International Journal of Heat and Mass Transfer 72 (2014) 452–460

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Numerical study of heat transfer during sapphire crystal growth by heat exchanger method

Wencheng Ma [a,b], Lili Zhao [c,*], Guoqiang Ding [a], Yang Yang [a], Tiezheng Lv [d], Ming Wu [b], Lijun Liu [b,*]

[a] Uonone Optoelectronics Technology Co., Ltd., Zhenjiang, Jiangsu 212132, China
[b] Key Laboratory of Thermo-Fluid Science and Engineering, Ministry of Education, School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an, Shaanxi 710049, China
[c] Department of Applied Chemistry, Harbin Institute of Technology, 92 Xidazhi Street, Harbin 150001, China
[d] College of Material Science and Engineering, Hunan University, Changsha, Hunan 410012, China

ARTICLE INFO

Article history:
Received 9 April 2013
Received in revised form 8 October 2013
Accepted 8 January 2014
Available online 3 February 2014

Keywords:
Transient global model
Crystal growth
Heat transfer
Internal radiation
Sapphire crystal

ABSTRACT

We developed a transient global model to study the heat transfer in sapphire crystal growth by heat exchanger method (HEM). Internal radiation in the semi-transparent sapphire was modeled with a rigorous discrete ordinate model and phase change was calculated based on a fixed-grid method in our simulations. The evolutions of thermal field, melt flow and melt-crystal (m-c) interface shape were predicted during the whole crystal growth process. Simulation results show that a U-shape distribution of axial temperature gradient appears along the centerline of the sapphire crystal and melt, respectively, due to the effect of internal radiation. The m-c interface is severely curved near the crucible wall and relatively flat at the central region with an increasing convexity when crystal grows. The sapphire melt flow is in a laminar state dominated by the thermal buoyancy force. The high growth rate at the final stage of the solidification process resulted in bubbles at the top layer of the grown crystal. The formation mechanism of the grain boundaries at the crystal periphery was discussed. The numerical simulation results show a good agreement with the experiment data. The developed global model herein provides important information to improve the growth process of sapphire crystal by the HEM technique.

1. Introduction

Sapphire crystal has been used for growing GaN-based high-brightness Light Emitting Diode (LED) device layers due to its relatively good lattice match, high thermal stability, chemical resistance and mechanical strength [1]. Heat exchanger method (HEM) is one of the most important techniques for producing large-size high-quality sapphire crystals [2] for the lately booming LED market. Precise control of the heat transfer in the whole HEM furnace is essential to grow perfect crystals because the quality of the grown crystal is quite sensitive to its thermal history in the furnace. Comparing to the time-consuming and high-cost experimental method, numerical simulation is definitely an effective tool to accurately predict and control the heat transfer in the HEM furnace to obtain high-quality sapphire crystals.

Different forms of heat transfer, such as thermal conduction, thermal convection, face to face thermal radiation between the solid surfaces, phase change as well as internal radiation in the melt and crystal, coexist in the sapphire crystal growth furnaces. Much work has been performed to investigate these heat transfer phenomena during the crystal growth by Czochralski [3–8] and Kyropoulos [9–12] methods. For the HEM method, Lu and his coworkers established 2D [13] and local 3D [14] quasi-steady-state models to investigate the effect of growth parameters on the convexity of melt-crystal (m-c) interface shape. They also [15] studied the effect of crucible geometry on the thermal field and found that the crucible with a curved base decreased the convexity of the m-c interface. However, the quasi-steady simulations in the previous studies ignored the thermal inertia of the system and the solidification rate during the actual transient crystal growth process. Fortunately, the thermal inertia of the thermal system can be fully taken into account and the solidification rate during the whole solidification process can be accurately calculated in the transient global simulation [16]. In addition, the modeling of internal radiation in the sapphire crystal is a complicated task and was neglected in the previous simulations of HEM method. To have an insight into the thermal history of the crystal growth, it is necessary to develop transient global model of heat transfer with accurate prediction of internal radiation in the HEM furnace.

In this paper, a transient global model is established for sapphire crystal growth process in a HEM furnace. The numerical model takes into account the phase change and internal radiation in the sapphire melt and crystal and is validated by experimental measurements. The temperature and velocity fields of the sapphire

* Corresponding authors. Tel./fax: +86 451 86413707 (L. Zhao). Tel./fax: +86 29 82663443 (L. Liu).
E-mail addresses: lilizhao@hit.edu.cn (L. Zhao), ljliu@mail.xjtu.edu.cn (L. Liu).

0017-9310/$ - see front matter © 2014 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.ijheatmasstransfer.2014.01.033

melt and crystal as well as the m-c interface shape during the growth process are investigated. The formation mechanisms of bubbles and grain boundaries in the grown crystal were discussed.

2. Experimental setup

A schematic diagram of the HEM furnace for growing c-plane sapphire crystal is given in Fig. 1. The HEM furnace mainly consists of tungsten crucible, tungsten resistance heater, heat exchanger shaft, tungsten radiation shields and chamber wall. The polycrystalline sapphire feedstock is loaded into the tungsten crucible ($270\ mm^{I.D.} \times 420\ mm^{H} \times 20\ mm^{T}$) and a c-plane crystal seed is located in the seed pocket at the bottom of the crucible. The height of the grown crystal is 216 mm. The crucible was supported by the heat exchanger shaft filled with circular cooling helium flow. The cylindrical thermal radiation shields are made of a multitude of closely spaced, parallel, highly reflective tungsten slices, surrounding the heater and crucible to prevent heat loss from the furnace. The furnace is well sealed with a water-cooled wall and operates in an argon atmosphere with a very low pressure. The sapphire crystal growth process mainly includes two phases: the seeding phase and the bulk crystal growth phase. At the seeding phase, the c-plane seed crystal should be preserved (not totally melted) by accurately adjusting the heating power and the cooling helium flow rate. Then, with the decrease of the heating power and the increase of the cooling flow rate, the temperature at the melt-seed interface is dropped below the melting point. The seed begins to grow upwards and the bulk crystal growth phase starts. During the bulk crystal growth phase, a suitable temperature gradient is maintained by adjusting the heating power and slowly increasing the cooling helium flow rate. During the crystal growth process, we lower a probe into the melt to measure the height of the solidified crystal every several hours.

