FEEDFORWARD CONTROL OF BATCH CRYSTALLISERS - AN APPROACH BASED ON ORBITAL FLATNESS

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Abstract: This contribution addresses the problem of feedforward control design for batch crystallisers based on moment models. Moment models, which are standard in the crystallisation literature, are shown to be orbitally flat, i.e. they are flat after an appropriate time scaling. The reciprocal of the crystal growth rate serves as the time scaling function such that a new notion of 'time' is defined by the increase in crystal length. For any desired final crystal size distribution (CSD) which is compatible with the crystalliser model it is possible, exploiting flatness, to analytically compute the corresponding temperature trajectory.

Keywords: batch crystallisation, population balance, orbital flatness, process control

1. INTRODUCTION

The quality of crystalline products in the chemical and pharmaceutical industry is not solely determined by their chemical composition but also by physical properties, primarily by the crystal size distribution (CSD). Therefore, the manufacturing of crystalline material with a pre-defined CSD is one of the current challenges in industrial crystallisation (Rawlings *et al.*, 1993). In batch cooling crystallisers, the CSD can be influenced by the cooling strategy, i.e. the temperature-time-profile, applied to the crystalliser.

The temporal evolution of CSD in the domain of time and crystal size can be modelled by a partial differential equation (PDE) referred to as the *population balance equation* (Randolph and Larson, 1988; Ramkrishna, 2000). This PDE is coupled to one or more ordinary differential equations (ODE) for, e.g., concentration and temperature of the liquid phase. Under certain conditions, a finite-dimensional model can be derived from this infinite-dimensional system. The resulting model does not describe the evolution of the entire CSD, but only of a number of its moments.

Based on a population balance model, it is of course possible to determine the final product CSD for a given temperature profile by simulation. The inverse problem, i.e. the design of a feedforward control which produces a desired CSD, is an area of active research. The most common approach is to use optimisation techniques in order to determine a temperature profile that produces a CSD which is optimal with respect to some cost function. (Jones, 1974) applied Pontryagin's maximum principle to obtain an optimal cooling policy. Dynamic optimisation was applied, for example, in (Miller and Rawlings, 1994; Lang *et al.*, 1999; Chung *et al.*, 1999; Zhang and Rohani, 2002).

In this contribution, optimisation techniques are *not* used. Instead the feedforward control problem

for batch cooling crystallisers is solved exploiting *differential flatness* (Fliess *et al.*, 1992; Fliess *et al.*, 1995*b*). The batch crystalliser model can be shown to be flat after an appropriate time scaling. Such systems are called *orbitally flat* (Fliess *et al.*, 1995*a*; Respondek, 1998; Guay, 1999). It is well known that flat systems possess a certain invertibility property and, therefore, trajectory planning and feedforward control design can be done in a very elegant way (Rouchon *et al.*, 1993).

This paper is organised as follows. In the following section, orbital flatness is defined. In Section 3, a population balance model for a batch cooling crystalliser is presented and a moment model for this system is derived. In Section 4, orbital flatness of the moment model is established. Furthermore, it is shown that, based on orbital flatness, a temperature profile can be computed which produces any given final CSD compatible with the model. This is, to the authors' knowledge, the first time this problem has been solved for batch crystallisation.

2. ORBITAL FLATNESS

The notion of flatness can be mathematically defined in a differential algebra setting (Fliess *et al.*, 1995*b*) or in the context of differential geometry of infinite jets and prolongations (Fliess *et al.*, 1999). In this section, a less formal, more intuitive definition of flatness is given (Rothfuß *et al.*, 1996). A finite-dimensional dynamic system

$$\dot{x}(t) = f(x(t), u(t)),\ x(t) \in I\!R^n,\ u(t) \in I\!R^m \quad (1)$$

is called *differentially flat*, or simply *flat*, if there exists a fictitious output $y(t) \in I\!R^m$ which satisfies the following conditions.

