucleation occurs, resulting in downstream processing inefficiencies. Working within these limits justifies the assumption that the number of seed crystals remains constant through an operation. Additional constraints are required for an evaporative semi-batch since simultaneous syrup feeding and water evaporation affect the vessel inventory. For instance the total mass (W) in a crystalliser is always within limits (W_i and W_f) and solid content ($sc = x_c/W$) in a crystalliser has an upper limit sc_{fin}, at which circulation will suffer.

The target of both batch and semi-batch problems is the minimum operating time but the objective function set for cooling mode can also be to maximise the growth at a specified time $OF_{cool} = \max\{x_c(t_{fin})\}$. In contrast the objective function of an evaporative semi-batch, referred to as a multiple objective function, is much more complicated as in equation (4). It is expensive to formulate, solve and more difficult to control an evaporative case but as a trade off, batch operating time is ten times faster than cooling.

$$OF_{evap} = \min\left\{ w_t time + w_w \sum_{i=1}^{i=n} \left[W(i) - W_{fin}\right]^2 + w_{sc} \sum_{i=1}^{i=n} \left[sc(i) - sc_{fin}\right]^2 \right\} \tag{4}$$

$w_{t,w,sc}$: weighing constants defined by users

2.2.2 Continuous operation

Simulations of start-up stages are performed first. The initial conditions and cooling strategy applied are similar to batch and semi-batch runs but the feed stream is seeded. Therefore, the number of seed particles in the vessel increases with time. As a result, one additional ODE describing the change in the number of seed crystals must be included in the state equations during the start-up. This ODE can be removed when the product stream is withdrawn from the system. A model of a continuous stirred tank reactor operating at a constant temperature is applied to describe the performance of the crystalliser at steady state.

3. Results And Discussions

Feed syrup at 39°Brix, 60°C and seeds having median size of 12 μm, seeding rate of 1.5% w/w of crystal-free solution were used in all experiments and simulations of cooling or evaporative operations, batch, semi-batch or continuous mode. Cooling crystallisations were conducted in a 2L stainless steel vessel, thermostated between 10°C and 70°C and equipped with a stirrer. Different cooling strategies were applied to evaluate the model. Sampling was conducted every thirty minutes for the initial three hours then hourly for five hours and for the last three hours of each batch (twenty five hours). The samples were analysed for concentration (°Brix), solid content (mass of lactose crystals/mass of content of the crystalliser), particle size (μm) and distribution. For comparison one evaporative run was conducted isothermally in a 4L glass vessel under vacuum, starting at the same initial conditions applied for cooling. Some sets of results are plotted with the simulation results in two following figures.

Three cooling strategies labelled as (1), (2) and (3) in Figure 1 were applied to test the model. The first two are upper and lower cooling limits measured in the crystalliser that can be achieved manually. The cooling profile (3o) is the solution obtained from the dynamic optimisation. However, for possible implementation as set point cooling temperature, this solution is approximated by three first order polynomials, labelled as (3). In the same figure the corresponding solid content profiles show that the model can predict reasonably well the growth for all cooling strategies within two limits. Optimal batch time lies in the range from five to ten hours. Crystal growth flattens after ten hours. Applying the best cooling strategy achieves the highest yield.

The model is extended and applied to other circumstances. Figure 2 plots solid contents and average size for semi-batch cooling (1), evaporative (2) and continuous cooling (3). Solving the dynamic optimisation of a cooling semi-batch case, in which syrup feed is pumped into the vessel, while heat is removed, gives a cooling strategy similar to the batch case at the maximum feed rate. These strategies were applied in simulations and bench-scale runs. Experimental results and prediction on both solid yield and particle size agree well for an eight-hour period as shown in curves (1), upper graphs of Figure 2. After eight hours measured particle size starts to deviate from prediction since it is more difficult to avoid nucleation, breakage and agglomeration in a high solid content solution.

Solving the dynamic optimisation of the evaporative semi-batch case, in which syrup feed is pumped into the vessel, while heat is supplied to evaporate water yields feed and evaporation rate profiles. It was difficult to implement both strategies in a bench scale experiment; therefore heat was supplied at a fixed rate, while feed was pumped in to maintain a constant level in the vessel. The final products were filtered, washed, dried and analysed to compare with prediction. Curves (2) plot simulation results. Between two and four hours, an evaporative run could achieve more than a cooling batch, which operated for more than twenty hours.

