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Numerical simulations of flow and mass transfer during large-scale potassium dihydrogen phosphate crystal growth via three-dimensional motion growth method

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ABSTRACT

A novel solution-based crystal growth method, namely, the three-dimensional motion growth method (3D MGM) had been suggested by our team to effectively utilize convection for simultaneous enhancement of morphological stability and mass transfer. To evaluate this new method and link it to practice, numerical simulations of flow and mass transfer during large-scale potassium dihydrogen phosphate (KDP) crystal growth via this new method are carried out. The supersaturation field on the crystal surface is presented as functions of translational distance and crystal size. The relationships between directions of solution flow adjacent to crystal surface and surface shear stress are summarized and discussed. Results indicate that 3D MGM benefits the large-scale crystal growth by improving the morphological stability and crystal quality.

1. Introduction

It is well known that convection has significant effects on morphological stability and mass transfer in the process of crystal growth from solution. Convection can enhance growth rate, however, it may also induce morphological instability of the crystal surface and resulting in step bunching and inclusion formation.

A number of experimental [1–5] and theoretical [6–9] studies were conducted to reveal the correlations of convection with morphological instability and inclusion formation. All these studies suggested that the solution flow direction adjacent to the solution/crystal interface determines the occurrence of morphological instability to a large extent. If the flow direction is opposite to the step motion, the morphological stability can be enhanced; by contrast, if the solution flows along the direction of step advancement, the morphological instability easily occurs. Potapenko [10] notably concluded that vicinal crystal faces were morphologically stable against any perturbation if the solution flow was exactly reversible. However, the reversible stirring in the conventional rotating-crystal method would not result in a reversible shear rate over the entire crystal surface. Thus, large portions of the crystal surface remain morphologically unstable, and the possibility of inclusion formation still exists.

Surface supersaturation distribution depending on convection directly involves in the occurrence of morphological instability and inclusion formation. Robey et al. [11,12] reported that the inhomogeneous supersaturation field on a crystal surface led to step "bending" and the formation of "valleys", which causes the morphological instability and results in inclusion formation. The degree and location of bending depended on several parameters, such as the growth and rotation rates of crystals. Vartak [13,14] posited that the specific portions of two faces of potassium titanyl phosphate crystals maintained low supersaturation during the entire growth progress when the direction of crystal optical axis is perpendicular to the direction of gravity. The simulated pattern of the low-supersaturation regions are very similar to those inclusion regions observed during actual experimentation; if the direction of crystal optical axis is parallel to the direction of gravity, the low-supersaturation region in the simulations disappears, and no inclusion is observed in the experiment [13,14].

Based on above facts, our team proposed a novel method named three-dimensional motion growth method (3D MGM), aiming at more sufficient use of convection, thus achieving higher growth

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velocity and better crystal quality. Numerical analysis of small-scale (2 cm) crystal growth via this method by Zhou et al. [15] showed that 3D MGM has advantages in surface supersaturation and surface solute distribution. The corresponding experiment by Yin et al. [16] also confirmed that crystal through this method claimed higher quality than that through traditional rotating-crystal method. But given the very high-energy Nd-glass lasers used for inertial confinement fusion research need large (about 40 cm) KDP plates for electro-optic switches and frequency converters, the flow state of crystal growth of this size will be turbulent flow and the surface supersaturation field will also be notably different with that of small-scale crystal growth. Therefore, it is necessary to conduct numerical analysis of flow and mass transfer for KDP crystals growth via 3D MGM under conditions of large-scale and turbulent flow.

2. Physical and mathematical models

2.1. Physical model

As shown in Fig. 1a, a KDP crystal with ideal morphology is placed into a rectangular crystallizer measuring $150 \times 150 \times 150\ \mathrm{cm}^3$, and it periodically translates along the path of $1 \rightarrow 2 \rightarrow 3 \rightarrow 4 \rightarrow 5 \rightarrow 6 \rightarrow 1$. During translations, the translational directions are parallel to *X*-, *Y*-, or *Z*-axis. On each direction, the crystal is accelerated firstly from rest state to the maximal velocity; it is then kept translation with the maximal velocity for a short time before decelerating to rest. The time scales of acceleration and deceleration are both set 0.2 s for all cases of simulations. Note that the bulk supersaturation is 8% and the maximal velocity is 0.4 m/s if without special illustration.