A The output $y(t)$ can be expressed as a function of the system state $x(t)$ and input $u(t)$ and finitely many time derivatives of the input

$$y(t) = \Phi\left(x(t), u(t), \dot{u}(t), \dots u^{(\alpha)}(t)\right). \quad (2)$$

B Reversely, the system state and input can be expressed as functions of the output $y(t)$ and finitely many of its time derivatives

$$x(t) = \Psi_1\left(y(t), \dot{y}(t), \dots, y(t)^{(\beta)}\right) \quad (3\text{-a})$$

$$u(t) = \Psi_2\left(y(t), \dot{y}(t), \dots, y(t)^{(\beta+1)}\right) \quad (3\text{-b})$$

The fictitious output $y(t)$ is then called a *flat output*. It completely determines the dynamic behaviour of system (1). If a sufficiently smooth trajectory of the flat output is given, the trajectories of the entire system state $x(t)$ and the system input $u(t)$ are determined by (3-a) and (3-b), respectively. They can be computed without solving a differential equation. Therefore, it is obvious that flatness is a property which facilitates the problems of trajectory planning and design of feedforward control considerably. Since the flat output constitutes an algebraic parameterisation of the system's dynamics, flatness also is a particularly useful property for the solution of dynamic optimisation problems.

Allowing an appropriate state-dependent time scaling, the advantages of flat systems can be extended to a somewhat larger class of systems (Fliess *et al.*, 1995*a*; Respondek, 1998; Guay, 1999). A new 'time'-variable τ is defined as follows:

$$[t_0 \quad t_{end}] \mapsto [\tau_0 \quad \tau_{end}]$$
$$\frac{dt}{d\tau} = s(x(t), u(t)), \quad \tau(t_0) = \tau_0. \quad (4)$$

The mapping of t to τ is bijective iff

$$0 < s(x(t), u(t)) < \infty, \quad \forall t. \quad (5)$$

This makes the time transformation invertible; hence, a control law $u(\tau)$ designed in new time τ can be transformed back and applied in real time t. In new 'time', the system (1) evolves according to

$$\frac{dx}{d\tau} = \underbrace{f(x(\tau), u(\tau))s(x(\tau), u(\tau))}_{=:g(x(\tau), u(\tau))}. \quad (6)$$

If the time scaled system (6) is flat then the original system (1) is called *orbitally flat*.

3. BATCH CRYSTALLISER MODEL

A batch cooling crystalliser (Figure 1) is operated as follows. Initially, the crystalliser contains undersaturated solution. As the solution is cooled, it becomes supersaturated. At this point, small seed crystals may be added. Supersaturation drives the formation of new crystals and the growth of existing crystals. Both processes, nucleation and growth, consume solute from the solution such that the concentration decreases while the amount of crystalline material increases. At the end of the batch the vessel is discharged and the product undergoes further processing steps such as filtering and drying. Product quality as well as the efficiency of downstream processing is heavily influenced by the CSD.

For modelling purposes, the size of crystals is defined by a characteristic length L. A number density function $f(L, t)$ representing the number of crystals per crystal length and volume of slurry formalises the concept of CSD (Randolph and Larson, 1988).

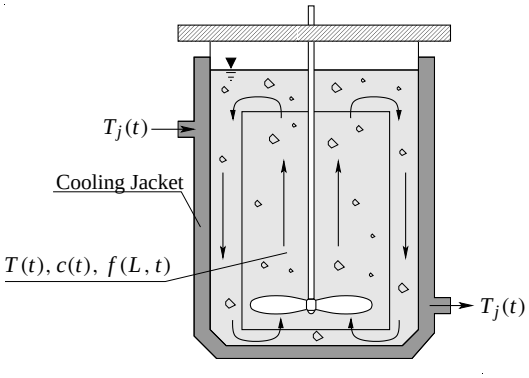

Fig. 1. Sketch of a batch cooling crystalliser

In the following, a standard model for batch crystallisers (Miller and Rawlings, 1994) is presented. It is assumed that all crystals grow at the same rate, i.e. the growth rate G is independent of crystal size, and that nuclei have a negligibly small size. Writing down a balance equation for the number of crystals in an infinitesimal interval of crystal length, a PDE is obtained which, together with appropriate initial and boundary conditions, describes the temporal evolution of the CSD

