# Interface shape evolution in the growth of BGO by HEM

Jong Hoe Wang and Do Hyun Kim

*Department of Chemical Engineering and Process Analysis Laboratory, Korea Advanced Institute of Science and Technology, 373-1, Kusung-dong, Yusung-gu, Taejon, 305-701, South Korea*

## 1. Introduction

Since Weber[1] measured the luminescence spectra and decay properties of bismuth germanium oxide ($Bi_4Ge_3O_{12}$:BGO) and predicted it to be a new scintillator material, the research on growth, performance and application of BGO has been developed successfully. Various techniques have been employed to grow BGO single crystals which have great potential for the technological application in solid-state devices[2].

We have already developed the conduction dominated HEM model for the growth of BGO crystals to simulate the heat transfer process[2]. The solutions from the model include the location of the melt/solid interface and temperature field in the crucible. The interface location of melt/solid is determined simultaneously by the equilibrium (isotherm) condition along the interface[3]. A reliable numerical method for transient simulation of the transition of melt/solid interface shape has been developed and a nonorthogonal mapping technique designed for computing it is introduced in ref. 2.

In the previous work[4], the effects of natural convection on the melt/solid interface shape in the HEM process was studied and the convective heat transfer should be considered to simulate the heat transfer process of HEM rigorously.

In this work, the model for heat and momentum transfer in the HEM for the growth of BGO crystals was introduced and the effects of various growth parameters on the interface shape, the temperature and flow field were examined.

## 2. Modelling

The heat exchanger method is a process which controls both the heat input and the heat extraction independently in a crystal growth furnace[5-7]. The schematic diagram of a HEM furnace is shown in Fig. 1a and the geometry of the mathematical model for HEM growth of BGO crystals is shown in Fig. 1b. The computational domain is the crucible which contains the melt and solid BGO.

Two-dimensional equations of continuity, momentum and energy in the melt of BGO are

$$\frac{1}{r}\frac{\partial}{\partial r}(ru)+\frac{\partial v}{\partial z}=0$$

$$\rho(\frac{\partial u}{\partial t}+u\frac{\partial u}{\partial r}+v\frac{\partial u}{\partial z})=-\frac{\partial P}{\partial r}+\mu\ [\frac{\partial}{\partial r}(\frac{1}{r}\frac{\partial}{\partial r}(ru))+\frac{\partial^2 u}{\partial z^2}]$$

$$\rho(\frac{\partial v}{\partial t}+u\frac{\partial v}{\partial r}+v\frac{\partial v}{\partial z})=-\frac{\partial P}{\partial z}+\mu\ [\frac{1}{r}\frac{\partial}{\partial r}(r\frac{\partial v}{\partial r})+\frac{\partial^2 v}{\partial z^2}]-\rho z$$

$$\rho C_p(\frac{\partial T}{\partial t}+u\frac{\partial T}{\partial r}+v\frac{\partial T}{\partial z})=k_m\ [\frac{1}{r}\frac{\partial}{\partial r}(r\frac{\partial T}{\partial r})+\frac{\partial^2 T}{\partial z^2}]$$

The energy equation for the solid phase is

$$\rho C_p\frac{\partial T}{\partial t}=k_s\ [\frac{1}{r}\frac{\partial}{\partial r}(r\frac{\partial T}{\partial r})+\frac{\partial^2 T}{\partial z^2}]$$

The interface location of the melt/solid BGO is obtained by the equilibrium condition along the interface[2-4].

The initial condition is

$$T(r,z,0)=T_m+\Delta T^{ex}$$

The boundary conditions are

$$T(r,0,t)=T_o, \qquad 0 \le r \le R_c$$

Adiabatic conditions are used for the temperature at the boundary except at the heat exchanger. No slip conditions for the velocity are used at the boundary except at the centerline where symmetric conditions are used. The latent heat effect is considered in the boundary condition at the interface.

