Nonlinear Controller for Batch Crystallization: Development and Experimental Demonstration

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Particle-size control in crystallization is important for product quality control, as well as plant productivity. Since rapid crystallization traps impurities within the crystal, it is desirable to manipulate solution desupersaturation rates so as to maintain quality. However, the crystal-size distribution (CSD) is influenced by supersaturation levels through complex nucleation and growth kinetics as well as through particulate phenomena such as agglomeration and attrition. Plant productivity is severely hindered when excessive fines are produced since the solid-liquid separation system must be slowed to compensate.

Significant advances have been made toward the development of dynamic CSD measurements, however, these sensors are not as yet widely accepted by industry. As a result, the conventional practice involving the control of seeding or desupersaturation profiles rather than CSD continues. The preferred CSD measurement is obtained from laser light scattering instruments, since these avoid the plugging problems associated with Coulter counters, require little calibration and are easily automated (Rawlings et al., 1993). However, combining the process and light scattering measurement model into a form suitable for model-based control is difficult.

Consider a crystallization and measurement process:

$$\dot{x} = f(x,u,d,p,t) \tag{1}$$

$$y_m = g(x) \tag{2}$$

where for the process model

x = state vector of number of crystals in a size increment
u = vector of manipulated inputs (solute supersaturation level)
d = disturbance variables (such as seed size and mass)
p = parameters (rates of nucleation, growth, agglomeration, breakage crystal shape).

and for the measurement model

y_m = model representation of the distributed measurement vector y from laser light scattering

The full identification of a crystallization process according to model Eqs. 1 and 2 is restricted by structural and parametric plant/model mismatch, mathematical conditioning problems and mixing pattern uncertainties. Structural mismatch is introduced in Eq. 1 by the size-axis discretation scheme used to reduce the population balance to an ODE and into Eq. 2 since the measurement itself is a subset of the process states. Parametric and structural mismatch occurs in Eq. 1 due to the difficulty in modeling, as well as identifying time-varying rate mechanisms representing nucleation, growth, agglomeration, breakage, and so on (Farrell and Tsai, 1994). Finally, the computations associated with reducing the Laser measurement to CSD are not only mathematically ill-conditioned, but also dependent on hydrodynamics, particle shape, spatial distribution of the suspended particles around the sensor and the focal plane of the laser beam (Rawlings et al., 1993; Boxman, 1992).

To avoid some of the conditioning problems associated with developing a model identification and control scheme based upon Eqs. 1 and 2, Witkowski et al. (1990) proposed using the bulk crystallizer transmittance measurement in place of Eq. 2 as a CSD related secondary measurement. Transmittance can be shown to have a one-to-one relationship to the projected crystal area (second moment). Using dynamic transmittance data together with concentration measurements, Rawlings et al. (1993) discuss the successful kinetic model identification of a batch KNO_3-H_2O crystallizer. Using the potassium nitrate model, Miller and Rawlings (1994) formulated a model-based approach to the open-loop computation of the cooling profiles that maximizes final product seed size while minimizing nucleated crystal mass.

The same investigators were not as successful in a study regarding crystallization of naphthalene from toluene. For this system, less accurate model parameter estimates were obtained (Witkowski et al., 1990). Miller and Rawlings (1994) attribute the identification problem to a lower information content of the naphthalene data set. Kinetic parameter identification using batch data has been shown to be problem dependent being especially difficult for higher nucleating systems (Farrell and Tsai, 1994). The proper identification of system models that include kinetic parameters may be too time consuming for many industrial applications.

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Reduced order models that do not require kinetic parameter identification can also be used to develop workable model based control schemes as demonstrated by Rohani and Bourne (1990). Unfortunately in this work, dynamic CSD related measurements were obtained from a sensor measuring fines loop nuclei density based upon calibration of the temperature difference measured before and after fines dissolution in a sample cell, which required considerable development (Rohani and Paine, 1987). To account for the time varying characteristics of the batch process, a minimum variance self tuning controller (MVSTC) was developed. The MVSTC control scheme includes on-line recursive input/output model identification (fines dissolution rate/measured temperature difference) and a minimum variance control law to maintain the desired setpoint (Rohani and Bourne, 1990).

