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Rapid Communication

Two dimensional population balance modelling of crystal growth behaviour under the influence of impurities

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ABSTRACT

This short communication investigates the impact of impurity on crystal growth behaviour in terms of crystal shape distribution. A two dimensional population balance model is applied to study the crystal shape changing behaviour in cooling crystallisation producing β-form L-glutamic acid crystals, in the presence and without the presence of an additive, L-phenylalanine. The simulation results were verified in experiments using a 1-litre crystalliser for crystallisation of L-glutamic acid.

1. Introduction

For crystalline particulate products obtained through crystallisation, crystal morphology is an important property that affects the processability of the solid crystals and the end-use properties of the final products. Although much work has been done in order to understand the growth behaviour of single crystals, for quite some time, there has been a lack of knowledge on how to describe, model and ultimately control the morphology for a population of crystals growing in a crystalliser. As a result, at process level, modelling, simulation and control has focused on crystal size distribution where the size of a crystal is ofen defined as the diameter of a sphere having the same volume of the crystal [1–7]. In recent years, there have been noticeable developments in modelling the dynamic evolution of crystal shape distribution using multi-dimensional and morphological population balance (PB) equations [8–15], as well as in on-line characterisation of crystal shape using imaging and image analysis [16–30]. These new developments paved the way for a practical solution to automatic control of crystal shape distribution. For example, Wan et al. [30] recently demonstrated that using a morphological PB model, an optimal supersaturation trajectory can be derived, and desired crystal shape distribution can be obtained by tracking the optimal supersaturation trajectory.

In addition to supersaturation, another effective means to influence crystal morphology is the use of impurities, or additives [31,32]. An impurity can have strong influence on crystal growth, production throughput, yield and robustness of formulations. Quantitative knowledge on their influence can be used not only to develop strategies for their removal or moderating their effects, but also to possibly make positive use of an additive as a manipulated variable for optimisation and control of crystal growth processes [33]. The effect of impurity is known to be highly selective: on the one hand, some impurity may inhibit the nucleation or growth of the crystals of one compound while promoting the nucleation or growth of the crystals of the other compound; on the other hand, the influence of the impurities on a single crystal may only demonstrate on certain crystallographic faces. Furthermore, the influence of impurities on crystallisation is also closely correlated with process environments such as supersaturation, cooling rate and pH [34]. The mechanism and kinetics for the effect of the impurities on crystallisation were studied intensively in the literature. Weissbuch et al. [35] investigated the effect of impurities on nucleation, growth and dissolution of crystals at the molecular level, and developed the tailor-made additives for blocking, docking and disrupting molecular packing arrangements during crystallisation. Molecular modelling was thereafter widely applied to explain the mechanism for the effect of the impurities on crystallisation of various compounds [36–39]. Davey et al. [40]

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proposed guidelines for selecting suitable additive molecules for preventing the appearance of undesirable polymorphs by conformational mimicry. It was concluded that any successful additive molecule should have the appropriate conformation for taking part in the bonding network with minimum disruption. Along with the success of using molecular modelling to explain the mechanism for the effect of impurities on crystallisation, kinetics relating the concentration of the added impurity as well as the supersaturation and faceted growth rates of crystals was also studied in literature. Kubota and Mullin proposed a kinetic model, the Kubota–Mullin model, to describe the faceted crystal growth rate as a function of impurity concentration [41]. Kubota–Mullin model or similar kinetic models were further applied to quantify the macro-scale effect of the added impurities on faceted growths of various crystals such as sodium chloride, hydroquinone, sucrose and L-glutamic acid [42–45].

An earlier experimental study on the crystallisation of benzamide in the presence of impurities demonstrated that controlled modification of crystal habit was feasible by adding tailor-made impurities [46]. The influence of added impurities was mainly studied for single crystals to quantify the kinetic effect individually rather than for the population of crystals in the crystalliser. Patience and Rawlings designed a closed-loop feedback control system for shape control in which the shape was monitored by an imaging system, and the control was achieved by manipulating the flow rate of a habit modifier stream [19].

The purpose of the current work is to use a two-dimensional population balance model to study the impact of impurity on crystal growth behaviour, in particular the crystal shape distribution. The work is different from previous work in that early work either was restricted to a single crystal or a few crystals, or treated each crystal as a sphere for a population of crystals. In this paper, a two dimensional population balance equations were built to model the effect of L-phenylalanine, as the impurity, on the shape changing behaviour of a growing crystal population during seeded crystallisation of β-form L-glutamic acid (L-GA). Faceted growth kinetics of β-form L-GA crystals with and without the added impurity L-phenylalanine is discussed in Section 2. The two dimensional PB model of seeded crystallisation with the consideration of the added impurity L-phenylalanine is illustrated in Section 3. The experimental apparatus for validating simulation results is introduced in Section 4. The simulation results as well as its experimental validation are discussed in Section 5, which is followed by final remarks in Section 6.