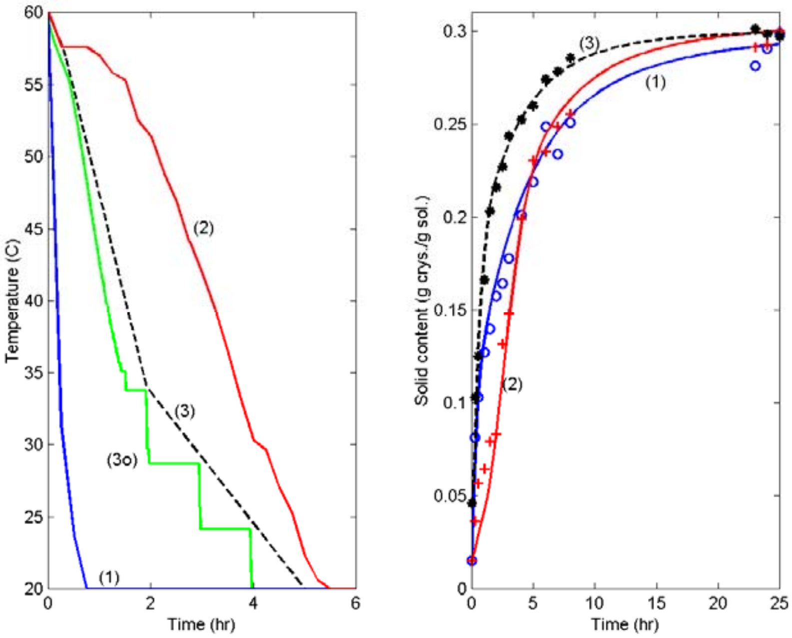

Figure 1. Cooling strategies and solid yields in cooling batch (1: fastest strategy; 2: slowest strategy; 3o: optimal operation; data points represent experimental results)

A continuous run includes two stages. During the start-up stage, seeded syrup feed is pumped in while heat is removed but no product is withdrawn. The first stage, case (3) is similar to the cooling semi-batch case except that seed particles are added in the feed stream. After withdrawing the product, it takes about six or seven residence times for the solid content to reach steady state as shown in the lower graphs of Figure 2. However, after hundred hours the size still increases. This indicates that seed particles are gradually washed out of the tank and nucleation eventually becomes the main source to provide nuclei. The model cannot describe the performance when nucleation dominates the process. A plug flow reactor instead of a continuous stirred tank reactor is being studied to overcome this drawback.

4. Conclusions

The process dynamic model of a seeded batch was revised for cooling and evaporative semi-batch and cooling in continuous mode. Using the values of two constants in the growth rate equation previously found and solving the dynamic optimisation problems gave optimal cooling, feed and evaporation strategies. These policies were implemented in bench scale crystallisations. Experimental results agreed well with predictions in a ten-hour period for cooling batch and semi-batch. Despite a low production yield limited by lactose solubility, cooling batch operation is commonly practiced in lactose factories, as the process requires low capital and maintenance costs, and product quality can be easily manipulated. In contrast, these results suggest that evaporative batch operation is much faster but implementation is more complicated and nucleation is hard to control because the gap between solubility and secondary nucleation threshold concentration is very narrow (from 37°Brix to 40°Brix, at 60°C). A further modelling and experimental investigation is being carried out on continuous cooling crystallisation. A more rigorous model is needed to describe the performance of a continuous cooling and seeding run to close the mass and population balances.

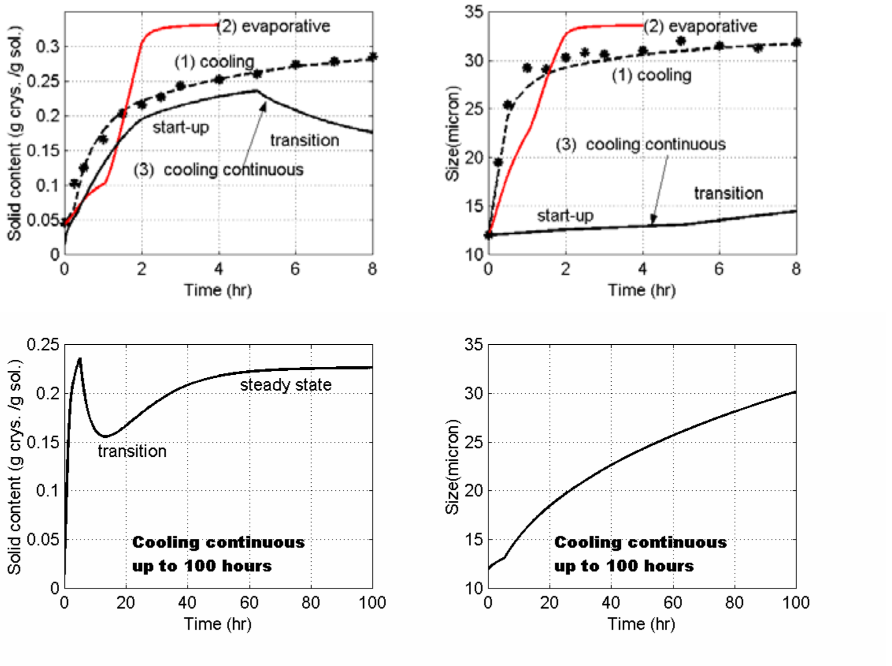

Figure 2. Solid yields and particle sizes in semi-batch cooling, heating and continuous cooling operations (1&2: semi-batch; 3: continuous mode)

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