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Numerical analysis of crystallization of high aspect ratio crystals with breakage

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ABSTRACT

Two-dimensional population balance model for continuous cooling crystallization of high aspect ratio crystals, including nucleation, size-dependent growth of the two characteristic crystal facets and binary breakage along the crystal length is developed and analysed. The population balance equation is solved numerically using the 2D quadrature method of moments. The behaviour of the crystallizer and the effects of kinetic and process parameters on the characteristics of the crystal size distribution are analysed by detailed numerical experimentation. The binary breakage is described by four breakage rate functions comparing their effects on the mean length and mean aspect ratio of crystals. The simulation results revealed several interesting phenomena caused by interactions of the nonlinear sub-processes playing a relevant role in formulation of high aspect ratio crystals. Interactions of nucleation and size-dependent breakage may induce decrease in production of crystals. Size-dependent crystal breakage, decreasing the mean length of crystals induces some increase in their mean width.

1. Introduction

Several properties of crystalline products, such as bioactivity, hydrophobicity or dissolution rate, depend strongly on the crystal shape. Also, the shape of crystals influences the effectiveness of downstream processing significantly thus it seems to be important to track the shape evolution of crystals in modelling crystallization processes. Crystals are, as a rule, anisotropic and their shape usually is not spherical thus it cannot be described by a single size dimension using some volume equivalent statistics-based linear size but all internal variables needed to characterize the actual shape must be taken into consideration. As a consequence, it leads to the need of modelling crystallization process by multi-dimensional population balance models and by estimation of the growth rates of the individual faces as a function of the operation conditions.

In crystallization from solution, especially in the pharmaceutical and fine chemicals industries, rode-like or needle-shape crystals, i.e. crystals of high aspect ratio are often encountered.

These crystals are characterized by two size parameters, i.e. by length and width thus two-dimensional population balance equation is required to describe the population of crystals. Such 2D population balance models were applied by Ma et al. [1] for simulation of crystallization of KDP crystals and by Puel et al. [2,3] for batch crystallization of a rod-like organic product. Gerstlauer et al. [4] presented a model with detailed kinetic expressions using two independent particle properties. Briesen [5] developed a modified moment method for reducing a 2D population balance model of crystallization to system of moment equations. Borchert et al. [6] presented a general multidimensional population balance approach accounting for dependence of the relative face-specific growth rates on supersaturation. Eggers et al. [7] formulated finding the 2D size distribution as an optimization problem.

Crystals of high aspect ratio are sensitive to breakage along their length thus breakage appears to be an important factor of forming their size distribution [8]. Several contributions have dealt with the simultaneous breakage process of high aspect ratio crystals. Biscans [8] studied the breakage of mono sodium glutamate crystals Bao et al. [9] presented a model of L-threonine crystals describing their growth and binary breakage. Population balance models were applied by Sato et al. [10] and Grof et al. [11] to characterize the breakage phenomenon of high aspect ratio crystals. Borsos and Lakatos [12,13] applied a standard moment method with nucleation, growth and binary breakage of crystals. However, detailed population balance model for studying several aspects of crystallization of high aspect ratio crystals simultaneously, including primary and secondary nucleation, nonlinear size-dependent growth and nonlinear size-dependent breakage as well as their interactions have not been published yet.

Different methods were proposed for solving bivariate population balance equations. High resolution algorithms were introduced by Ma et al. [1] for nucleation and growth problems, Puel et al. [2,3] and Ma et al. [14] applied the method of classes, Alexopoulos and Kiparissides [15] applied an improved sectional grid technique, Chakraborty and Kumar [16] used discretization, Kumar et al. [17] used an extension of the cell average technique while Singh et al. [18] applied for that

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purpose unstructured triangular grids. The Monte Carlo method has proved to be an effective solution method for bivariate cases [19–22] but this method can be applied even for solution of higher dimensional population balance models [23,24].

The method of moments seems to be an attractive alternative [25] although the standard moment method may be applied only for special forms of constitutive expressions of size-dependent growth and breakage processes [12,13]. However, applying some closure model, such as the cumulant-neglect closure [26] or the quadrature method of moments, first proposed by McGraw [27] for 1D population balance equation and extended to bivariate cases by Wright et al. [28], makes the moment method very useful also for solving general nonlinear problems. We apply here the 2D quadrature method of moments which has proved successful in solving several bivariate problems [29–37].

The aim of the present work is to develop and analyse a detailed 2D population balance model for continuous cooling crystallization of high aspect ratio crystals including primary and secondary nucleation, nonlinear size-dependent growth of facets and size-dependent breakage along the crystal length. Using the 2D population balance equation, a nonlinear and unclosed moment equations system is determined for the bivariate mixed moments of length and width variables of crystals which are closed and solved by using the quadrature method of moments. Extended numerical experiments are carried out to investigate the behaviour of the system and the effects of kinetic and system parameters on the properties of crystals and crystals population. Several phenomena caused by interactions of the nonlinear sub-processes playing a relevant role in formulation of high aspect ratio crystals are presented and analysed.