3. Numerical modeling

3.1. Computational model of heat transfer

A transient global model was developed by taking into account the thermal conduction, thermal radiation between solid surfaces, melt convection, cooling helium flow as well as phase change and internal radiation in sapphire domain. To compromise the computational accuracy and convenience, the following assumptions were made in the current global model: (1) the furnace configuration is axisymmetric; (2) the sapphire melt flow is incompressible and the Boussinesq assumption is applied; (3) the radiative surfaces of all solids located in non-participating medium are diffuse-gray; (4) the melt free surface is flat and the effect of Marangoni surface tension is ignored. A structured/unstructured combined mesh scheme [17] is applied in our simulations due to the presence of the irregular argon domain. The algorithms for the global modeling of heat transfer in a crystal growth furnace have been described in our previous paper [18]. The cooling helium flow was assumed to be laminar as the Reynolds number at the inlet is about 1400.

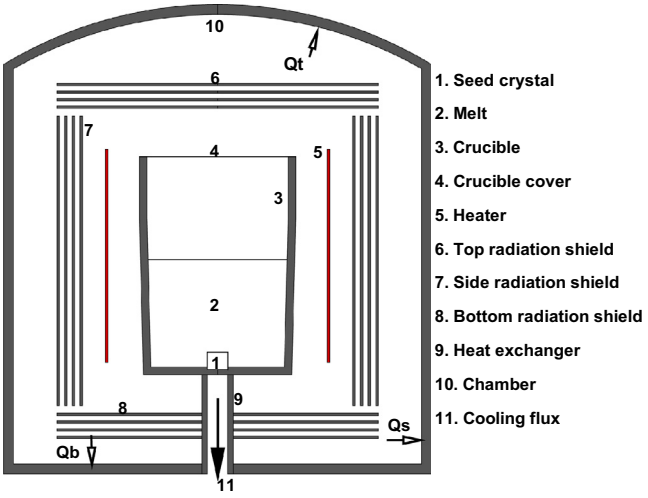

Fig. 1. Schematic diagram of the HEM furnace.

The heat transfer in the resistance heater is governed by

$$\rho C_P \frac{\partial T}{\partial t} = \lambda \nabla \cdot (\nabla T) + \dot{q}, \tag{1}$$

where ρ is the density, C_p is the heat capacity, T is the temperature, λ is the heat conductivity. $\dot{q}$ is the heat source per unit volume in the heater. It is uniformly distributed in the heater and controlled by the temperature measured by the optical pyrometer.

The energy conservation equation governing the thermal fields in other solid conductive elements is written as

$$\rho C_P \frac{\partial T}{\partial t} = \lambda \nabla \cdot (\nabla T). \tag{2}$$

The solid elements include crucible, radiation shields, heat exchanger shaft and chamber wall.

The high conductivity of the tungsten crucible will result in a large radial heat flux into the sapphire from the crucible side wall. On the other hand, the cooling helium flow will extract lots of heat from the central bottom of the crucible. This might result in a highly curved m-c interface shape during the crystal growth in the present HEM furnace. To accurately track such interface evolution, an enthalpy formulation based on fixed-grid methodology [19] was used to model the phase change during the growth process. The governing equations for the sapphire domain can be expressed in the following forms:

$$\nabla \cdot \vec{u} = 0, \tag{3}$$

$$\rho \frac{\partial \vec{u}}{\partial t} + \rho \vec{u} \cdot \nabla \vec{u} = -\nabla p + \nabla \cdot \left[\mu(\nabla \vec{u} + \nabla \vec{u}^T)\right] + \rho \vec{g}\, \beta (h - h_{ref})/C_p + S_m, \tag{4}$$

$$\rho \frac{\partial (h)}{\partial t} + \rho \nabla \cdot (\vec{u} h) = \nabla \cdot \left(\frac{\lambda}{C_p} \nabla h\right) + \nabla \cdot \vec{q}_{rad} + S_h, \tag{5}$$

where $\vec{u}$ is the velocity, p is the pressure, μ is the dynamic viscosity, $\vec{g}$ is the gravity acceleration vector, β is the thermal expansion coefficient, h is the sensible enthalpy and h_{ref} is the reference sensible enthalpy. The source term S_m in Eq. (4) is defined as $S_m = -A\vec{u}$, where A increases from zero to a large value as the solid fraction of a grid cell increases from 0 at full liquid state to 1 at full solid state. In the liquid region (sapphire melt) it takes a zero value and the momentum equation is in terms of the actual fluid velocities. In the solid region (sapphire crystal) the source term dominates all other terms in the momentum equation and the predicted superficial velocity of the cell approaches to zero. In this way, the source term S_m in Eq. (4) is used to drop the velocity to zero when the melt has fully solidified in a grid cell during the phase change. The second term on the right-hand side in Eq. (5) is the internal radiation contribution, which will be discussed in detail below. The source term S_h in Eq. (5) accounts for the rate of volumetric latent heat change during the growth process, which can be described as [19]

$$S_h = -\rho \frac{\partial (\Delta H)}{\partial t}, \tag{6}$$

where ΔH represents the latent heat content. Instead of tracking the m-c interface explicitly, a concept of liquid fraction g_l in each cell