## 3. Numerical analysis

The moving boundary problem for the temperature field in the whole calculation domain, the flow field in the melt phase and the melt/solid interface shape are solved using the modified finite element isotherm-Newton method which maps the problem to a new fixed domain in a nonorthogonal coordinate system[2-4,8]. As mentioned previously, nonorthogonal mapping rules designed for solving moving boundary problem is introduced in ref. 2.

The temperature field in both phases and velocities in the melt are represented by Lagrangian biquadratic finite element basis functions in a transformed coordinate system. Discontinuous piecewise linear basis functions defined at the centroid node of each element are used for the dynamic pressure in the melt and the one-dimensional quadratic basis functions are used for the melt/solid interface locations. An implicit Euler algorithm is used for the time integration. The solutions are obtained in a new fixed domain and mapped back to the original coordinate system. Typical finite element meshes mapped back to the original coordinate system are shown in Fig. 2.

## 4. Results and discussion

Calculations were performed using the growth parameter values in Table 1 for the model of the HEM growth of BGO crystals. The effects of the growth parameters on the interface shape, temperature and flow were investigated.

The temperature field in the crystal, flow field in the melt in terms of the stream function values and the location of the moving interface at several time steps for the standard case are shown in Fig. 3. As shown in Fig. 3, the hemispherical interface shape of the solid/melt BGO becomes planar when the interface crosses the corner of the crucible.

To investigate the curvature of interface, the deflection of the melt/solid interface $\Delta$ at each time step is defined as

$$\Delta \equiv \max{}_i(h_z)_i-\min{}_i(h_z)_i, \qquad i=1,\cdots,M.$$

The deflection of melt/solid interface is shown in Fig. 4. The maximum interface deflection occurs when the melt/solid interface is at the corner of the crucible and the interface shape changes its type from hemispherical to planar.

### *Effects of the excess heating temperature*

The excess heating temperature is one of the important growth parameters which affect the deflection of the melt/solid interface. As the excess heating temperature was increased, the maximum interface deflection decreased, because more heat should be extracted. As it is increased, more growth time for the whole process is required but the quality of the BGO crystal may be improved due to higher planarity of the interface.

### *Effects of the heat exchanger temperature*

The heat exchanger temperature can be controlled by the flow rate of the helium gas through the heat exchanger. The heat exchanger temperature hardly affects the maximum deflection of melt/solid BGO interface in the investigated temperature range.

## 5. Conclusions

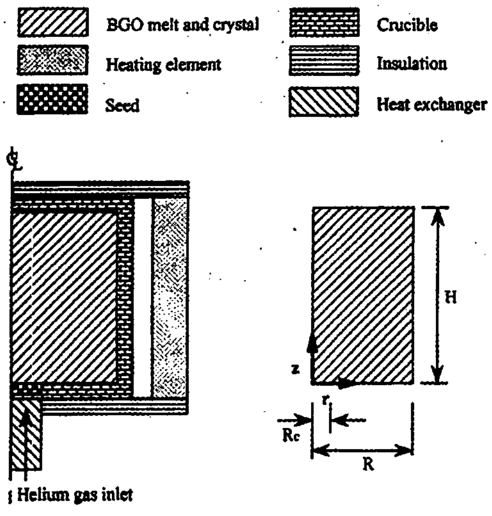
The model for heat and momentum transfer in the HEM for the growth of BGO crystals was introduced and the effects of various growth parameters on the interface shape, the temperature and flow field were examined. The hemispherical interface shape of the solid/melt BGO becomes planar as the crystal is grown by HEM. The maximum interface deflection occurs when the melt/solid interface is at the corner of the crucible.