In the present study, we define a reduced order time dependent single-input-single-output model relating transient solubility to the measured LASENTEC mean computed from the distributed particle counts in 38 channels (LASENTEC is available from Laser Sensor Technology Inc., Redmond, WA). Most crystallizations require that growth rates (weight mean) and nucleation rates (number mean) be monitored, suggesting a single-output controlled variable of the form:

$$y_m = \alpha(t) L_{wt} + (1 - \alpha(t)) L_n \tag{3}$$

For growth dominant processes, weight mean ($\alpha = 1$) is adequate.

The model is constructed by correlating successful batches according to $h(y^*(t), u^*(t))$, where for growth dominance: y is the weight mean size as measured by the LASENTEC; u is the crystal solubility.

The slope of the correlation function is used to define a time dependent input-output model of the measurement trajectory:

$$\dot{g}(x) = \Delta y_m = K_p(t)\,\Delta u \tag{4}$$

The generic model control algorithm (GMC) (Lee and Sullivan, 1988) is used to construct a model based control law from Eq. 4.

Review of GMC

As presented by Lee and Sullivan (1988), for setpoint deviations, a reference rate for returning the process to the setpoint ($y^*(t)$) is defined:

$$r^* = k_1(y^* - y) + k_2 \int_0^t (y^* - y)\,dt \tag{5}$$

The direct computation of the control signal u (provided the elements of y and u in the process model are equal) is found directly from:

$$0 = \dot{g}(x) - r^* = \dot{g}(x) - k_1(y^* - y) - k_2 \int_0^t (y^* - y)\,dt \tag{6}$$

where generic constants k_1 and k_2 are chosen to dampen the process output for a specified response. Note that parametric model error is handled in classical GMC by the integral term in the reference rate. For overdamped response specifications, the integral term may not entirely eliminate offset.

For time variant nonlinear models, parameter adaptation has been incorporated into GMC whereby the model parameters in nonlinear models similar to Eq. 1 are updated on-line. Most previous applications involve the parameter update of steady-state models [Eq. 1 is $f(x, u, d, p, t) = 0$]. Mahuli et al. (1992, 1993) studied pH control with updates of parameters related to the titration curve while Pandit et al. (1992) worked on continuous distillation control updating tray efficiency parameters in a nonlinear shortcut model (Eq. 1). Implementation for a nonsteady-state model is reported by Cott and Macchietto (1989) on the temperature control of a batch exothermic reactor incorporating on-line estimation of the heat release.

As an alternate to parameter adaptation, Lundberg and Bezanson (1990) proposed the robust generic model control structure (RGMC) whereby the reference rate is modified for derivative feedback:

$$0 = \dot{g}(x) - r^* - [\dot{y}_m(t) - \dot{y}(t)] \tag{7}$$

As explained by Rani and Gangiah (1991), the RGMC approach will be effective for any closed-loop reference specification whereas the integral correction of Eq. 5 becomes less effective for overdamped reference rate specifications. A successful implementation of RGMC on multilayer bonding presses is presented by Bezanson et al. (1989).

BGMC Applied to Batch Crystallization

For batch processes, we define the GMC controller to follow the reference trajectory [$r^*(t)$] and the desired process trajectory [$\dot{y}^*(t)$], and Eq. 8 is termed batch generic model control (BGMC):

$$\dot{g}(x) - r^* - \dot{y}^* = 0 \tag{8}$$

Substituting Eq. 4 for the model $\dot{g}(x)$ and solving for discrete control action:

$$(\Delta u)_{t_k} = \frac{r^*_{t_k} + (\dot{y}^*)_{t_k}}{(K_p)_{t_k}} \tag{9}$$

Note that the desired trajectory of the batch process measurement is included directly in the control law. Model error is handled as in conventional GMC by the integral action of Eq. 5.

Specifying the reference rate generic constants (k_1, k_2) for a slightly overdamped second-order response has two advantages for batch crystallization: (1) overshoot which can translate into crystal solution as opposed to dissolution is avoided; (2) some offset elimination due to integral action in Eq. 5 is retained.