2.2. Mathematical model

Continuity and Navier-Stokes equations are employed for conservation of mass and momentum in the present solution system. The species transport equation is used to model the evolution of concentration field. All these equations neglect the change of crystal size since the tiny time scale of simulations. The corresponding equations are expressed as follows:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \boldsymbol{U}) = 0 \tag{1}$$

$$\frac{\partial (\rho \boldsymbol{U})}{\partial t} + \rho \boldsymbol{U} \cdot \nabla \boldsymbol{U} = -\nabla P + \nabla \cdot \boldsymbol{\tau} - \rho g \boldsymbol{e}_y \tag{2}$$

$$\frac{\partial (\rho c)}{\partial t} + \nabla \cdot (\rho \boldsymbol{U} c) = \nabla \cdot \left[\left(\rho D + \frac{\mu_t}{Sc_t} \right) \nabla c \right] \tag{3}$$

where $\boldsymbol{U}$ is the velocity vector, p is the pressure, $\boldsymbol{e}_y$ is the unit vector of the *Y* direction producing the buoyancy convection, c is the KDP mass fraction, D ($D = 1 \times 10^{-9}\ \mathrm{m}^2/\mathrm{s}$) is the solute diffusion coefficient, and ρ is the solution density, which is a function of the mass fraction of KDP [17]. μ_t is the turbulent viscosity, which is derived from the Spalart–Allmaras one-equation turbulence model [18], $Sc_t (Sc_t = 0.7)$ is the turbulent Schmidt number. Due to the high Reynolds number, the solution flow is turbulent. For the description of turbulent flow, the viscous stress tensor $\boldsymbol{\tau}$ is written as

$$\tau_{ij} = \mu_{eff} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) - \frac{2}{3} \mu_{eff} (div U) \delta_{ij} \tag{4}$$

where δ_{ij} is the Kronecker delta, and μ_{eff} is the effective turbulent viscosity, which is calculated by

$$\mu_{eff} = \mu + \mu_t \tag{5}$$

where μ is the dynamic viscosity (0.000564 kg/m·s).

The no-slip boundary condition ($\boldsymbol{U} = 0$) is used on all vessel walls and crystal surfaces. The top surface of the solution is a free surface with zero shear stress. The velocity magnitude on the crystal surface is a given value, and its direction is determined by the translational schedule shown in Fig. 1a. The mass transfer rate on the crystal surface m is defined by

$$m = (\rho_s - \rho_0 c_e) R = \rho_0 D \left. \frac{\partial c}{\partial n} \right|_0 . \tag{6}$$

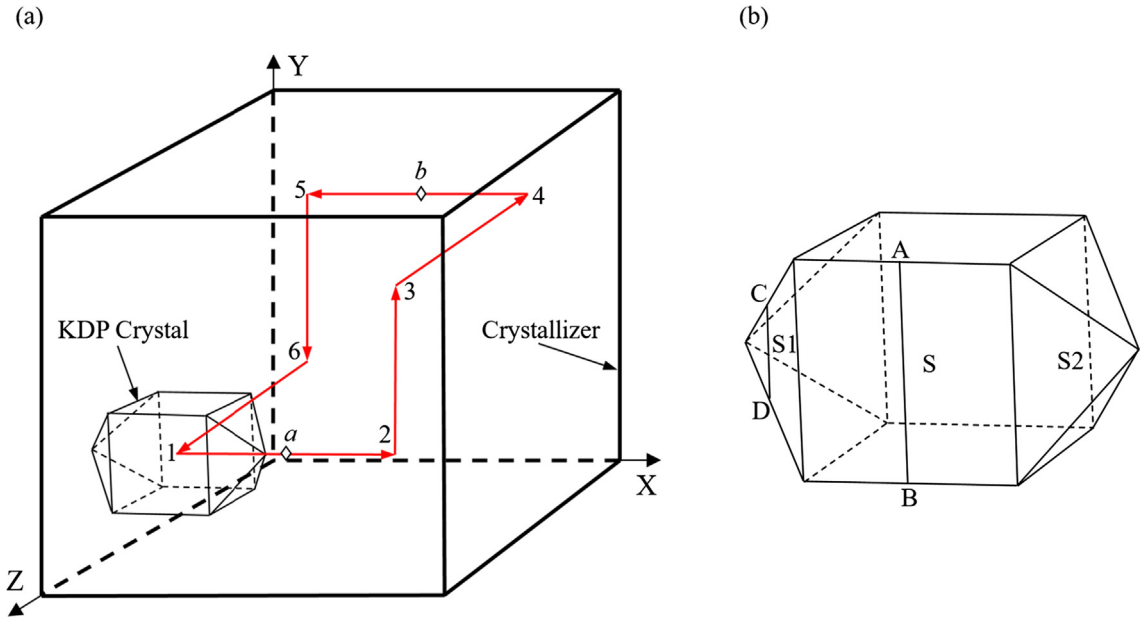

Fig. 1. (a) Schematic of 3D MGM, the red arrows indicate the translation path of crystal center and the positions of *a* and *b* correspond to the midpoint of related moving direction. (b) Related signs on the crystal surface. Segments AB and CD are obtained through the geometrical centers of prismatic face S and left pyramidal face S1 and will be used hereafter to collect data for surface supersaturation analysis. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)