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Numerical solution of a multi-dimensional batch crystallization model with fines dissolution

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ABSTRACT

In this article a mathematical model for two-dimensional batch crystallization process with fines dissolution is presented. The fines dissolution is useful for improving the quality of a product and facilitates the downstream process like filtration. The crystals growth rates can be size-dependent and a time-delay in the recycle pipe is incorporated in the model. The high resolution finite volume schemes, originally derived for general systems in divergence form, are used to solve the resulting model. The schemes have already been used for the simulation of complex problems in gas dynamics and were found to be computationally efficient, accurate, and robust. The numerical test problems of this manuscript verify the capability of the proposed schemes for solving batch crystallization models.

1. Introduction

Crystallization from the supersaturated solution is a core technology in the chemical and pharmaceutical industry for the formation of solids and high-purity separations. Supersaturation of the solution is a main driving force in the crystallization process. Several interacting physical phenomena are involved in this process, such as nucleation, growth, mass transfer, attrition, breakage and agglomeration. An understanding and optimization of crystallization processes are desirable for improving the product quality and for the minimization of production costs. The quality of a product is determined by its crystal size distribution (CSD). Modeling and simulation of underlying process enables one to achieve the desired goals and to investigate the effect of different operating conditions.

In batch crystallization process, dissolution of small crystals (fines dissolution) can be used for improving the quality of a product. Normally, large size crystals are needed in the chemical industry. The fine dissolution reduces undesirable small crystals and helps in achieving the desired CSD. Moreover, it facilitates downstream processes like filtration.

The population balance based modeling is a well established and commonly used modeling approach for describing several chemical engineering processes since mid-1960s (e.g. Hulburt and Katz, 1964; Randolph and Larson, 1988). This is a partial differential equation, sometimes with additional integral parts representing breakage, attrition, and aggregation phenomena. The population balance equation (PBE) is coupled to one or more ordinary differential equations (ODEs) describing the mass balance of a solute in the liquid phase and, if necessary, an energy balance of the system.

Analytical solutions of PBEs are only possible in simple cases. Therefore, in practical situations numerical methods are the only tools to solve the given PBE, see Ramkrishna (2000) for a review. Besides Hulburt and Katz (1964) and Randolph and Larson (1988), further literature on population balance modeling can be found in Hounslow, Ryall, and Marshall (1988), Hounslow and Wynn (1993), Kumar and Ramkrishna (1996), Lim et al. (2002), Ma, Tafti, and Braatz (2002), Marchisio, Vigil, and Fox (2003), McCoy (2002), Qamar, Elsner, Angelov, Warnecke, and Seidel-Morgenstern (2006), Ramkrishna (2000) and Rawlings, Witkowski, and Eaton (1992), and references therein.

In this article, a mathematical model is derived for the simulation of two-dimensional batch crystallization process with fines dissolution. A simplified dynamic model of ideally mixed batch crystallizer is considered. It is assumed that fines completely dissolve at the end of recycle pipe attached to the crystallizer, see Fig. 1. The crystal growth rates can be size-dependent and a time-delay in the recycle pipe is included in the model. In this model the growth and nucleation play a dominant role while aggregation

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Nomenclature

b	nucleation order (–)
B_0	nucleation rate at (x_0, y_0) (min^{-1})
$\mathcal{F}$	x-component flux ($G_1 n$) ($\mathrm{m}^{-1}\,\mathrm{min}^{-1}$)
g_1, g_2	growth orders (–)
G_1	growth rate along x-direction ($\mathrm{m\,min}^{-1}$)
G_2	growth rate along y-direction ($\mathrm{m\,min}^{-1}$)
$\mathcal{GG}$	y-component flux ($G_2 n$) ($\mathrm{m}^{-1}\,\mathrm{min}^{-1}$)
h	death function (–)
k_b	nucleation rate constant ($\mathrm{m}^{-3}\,\mathrm{min}^{-1}$)
k_{g_1}, k_{g_2}	growth rate constants ($\mathrm{m\,min}^{-1}$)
m	solute mass (kg)
m_{solv}	mass of the solvent (kg)
$\dot{m}$	mass flow rate ($\mathrm{kg\,min}^{-1}$)
n	crystal size distribution (CSD) (m^{-2})
N_x	number of grid cells in x-direction (–)
N_y	number of grid cells in y-direction (–)
$\mathbb{R}_+$	half space $(0, \infty)$
S	supersaturation of the solute (–)
t	time (min)
t_{p}	time-delay in the pipe (min)
T	temperature (°C)
V_{c}	volume of the single crystal (m^3)
V_{crz}	volume of the crystallizer (m^3)
V_{p}	volume of the pipe (m^3)
$\dot{V}$	volumetric flow rate ($\mathrm{m}^3\,\mathrm{min}^{-1}$)
w	mass fraction (–)
w_{sat}	saturated mass fraction (–)
x	width of the crystal (m)
y	length of the crystal (m)
$x_0, x_{\max}$	minimum and maximum crystal widths (m)
$y_0, y_{\max}$	minimum and maximum crystal lengths (m)