Experimental

The process involves seeded, cooling-drowning-out batch crystallization of potassium sulfate from an aqueous solution

Table 1. Experimental Conditions

Vessel vol.	0.5 L
Operating temp.	25°C (start), 10°C (end)
Cooling policy	Linear Cooling
Seed size	75–125 μm
Initial saturation (25°C)	0.1195 kg K_2SO_4/kg H_2O
Initial mass of solution	0.5 kg
Time for adding acetone	4,800 s
Acetone addition policy	Constant Addition
Conc. of acetone solution	50 wt.% (1 kg/kg H_2O)
Total 50 wt. % acetone added	110 mL
Open-Loop Seed Mass Perturbation Results	
Seed mass, LASENTEC L_{wt}	0.6 g, 304 μm
Seed mass, LASENTEC L_{wt}	1.2 g, 253 μm

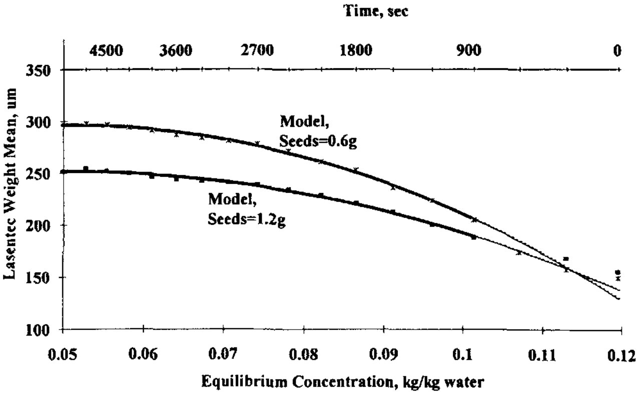

Figure 1. Reduced-order input-output relationship.

using acetone as previously studied by Jones and Teodossiev (1988). Solubility data for potassium sulfate in water-acetone solutions is obtained as a function of temperature and acetone concentration $c(T, x_a)$ from Mydlarz and Jones (1990). Previous work (Jones and Mullin, 1974) indicates the process to be growth dominant (negligible nucleation rates, product weight ratio of seed crystals/nucleated crystals > 99/1). Starting with a saturated aqueous solution at 25°C, each batch is desupersaturated for 80 min by cooling and addition of 50% acetone solution. The LASENTEC mean weight is obtained at 20 s intervals and noise is filtered via a least-squares model estimate obtained by a linear regression of the recursive sequence of the most recent 10 measurements. Outliers (deviation > 3 μm) are rejected by comparison of the data point to the regressed measurement prediction.

For closed-loop runs, the controller is activated 900 s into the batch, to reduce the effect of measurement error at low crystal concentrations when the LASENTEC scan count readings are low. Discrete control is performed thereafter at $T_s = 300$ s intervals, according to Eq. 11 below, resulting in a new setpoint for the crystallizer bath temperature. Differentiating the solubility relationship:

$$(\Delta C)_{t_k} = \left\{\left[\frac{\partial c}{\partial T}\Delta T\right] + \left[\frac{\partial c}{\partial x_a}\Delta x_a\right]\right\}_{t_k} \qquad (10)$$

rearranging to the form of Eq. 9:

$$(\Delta T)_{t_k} = \left(\frac{1}{\dfrac{\partial c}{\partial T}}\right)_{t_k} \left(\frac{r^* + (\dot{L}^*_{wt})}{(K_p)} - \left[\frac{\partial c}{\partial x_a}\Delta x_a\right]\right)_{t_k} \qquad (11)$$

At each control interval, the target trajectory model (Eq. 4) provides $(\dot{L}^*_{wt})$, K_p, L^*_{wt} and r^* is computed according to:

$$r^* = k_1(L^*_{w_t} - L_{w_t})_{t_k} + k_2 \sum_{0}^{NS} (L^*_{w_t} - L_{w_t})_{t_k} T_s \qquad (12)$$

where $NS = (t - 900)/T_s$. Values of $\partial c/\partial T$, $\partial c/\partial x_a$, Δx_a are continually updated based upon batch temperature and acetone concentration.

Results

Referring to Table 1, the effect of growth dominance is reflected by the significant influence of seed mass on the product size. Open-loop runs using low seeds (0.6 g) produces a product with measured mean size 304 μm, whereas high seed mass (1.2 g) reduces the product mean to 253 μm, provided the desupersaturation policies are unchanged.