$$\frac{\partial f(L,t)}{\partial t} = -\frac{G(t)\partial f(L,t)}{\partial L} \tag{7-a}$$

$$f(0,t) = \frac{B(t)}{G(t)} \tag{7-b}$$

$$f(L,0) = f_{seed}(L). \tag{7-c}$$

The nucleation rate is denoted by $B(t)$. The size distribution of seed crystals is $f_{seed}(L)$. Equation (7-a) is called the *population balance*. An ordinary differential equation for the solute concentration $c(t)$ is obtained from the mole balance for the liquid phase

$$\frac{dc(t)}{dt} = -3\rho_c k_v h \int_0^\infty L^2 G(t) f(L,t)\, dL, \tag{8}$$

where ρ_c is the density of crystals, h is a conversion factor (volume of slurry per mass of solvent) and k_v is a volume shape factor defined such that the volume of a crystal with length L is $V_{crystal}(L) = k_v L^3$.

The dependence of (secondary) nucleation and growth rates on supersaturation $S(t)$ and CSD $f(L,t)$ is empirically modelled by

$$G(t) = k_g S(t)^g \tag{9}$$

$$B(t) = k_b S(t)^b k_v \int_0^\infty L^3 f(L,t)\, dL, \tag{10}$$

with supersaturation

$$S(t) = \frac{c(t) - c_{sat}(t)}{c_{sat}(t)}. \tag{11}$$

The saturation concentration c_{sat} is modelled by the empirical relation

$$c_{sat}(t) = A_0 + A_1 T(t) + A_2 T(t)^2. \tag{12}$$

The parameters k_g, k_b, g and b depend on the material to be crystallised, the crystalliser geometry and operating conditions whereas A_0, A_1 and A_2 are solely determined by the combination of solute and solvent substances.

According to (9)-(12), the rates of nucleation $B(t)$ and growth $G(t)$ depend on the temperature $T(t)$. Hence, $T(t)$ can be used as a manipulated variable to affect the CSD. As the temperature $T(t)$ cannot be influenced directly, a subsidiary feedback controller may be used which controls the temperature $T(t)$ by manipulating the cooling jacket temperature $T_j(t)$.

From population balance (7-a) with boundary condition (7-b), a set of ODEs for the moments of the CSD

$$\mu_i(t) := \int_0^\infty L^i f(L,t)\, dL, \quad i = 0, 1, 2, \ldots \tag{13}$$

can be derived. The zeroth moment $\mu_0(t)$ gives the overall number of crystals. The second moment $\mu_2(t)$ is proportional to the overall crystal surface, and the third moment $\mu_3(t)$ is proportional to the volume of the crystalline material in the crystalliser.

Since the duration of the batch, t_{end}, is finite and the growth rate $G(t)$ is bounded it follows that the size of crystals is bounded also. Hence, there is a maximum length L_{max} such that the following is true

$$f(L,t) = 0, \quad \forall L > L_{max}. \tag{14}$$

Consequently, by partial integration it follows from (7-a), (7-b) that

$$\frac{d\mu_0(t)}{dt} = B(t)$$
$$\frac{d\mu_i(t)}{dt} = iG(t)\mu_{i-1}(t), \quad i = 1, 2, \ldots \tag{15}$$

Since the overall mass of solute in the crystalliser is constant, an additional algebraic equation can be derived, relating the third moment $\mu_3(t)$ and the solute concentration $c(t)$

$$c(t) = c_0 + \rho_c k_v h(\mu_{3,Seed} - \mu_3(t)), \tag{16}$$

where c_0 is the initial solute concentration and $\mu_{3,Seed} := \int_0^\infty f_{seed}(L)\, dL$ is the third moment of the seed CSD. Note that the integral expression in (10) is the third moment, i.e. $B(t) = k_b k_v S(t)^b \mu_3(t)$. Because of (9),(11),(12) and (16),

