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Nonlinear model reduction of a continuous fluidized bed crystallizer

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ABSTRACT

This work considers a system of a fluidized bed crystallizer and an ultrasonic attenuator, which separates an enantiomer from a liquid solution. A population balance model of the system shows autonomous oscillations over a wide range of operation conditions. Proper orthogonal decomposition is applied to obtain nonlinear reduced models of low order. An *a posteriori* error estimator is used to assess the quality of the reduced model during run time.

1. Introduction

Enantiomers are a class of molecules that appear in two variants, which have the same sum formula and consist of the same molecular groups, but with an opposite orientation in space, like mirror images of each other or like right and left hand. Enantiomers occur quite frequently and are of great importance for the food industry, chemical industry, and pharmaceutical industry, because the two enantiomers of a substance (often called D- and L-enantiomer) may have completely different effects on biological organisms [1,2]. For example, they may differ in taste, or when used as drugs, one enantiomer may have a desired physiological effect, whereas the other is inactive or even toxic. Usually, chemical synthesis produces a 50:50% mixture of both enantiomers, a so-called racemate, which has to be separated into the two enantiomers in subsequent purifying steps. As both enantiomers have identical physical properties like density or boiling point, the separation task is challenging. Classical techniques like distillation are not applicable, but more advanced approaches like adsorption, membrane separation, or crystallization are needed [3,4].

A set-up for selective crystallization of enantiomers was recently investigated in [5]. It was shown that crystals of both enantiomers can be recovered continuously in high purity by coupling two crystallizers in a suitable way. In this work, a simplified scheme containing only one crystallizer is studied. Fig. 1 shows a sketch of the scheme. The system consists of a fluidized bed crystallizer and an ultrasonic attenuator connected to the bottom of the crystallizer. The simplified scheme allows the generation of crystals of one of the two enantiomers, while the counter enantiomer remains in the liquid solution. This desired behavior is achievable by feeding the crystallizer with a solution that contains seeding crystals of only one enantiomer. If the conditions are chosen such that no new crystals can nucleate, but only existing crystals can grow, then

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crystals of one enantiomer grow selectively in the crystallizer. The ultrasonic attenuator has the task to remove large crystals, which have sunk to the bottom, to break them into smaller fragments and to recycle them to the crystallizer.

The intended process behavior is only achieved in a small operation window, and keeping the process in the desired state is quite delicate. A successful process design requires a thorough understanding of the complex interactions between fluid dynamics, crystal formation, and crystal growth.

Detailed first principle models are needed to provide the required insight. The behavior of the liquid phase may be described by the Navier–Stokes equations in combination with mass and energy balances, or by simplified continuous models using idealizing assumptions for the flow field. The particle phase may be modeled by tracking the movement of an ensemble of particles in the flow field in the most detailed case. More often, population balances are formulated instead, assuming an infinite number of particles in the system [6]. This adds internal coordinates for the particle properties – like crystal size or crystal composition – to the external or geometrical coordinates of the system. In all cases, the solution of the detailed physical models is expensive and time consuming [7]. For process design by means of mathematical optimization, for controller design, or for online applications like model predictive control or state estimation, these models are less suited [8]. There is a need for reduced models with smaller computational demands.

Model reduction of systems with particle populations in fluid flow has been studied in many publications. In most cases, the internal and external coordinates are treated separately. Reduction of the external coordinates is achieved by simplifying assumptions on the flow field and the spatial gradients, and by dividing the space domain into compartments with different idealized flow fields like perfect mixing or plug flow. For a condensed description of the system's dependence on internal coordinates, generalized methods of moments are most widely used [9]; in this case, the reduced model does not preserve the full information on the particles' property distributions, but only on some of their moments. The application of most model reduction methods requires in-depth physical process understanding and experience with the reduction technique from the user. More easily applicable reduction techniques are desirable, with the potential of a largely automated reduction step. Model reduction by proper orthogonal decomposition [10,11] is a promising candidate for automatic reduction of particle system models. It has been applied successfully to crystallization and granulation processes [12,13]. The new challenge of the fluidized bed crystallizer considered in this work lies in the feedback of the particle phase on the fluid flow, which causes a bidirectional coupling between continuous phase and particle phase. As another new aspect in this work, a novel *a posteriori* error estimation method from [14] is tested for population balance systems.

The work is structured as follows. Section 2 introduces a population balance model of the crystallization system, which serves as a starting point and reference for the model reduction. The model reduction approach is described in Section 3. Simulation results for the reference model and the reduced model follow in Section 4.

2. Reference model

The studied crystallizer has the shape of a cylinder whose diameter narrows towards the crystallizer's bottom. Liquid solvent without crystals enters the crystallizer at the bottom. A mixture of solvent and crystals leaves the crystallizer at the top. A side stream for product removal is not considered at the moment, as the focus of this work is on understanding the interaction between fluid flow, crystal growth, and the attrition of crystals in the ultrasonic attenuator. In the crystallizer, small crystals move upwards with the fluid flow, while larger crystals sink to the bottom due to gravity. An additional fluid flow near the crystallizer's bottom transports crystals to the ultrasonic attenuator where they are broken into smaller fragments. The fragments are sent back to the crystallizer.

The reference model for this process is a variant of the models described in [15,16]. The model follows the widely used population balance approach, i.e. not single particles are modeled, but the properties of a particle population consisting of an infinite number of individuals [6]. The main model assumptions are:

- spatial gradients perpendicular to the coordinate x in the direction of the fluid flow are negligible.
- The fluid flow is a plug flow with flow velocity v_f.
- Crystals grow with a constant growth rate G.
- The number of crystals is sufficiently high so that the particle phase may be described by a particle population with a number size density $n(x, L, t)$ denoting the number of particles with size L per volume at a point x in space.
- Close to a point x_{US}, particles are withdrawn from the crystallizer, are sent to the ultrasonic attenuator, are broken into smaller fragments, and are finally recycled into the crystallizer.
- The ultrasonic attenuator has a negligible hold-up. It generates a particle population with a fixed size distribution proportional to $n_{US}(L)$.
- The gravity force, the buoyancy force, and the drag force acting on a particle are in equilibrium.

The population balance equation of the system reads:

$$A(x)\left.\frac{\partial n}{\partial t}\right|_{x,L,t} = -\frac{\partial}{\partial x}\left(A(x)\; v_P(x,L,t)\; n(x,L,t)\right) + D\frac{\partial}{\partial x}\left(A(x)\left.\frac{\partial n}{\partial x}\right|_{x,L,t}\right) - A(x)\; G\left.\frac{\partial n}{\partial L}\right|_{x,L,t}$$
$$+ \dot{V}_{US}\left(n_{US}(L)\frac{\int_0^\infty n(x,l,t)\; l^3\; dl}{\int_0^\infty n_{US}(l)\; l^3\; dl} - n(x,L,t)\right)\hat{\delta}(x - x_{US}) \quad (0 < x < H,\; 0 < L,\; t > 0) \tag{1}$$