Figure 1 shows the reduced-order model relationship of mean weight vs. solubility for each of the open-loop runs obtained by correlating data between 900–4,800 s. Computing the slopes:

$$\text{for high seeds}, -450 > Kp > -2{,}760 \quad \mu\text{m}/(\text{kg } K_2SO_4/\text{kg } H_2O)$$

$$\text{for low seeds}, -620 > Kp > -3{,}920 \quad \mu\text{m}/(\text{kg } K_2SO_4/\text{kg } H_2O)$$

Note that the process gain changes by a factor of eight, being less sensitive to solubility near the end of the batch.

Figure 2 compares uncontrolled vs. controlled desupersaturation runs for a desired low seed target trajectory process which has been intentionally overseeded (1.2 g rather than 0.6 g) as a disturbance. For the controlled runs, the model (Eq. 4) is constructed from the desired low seed target. The GMC constants ($k_1 = 0.0033$, $k_2 = 0.000003$) are defined to produce an overdamped second-order response ($\xi \sim 0.96$) (Lundberg et al., 1990). Note that the controlled vessel tem-

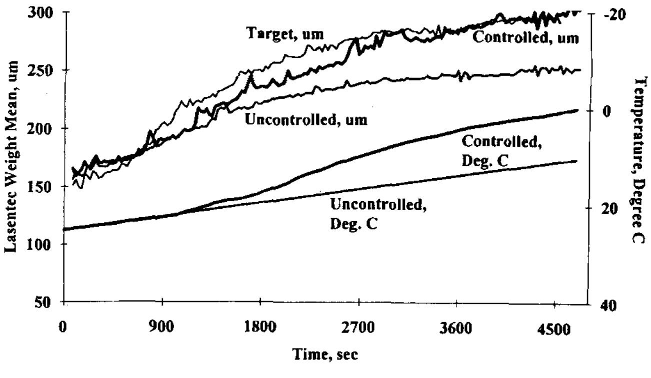

Figure 2. Experimental response to overseeding.

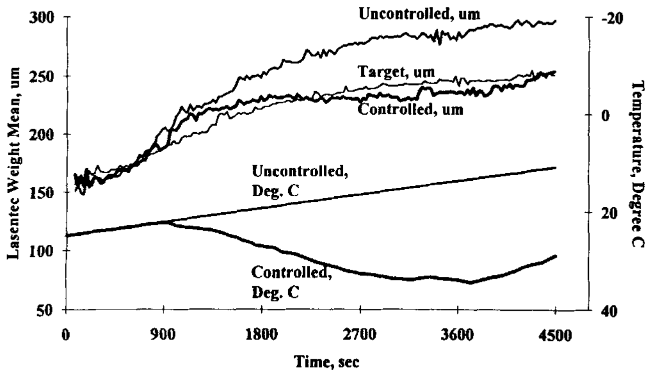

Figure 3. Experimental response to underseeding.

perature is adjusted to cool more rapidly so as to effectively increase growth rate to return to the higher target mean size.

Control in the opposite direction is demonstrated in Figure 3 for a desired high seed trajectory process which has been underseeded (0.6 g rather than 1.2 g). Here, the model (Eq. 4) is constructed from the high seed target. The controller slows cooling to effectively decrease growth rate to return to the higher target mean size.

Conclusions

The generic model control algorithm is successfully applied to a batch crystallization process. The BGMC algorithm utilizes a time variant reduced-order input-output model derived by correlating historical data of solubility vs. weight mean size as measured *in situ* by the LASENTEC scanning laser microscope sensor. Control of the weight mean size trajectory in response to seed disturbances is successfully demonstrated. The approach demonstrates that rigorous mechanistic modeling of process and sensor with regard to the population balance is not necessarily a prerequisite for implementation of model based control.

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Notation

c = equilibrium concentration of solution, kg/kg H_2O
d = system disturbance
f, g, h = arbitrary functions
k_1, k_2 = generic control loop constants
K_p = slope of output vs. input model
L_n = number mean, μm
L_{wt} = weight mean, μm
p = system parameter
r^* = reference rate
t = time, s
T = temperature, °C
T_s = control interval, s
u = system input
x = system state variable
x_a = weight fraction acetone in solution
y = system output
α = measurement weighting factor
ξ = damping coefficient

Subscripts and superscript

k = discrete time index
m = model
$*$ = setpoint

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