$B(t)$ and $G(t)$ are entirely determined by $\mu_3(t)$ and $T(t)$. Hence, the differential equations for the first four moments can be written as

$$\frac{d\mu_0(t)}{dt} = B(\mu_3(t), T(t)) \quad (17\text{-a})$$
$$\frac{d\mu_1(t)}{dt} = G(\mu_3(t), T(t)) \cdot \mu_0(t) \quad (17\text{-b})$$
$$\frac{d\mu_2(t)}{dt} = 2G(\mu_3(t), T(t)) \cdot \mu_1(t) \quad (17\text{-c})$$
$$\frac{d\mu_3(t)}{dt} = 3G\left(\mu_3(t), T(t)\right) \cdot \mu_2(t), \quad (17\text{-d})$$

This constitutes a simplified model for the batch crystalliser. It is clearly nonlinear, but finite-dimensional. The moments $\mu_0(t) \dots \mu_3(t)$ are the system states, and temperature $T(t)$ serves as the control input. This model exactly describes the dynamics of the moments of the CSD but it does, of course, not describe the evolution of the entire CSD. However, as mentioned above, the moments have a clear physical meaning and for many applications they represent the most important aspects of the CSD.

4. FEEDFORWARD CONTROL

4.1 *Flatness of crystalliser model*

In this paragraph, it is demonstrated that system (17-a)-(17-d) is orbitally flat. With scaling function

$$s(t) = \frac{1}{G(\mu_3(t), T(t))}, \quad (18)$$

a new notion of time is defined by

$$d\tau = G(\mu_3(t), T(t))dt, \quad \tau_0 = 0. \quad (19)$$

As G represents the crystal growth rate, the new 'time' τ is the increase in length which crystals have gained since the beginning of the batch. This is a very natural way to describe the progression of the batch. According to (5), the scaling function has to be strictly positive and finite. Because of (9) and (11), this implies that the liquid in the crystalliser has to be kept supersaturated, i.e. $c(t) > c_{sat}(t)$, $\forall t$. Since in a crystalliser crystals are to be grown rather than dissolved this condition makes eminent sense from a practical point of view. Using new time τ, the system (17-a)-(17-d) is transformed to

$$\frac{d\mu_0(\tau)}{d\tau} = \frac{B(\mu_3(\tau), T(\tau))}{G(\mu_3(\tau), T(\tau))} \quad (20\text{-a})$$
$$\frac{d\mu_1(\tau)}{d\tau} = \mu_0(\tau) \quad (20\text{-b})$$
$$\frac{d\mu_2(\tau)}{d\tau} = 2\mu_1(\tau) \quad (20\text{-c})$$
$$\frac{d\mu_3(\tau)}{d\tau} = 3\mu_2(\tau). \quad (20\text{-d})$$

It is now shown that for the output

$$y(\tau) = \mu_3(\tau) \quad (21)$$

both conditions A and B in the definition of flatness hold. As $\mu_3(\tau)$ is a state variable, (2) and therefore requirement A hold trivially. Differentiating the output $y(\tau)$ four times with respect to τ yields

$$\frac{dy(\tau)}{d\tau} = 3\mu_2(\tau) \quad (22\text{-a})$$
$$\frac{d^2y(\tau)}{d\tau^2} = 6\mu_1(\tau) \quad (22\text{-b})$$
$$\frac{d^3y(\tau)}{d\tau^3} = 6\mu_0(\tau) \quad (22\text{-c})$$
$$\frac{d^4y(\tau)}{d\tau^4} = 6\frac{B(\mu_3(\tau), T(\tau))}{G(\mu_3(\tau), T(\tau))}. \quad (22\text{-d})$$

From (21), (22-a)-(22-c) it is obvious that the states $\mu_3(\tau) \dots \mu_0(\tau)$ can be computed from $y(\tau)$ and its first three derivatives. The input $T(\tau)$ can be determined from (22-d) by additionally using the fourth derivative. Hence, equations (3-a), (3-b) and therefore requirement B also hold. Consequently, $y(\tau)$ is a flat output, and the transformed system (20-a)-(20-d) is flat. This implies orbital flatness of the moment model (17-a)-(17-d) in original time.