Greek symbols

$\alpha_i\,(i = 1, 2, 3, 4)$	constants for size-dependency of growth (–)
δ	Dirac delta distribution
Δ	central difference
$\mu_{i,j}$	ij th moment of CSD
ϕ	limiting function in $HR - \kappa = 1/3$ scheme
φ	limiting function in $HR - \kappa = -1$ scheme
ν^x, ν^y	flux difference ratios in $HR - \kappa = 1/3$ scheme
ρ_c	crystal density ($\mathrm{kg\,m}^{-3}$)
ξ, η	flux difference ratios in $HR - \kappa = -1$ scheme

Subscripts

in	inflow stream to the crystallizer
out	outlet stream from the crystallizer

Abbreviations

CSD	crystal size distribution
ODE	ordinary differential equation
PBE	population balance equation

and breakage phenomena are neglected. The proposed model is indeed simple considering the large number of phenomena occurring during real crystallization processes. The coalescence and aggregation phenomena play a major role in multiphase population balance problems of liquid–liquid systems (e.g. emulsions) or during nanoparticles formation in precipitation processes. However, in relatively slow crystallization processes (solid liquid systems) nucleation and growth are the most essential phenomena and coalescence (aggregation) can be often neglected. The high resolution scheme of Koren (1993) and the one given by LeVeque (2002) are implemented for solving the resulting model. These schemes were originally derived for general systems in divergence form. The schemes have already been used for solving complex problems in gas dynamics and were found to be computationally efficient, accurate, and robust. The batch crystallization model in this manuscript also form a hyperbolic equation (divergence form), therefore the application of the proposed schemes for solving this model is very natural. In this article, the accuracy of the schemes is analyzed with the help of two test problems for which analytical solutions are available.

This paper is organized as follows. In Section 2, a two-dimensional batch crystallization model with fines dissolution is presented. In Section 3, the proposed numerical schemes are derived. In Section 4, numerical test problems are presented. Finally, Section 5 gives conclusions and remarks.

2. Two-dimensional batch crystallization model

In this section, a mathematical model is derived for the simulation of a batch crystallizer equipped with a fines dissolution loop, see Fig. 1. A simplified dynamic model of an ideally mixed batch crystallizer is considered. In the current simple situation, a recycle pipe is attached to the crystallizer and it is assumed that fines completely dissolve at the end of the pipe. It is also assumed that initially the pipe is filled with the same solution as in the crystallizer. Moreover, a delay in the recycle pipe is also incorporated in the model. In the two-dimensional case, the evolution of CSD $n(t, x, y) \geq 0$ is given by the population balance equation (PBE) (e.g. Randolph & Larson, 1988; Rawlings, Miller, & Witkowski, 1993)

$$\frac{\partial n(t,x,y)}{\partial t} + \frac{\partial [G_1(t,x)n(t,x,y)]}{\partial x} + \frac{\partial [G_2(t,y)n(t,x,y)]}{\partial y} = -\frac{\dot{V}}{V_{\mathrm{crz}}} h(x,y)n(t,x,y) + B_0(t)\delta(x - x_0, y - y_0), \tag{1}$$

$$n(0,x,y) = n_0(x,y), \quad (t,x,y) \in \mathbb{R}^3_+, \tag{2}$$

where $\mathbb{R}_+ := (0, \infty)$. Here, $n_0(x,y) \in \mathbb{R}^2_{\geq 0}$ denotes the CSD of seed crystals added at the beginning of the batch process, $G_1(t,x) \geq 0$ and $G_2(t,y) \geq 0$ are the crystals growth rates along the characteristic directions x and y, $B_0(t) \geq 0$ is the nucleation rate at minimum crystal size (x_0, y_0), and δ is the Dirac delta distribution. Moreover, V_{crz} is volume of the crystallizer and $\dot{V}$ is volumetric flow rate. The death function $h(x,y)$ describes the dissolution of small particles below some critical size. The ij th moment of CSD is defined as

$$\mu_{i,j}(t) = \int_0^\infty \int_0^\infty x^i y^j n(t,x,y)\,dx dy, \quad i,j = 0, 1, 2, \ldots. \tag{3}$$

We consider the case of fines dissolution with time-delay in the dissolution pipe. In other words, small nuclei (fines) are taken from the crystallizer along with the solution and are dissolved in the dissolution unite. The solution from the recycle pipe comes back into the crystallizer after certain time. For dissolving nuclei in the dissolution unit (pipe) a heat exchanger is equipped with the recycle unit. However, the solution is cooled down back to the crystallizer temperature before putting back into the crystallizer. This avoids a flow of hot liquid into the crystallizer which can effect the crystallization process. Due to above assumptions, a balance for the liquid phase yields an ordinary differential equation (ODE) for the solute mass