2. 2D population balance model

High aspect ratio crystals, often called rod-like or needle-shape crystals can be characterized by two size dimensions L_1 and L_2 with condition $L_1 >> L_2$, as it is shown for an idealized model form of crystals in Fig. 1, which give sufficient information to compute the volume of crystals needed to close the macroscopic mass and energy balances of crystallizers. A volumetric form factor k_V can be introduced simply for characterizing different forms but it seems to be useful also in taking into consideration breakage of crystals along the length L_1. Naturally, in the case of the parallelepiped in Fig. 1 $k_V = 1$.

The population of crystals is described by the 2D population density function $(L_1, L_2, t) \rightarrow n(L_1, L_2, t)$ by means of which $n(L_1, L_2, t)dL_1dL_2$ provides the number of crystals from the size domain $(L_1, L_1 + dL_1)x(L_2, L_2 + dL_2)$ in a unit volume of suspension at time t. As a consequence, the volume of a crystal takes the form

$$v_c = k_V L_1 L_2^2 \tag{1}$$

while the macroscopic volume of crystals population in a unit volume of suspension takes the form

$$\mathcal{V}_c = k_V \mu_{1,2} = k_V \int_0^\infty \int_0^\infty L_1 L_2^2 n(L_1, L_2, t) dL_1 dL_2 \tag{2}$$

by means of which the volumetric ratio of solution ε is expressed as

$$\varepsilon(t) = 1 - \mathcal{V}_c(t). \tag{3}$$

In Eq. (2), $\mu_{1,2}$ denotes one of the mixed moments of the length and width of crystals

$$\mu_{l,m}(t) = \int_0^\infty \int_0^\infty L_1^l L_2^m n(L_1, L_2, t) dL_1 dL_2, \quad l, m = 0, 1, 2... \tag{4}$$

Assuming that the continuous crystallizer to be modelled is perfectly mixed at both the macro-scale and micro-scale, is unseeded, the agglomeration is negligible, and breakage of crystals may occur along the length L_1 with negligible effects on the width L_2 then the 2D population balance equation takes the form

$$\begin{aligned}
&\frac{\partial n(L_1, L_2, t)}{\partial t} + \frac{\partial [G_1(t, L_1) n(L_1, L_2, t)]}{\partial L_1} + \frac{\partial [G_2(t, L_2) n(L_1, L_2, t)]}{\partial L_2} = \\
&- \frac{n(L_1, L_2, t)}{\tau} + [B_p(t) + B_b(t)] \delta(L_1 - L_n, L_2 - L_n) \\
&- k_{br} \int_0^{L_1} \int_0^{L_2} S_{br}(L_1, L_2) b_{br}^1(\lambda_1, L_1) \delta(\lambda_2 - L_2) n(L_1, L_2, t) d\lambda_1 d\lambda_2 \\
&+ k_{br} \int_{L_2}^\infty \int_{L_1}^\infty S_{br}(\lambda_1, \lambda_2) b_{br}^1(L_1, \lambda_1) \delta(L_2 - \lambda_2) n(\lambda_1, \lambda_2, t) d\lambda_1 d\lambda_2
\end{aligned} \tag{5}$$

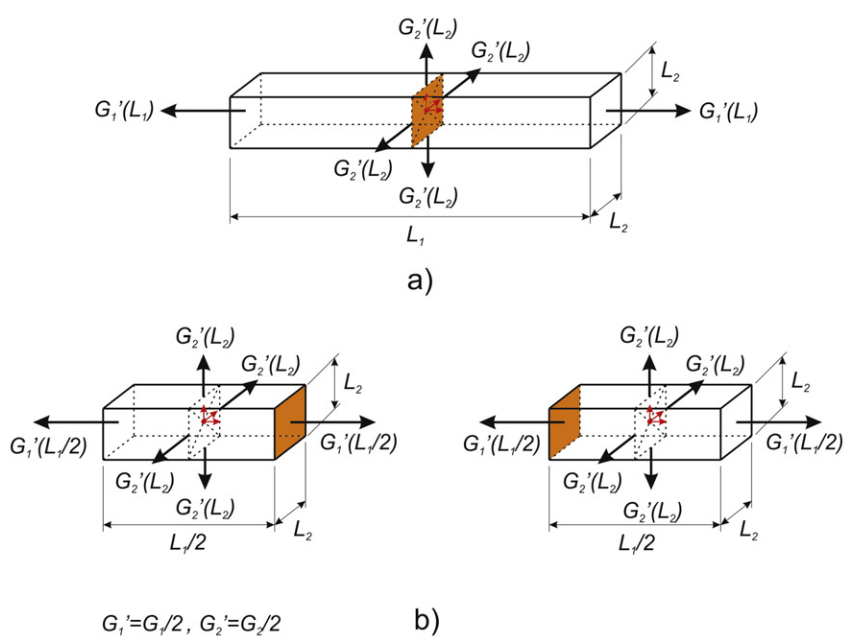

Fig. 1. a) Idealized model form of a high aspect ratio crystal with growth rate G_1 along the length and G_2 along the width, and b) after a symmetrical binary breakage.