4.2 *Control synthesis procedure*

In the following, two characteristics of the batch crystalliser model are exploited to facilitate feedforward control design. These characteristics are, on the one hand, the orbital flatness property of the moment model and, on the other hand, the simple form of the population balance equation (7-a) when rewritten in new time τ. Applying the time scaling (19) to the original PDE (7-a) yields the simple transport equation

$$\frac{\partial f(L,\tau)}{\partial \tau} = -\frac{\partial f(L,\tau)}{\partial L}. \quad (23)$$

This implies that $f(L,t)$ is constant on straight lines in the (L,τ)-domain with $\frac{dL}{d\tau} = 1$, see Figure 2. Furthermore, the size distribution $f(L,\tau)$ can be split into two parts, where one part represents grown seed crystals

$$f_s(L,\tau) = f(L,\tau), \quad \text{for } L \geq \tau \quad (24)$$

and the other part describes the distribution of crystals produced by nucleation

$$f_n(L,\tau) = f(L,\tau), \quad \text{for } L < \tau. \quad (25)$$

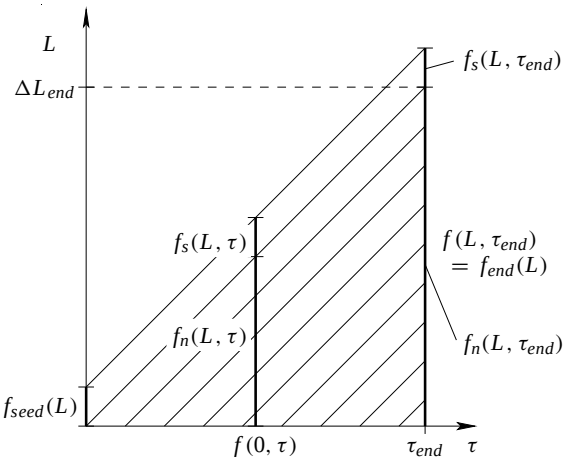

Fig. 2. Evolution of CSD in the (L,τ)-domain

Obviously, the distribution of grown seed crystals $f_s(L,\tau)$ cannot be influenced by control since it is equivalent to the initial seed distribution shifted in size by the length $\Delta L = \tau$, i.e. $f_s(L,\tau) = f_{seed}(L+\tau)$. In contrast, the distribution of particles created by nucleation $f_n(L,\tau)$ can be influenced by appropriate manipulation of the crystalliser temperature $T(\tau)$, since nucleation rate $B(\tau)$ depends on temperature. Consequently, a necessary condition for a desired CSD at the end of the batch $f_{end}(L)$ to be attainable is

$$f_{end}(L) = f_{seed}(L - \Delta L_{end}), \quad \text{for } L \geq \Delta L_{end}, \tag{26}$$

where $\Delta L_{end} = \tau_{end}$ is the increase in size which a crystal gains over the whole batch run.

Since $f(L,\tau)$ is constant on the characteristic lines $\frac{dL}{d\tau} = 1$, the values of a desired $f_{end}(L)$ in the size range $0 \leq L < \Delta L_{end}$ can be traced back to values of the CSD at the lower boundary of the size range $f(0,\tau)$ for $0 < \tau \leq \tau_{end}$. Consequently, if the CSD at the end of the batch is fixed to a certain desired distribution

$$f(L,\tau_{end}) = f_{end}(L), \tag{27}$$

then the time profile of the boundary condition that is necessary to produce the desired distribution $f_{end}(L)$ is determined by

$$f(0,\tau) = f_{end}(\tau_{end} - \tau), \quad 0 < \tau \leq \tau_{end}. \tag{28}$$

Note that τ_{end} is equivalent to the maximum length of nucleated crystals ΔL_{end}, which is also fixed when choosing the desired final CSD $f_{end}(L)$. Because of (28) the desired $f_{end}(L)$ determines the trajectory of the boundary condition (7-b) and according to (22-d) this determines the fourth derivative of the flat output

