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Time-dependent, three-dimensional flow and mass transport during solution growth of potassium titanyl phosphate

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Abstract

A finite-element, numerical model is used to compute time-dependent, three-dimensional fluid flow, mass transfer, and continuum growth kinetics in the potassium titanyl phosphate (KTP) solution crystal growth system of Bordui et al. The effects of a periodically-reversing crystal rotation schedule are analyzed for two different crystal-mounting geometries. Results suggest a lower probability of the occurrence of defects when the mounting geometry is designed to take advantage of periodic flow reversal effects on the supersaturation field.

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1. Introduction

The growth of large, single crystals from liquid solutions results from a complex interaction of continuum transport phenomena, crystal growth kinetics, and thermodynamics. Although kinetic models have formed the basis of many theoretical analyses of growth stability [1–5], it has long been known that mass transfer limitations have a profound effect on solution crystal growth systems, leading to unacceptably slow growth rates, and in some cases morphological instability. An important aspect of increasing growth rates while maintaining high crystal quality is the use of forced convection [6–10]. Because of the low rate of diffusion inherent in liquids, even modest levels of forced convection greatly influence solute transport from the solution to the crystal interface, thereby affecting growth rate and morphological stability [8]. Forced convection can be created in

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several ways, for example by rotating the growth vessel, as in the accelerated crucible rotation technique [6], or by rotating a support structure holding the crystal, as in the KDP rapid growth system developed at Lawrence Livermore National Laboratory [9,10].

In a previous paper [11], we used continuum transport modeling to study the effects of forced convection on the transport-limited growth of large, single crystals of potassium titanyl phosphate (KTP) in a system developed by Bordui et al. [12,13]. Two variants of this system, referred to here as the 0° and 90° orientations, are illustrated in Fig. 1a and b, respectively. A KTP seed crystal is mounted onto a platinum support and immersed into a high temperature, supersaturated solution of $K_6P_4O_{13}$. The temperature of the solution, kept in a nearly isothermal state by external heaters, is slowly lowered to maintain supersaturation as crystal growth proceeds and solute is depleted. The crystal support rod is rotated to create forced convection in an attempt to increase mass transfer and reduce solute concentration variations at the crystal interface.

Bordui and Motakef [14] studied the rate of growth and quality of crystals grown in this system using crystal support orientations of 0°, 10°, 45°, and 90°. They also performed flow visualization experiments in a model experimental system. Accelerated rotation of the crystal support, with periodic reversal of rotation direction, was used in these studies. Crystals grown in the 0° orientation were plagued by large numbers of liquid inclusions formed in the equatorial region of the crystal. In contrast, the 90° orientation produced superior crystals that were nearly free of inclusions. The 90° orientation also produced the highest growth rates. Bordui and Motakef used results of the flow visualization studies to argue that hydrodynamics alone were responsible for the difference in outcomes in the different geometries. They postulated that localized solute depletion occurred in the fluid wakes that form over certain regions of the crystal and that step trains emanating from regions of high supersaturation slow down and bunch together in these depleted regions, causing inclusions to form.

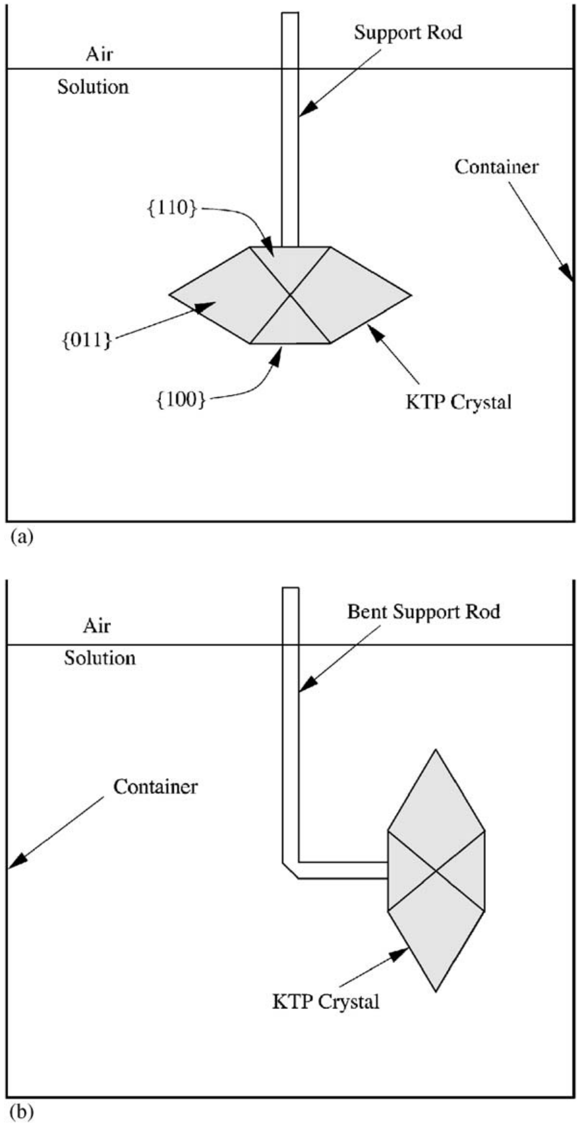

Fig. 1. Schematic of two configurations of the KTP growth system: (a) 0° orientation and (b) 90° orientation.

Validating the hypothesis of Bordui and Motakef requires detailed information on the distribution of supersaturation at the crystal interface. However, harsh operating conditions during high-temperature solution growth place severe restrictions on the range of experimental and visualization techniques that can be used to study flow and mass transport in this system. For the most part, previous studies of 3D transport effects in solution