2. Faceted growth kinetics of β-form L-GA crystals with and without the presence of impurity L-phenylalanine

The growth kinetics of both α-form and β-form L-GA crystals was investigated in the literature [45,47]. Various methods were used to measure faceted growth kinetics relating to the operating conditions such as the relative supersaturation. For direct measurement, a single crystal can be mounted in the growth cell for online image-based measurement using a video system [45]. An indirect approach was to estimate crystal growth rates using a predetermined growth pattern, where the initial and final crystal size distributions were measured offline [48]. The faceted growth rates of crystal faces during crystallisation can also be measured in a statistical way using real-time in-process imaging techniques, where multiple crystals were snapshot from the reactor and a multi-scale segmentation algorithm was used to effectively extract objects from their backgrounds [18,22]. For example, the shape of a β-form L-GA crystal is shown in Fig. 1(a), which can be further simplified as a rod-like parallelepiped with the length L and the width W depicted in Fig. 1(b). The obtained faceted growth rates of β-form L-GA crystals in the length and the width directions as a function of the relative supersaturation σ were as follows [11]:

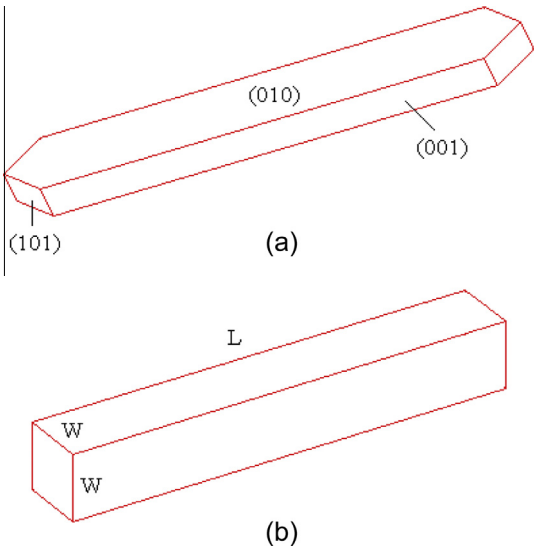

Fig. 1. A β-form L-glutamic acid crystal (a), can be simplified as a rod-like structure (b).

$$G_L = 3.44 \times 10^{-7} e^{(\sigma-0.49)/0.02} / \left[1 + e^{(\sigma-0.49)/0.02}\right]^2 \quad (1)$$

$$G_W = (-0.51 + 2.15\sigma - 2.22\sigma^2) \times 10^{-6} \quad (2)$$

where the corresponding experiment was performed around a relative supersaturation of 0.5 in the solution and G_L, G_W are the growth rates in the length and the width directions respectively. It is worth noting that the obtained growth kinetics fits better with the corresponding experimental data than those using traditional Kubota-Mullin models for this case.

Considering the effect of the added impurity L-phenylalanine on the growth of β-form L-GA crystal, the growth kinetics was also measured using the single-crystal method [45]. It was observed that the added impurity only inhibited the growth in the length direction and the degree of the inhibition was closely related to the concentration of the added impurity as well as the solution supersaturation. Then the modified overall growth rate in the length direction can be described as follows:

$$G_L^* = G_L \left[1 - \ell \left(K C_p^n\right)^{\frac{1}{2}}\right], \quad (3)$$

where ℓ, K, n are parameters related to the corresponding molecular adsorption mechanism, C_p denotes the concentration of L-phenylalanine, and G_L is defined by Eq. (1) [45]. It can be seen that the modified growth rate G_L^* depends on both the relative supersaturation of the solution and the concentration of the added impurity. It needs to point out here that while studying the facet growth kinetics by observing a few single crystals, as Kitamura and Ishizu did [45,47], is fundamental, just like measuring hear transfer or mass transfer coefficients, it cannot reflect the growth behaviour for a population of crystals. To simulate the growth behaviour of a population of crystals, the measured facet growth kinetics for single crystals needs to be incorporated into a morphological population balance model. Therefore the work presented in this paper represents a step forward from the single crystal work of Kitamura and Ishizu [45] and is the main thrust of this contribution.

3. Population balance model

A two dimensional PB model for seeded cooling crystallisation that generates β-form L-glutamic acid crystals was built previously (Ma et al. [11]). It was a simplified PB model that used two