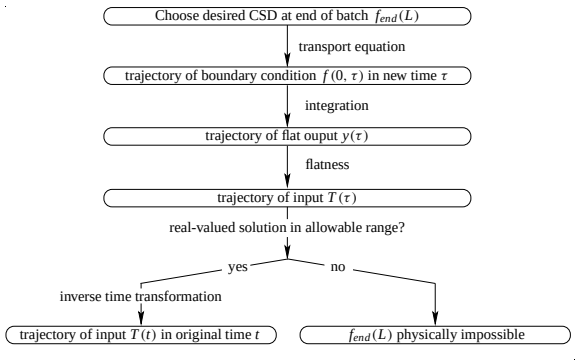

Fig. 3. Open Loop Control Design Procedure

$$\frac{d^4 y(\tau)}{d\tau^4} = 6 f_{end}(\tau_{end} - \tau). \tag{29}$$

The flat output as a function of new time τ is obtained by integrating (29) four times. According to the definition of flatness, all system states and the system input can be determined from the flat output and its derivatives. In particular, the feedforward control $T(\tau)$ which produces the desired final CSD $f_{end}(L)$ can be computed. Eventually, the time transformation (19) has to be inverted to obtain the control $T(t)$ in original time. Depending on the parameterisation of the flat output, the time transformation may not be invertible analytically. Therefore, the inversion may have to be done numerically. In these cases, the open loop control $T(t)$ is determined at a number of time instances but not as an explicit function of time. However, this is not a severe restriction for practical implementation.

Due to the quadratic expression in (12), the solution of (22-d) yields two results for $T(t)$ of which at most one is physically meaningful. If both results are not meaningful, e.g. in the case of complex conjugate solutions, this implies that the desired CSD $f_{end}(L)$ is not compatible with the model, i.e. it cannot be produced by the system from the given initial CSD $f_{seed}(L)$.

Summarising the results of this section, a procedure is obtained which makes it possible to check whether a desired final CSD $f_{end}(L)$ is physically possible and, if so, to compute the corresponding feedforward control $T(t)$. This procedure is depicted schematically in Figure 3. It works as follows. First, a desired CSD $f_{end}(L)$ complying with (26) is chosen. Then, because of the simple structure of the time-transformed population balance, the trajectory of the boundary condition $f(0,\tau)$ in new time is determined by (28). The computation of the flat output $y(\tau)$ basically requires integrating the boundary condition four times. Then, by flatness, the temperature trajectory $T(\tau)$ can be obtained from $y(\tau)$ and its first four derivatives. Finally, the time transformation (19) is inverted

to obtain the feedforward control $T(t)$ in original time.

5. CONCLUSION

It was shown that moment models for batch crystallisation, which are commonly used as a basis for design of feedforward control for these processes, are orbitally flat. The state dependent time scaling function used to render the system flat is physically meaningful: it represents the growth rate of crystals. This leads to a new notion of 'time' which is very natural for the crystallisation process, namely the increase in length of crystals.

The flatness property of the model was exploited for feedforward control design. A procedure was presented to check whether a desired product CSD is achievable and, if so, to *analytically* compute the corresponding control signal, i.e. the temperature trajectory, producing this specific CSD. This is a problem which, so far, has been unsolved in batch crystallisation.

Due to the fact that the flat output completely parameterises the system dynamics, flatness also greatly facilitates the problem of dynamic optimisation of the CSD. Conventionally, the control signal is parameterised, e.g. piecewise linearly. In each optimisation step, the cost function, depending on the final CSD, is determined by numerical solution of the system's differential equations. In a flatness based approach, in contrast, the desired final CSD can be parameterised directly. The corresponding trajectories of system states and input are computed as shown in Section 4 without solving differential equations. Thus, the dynamic optimisation problem is reformulated such that it does not involve the solution of differential equations. This constitutes a significant simplification of this type of problems (Guay *et al.*, 2001). Details on flatness based optimisation of batch crystallisers are presented in (Vollmer and Raisch, 2003).

Furthermore, future work will focus on the extension of the methods presented in this contribution to less restrictive models. In particular, the case of a crystal growth rate that depends on crystal size is of primary interest.

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