## References

[1] M.J. Weber and R.R. Monchamp, J. Appl. Phys. 44 (1973) 5495.
[2] J.H. Wang, D.H. Kim and D.S. Chung, Korean J. Che. E. 13 (1996) 503.
[3] H.M. Ettouney and R.A. Brown, J. Comput. Phy. 49 (1983) 118.
[4] J.H. Wang and D.H. Kim, J. KACG 7 (1997) 41.
[5] F. Schmid and D. Viechnicki, J. Am. Ceram. Soc. 53 (1970) 528.
[6] D. Viechnicki and F. Schmid, J. Crystal Growth 26 (1974) 162.
[7] J.L. Caslavsky, in: Crystal Growth of Electronic Materials, Ed. E. Kaldis (Elsevier, Amsterdam, 1985) p. 113.
[8] L.H. Ungar, N. Ramprasad and R.A. Brown, J. Sci. Comput. 3 (1988) 77.

Table 1. Thermophysical properties and the growth parameters

| Growth Parameters | Meaning | Values |
|---|---|---|
| $\rho$ | density of BGO | 7.12 g/cm3 |
| $C_p$ | specific heat | 0.28 J/gK |
| $k_s$ | thermal conductivity of solid BGO | 0.019 W/cmK |
| $k_m$ | thermal conductivity of melt BGO | 0.0095 ~ 0.038 W/cmK |
| $T_m$ | melting temperature of BGO | 1050 °C |
| $h_f$ | latent heat | 150.37 J/g |
| $\beta$ | thermal expansion coefficient | $0.8 \times 10^{-4}$ $K^{-1}$ |
| $\mu$ | viscosity | 0.9256 g/cm • sec |
| $H$ | crucible height | 3 cm |
| $R$ | crucible radius | 1.5 cm |
| $R_c$ | cooling zone radius | 0.375 cm |
| $\Delta T^{ex}$ | excess heating temperature of melt | 25 ~ 100 °C |
| $T_0$ | heat exchanger temperature | 0 ~ 500 °C |



(a) (b)

Fig. 1. A typical HEM furnace and the numerical domain of the model.

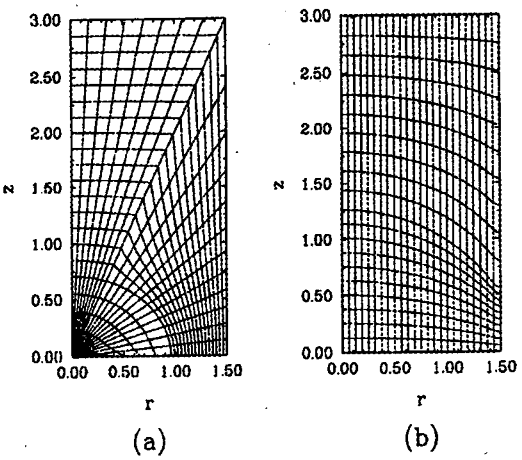
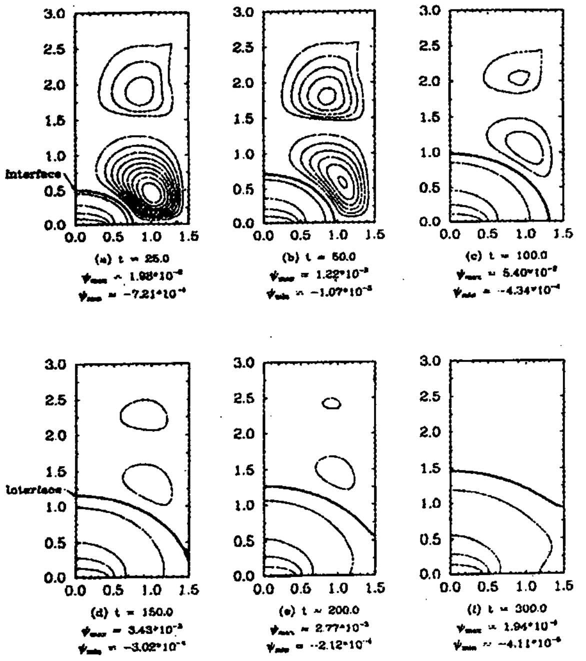


Fig. 2. Typical finite element meshes projected into the original coordinate system.



(a) t = 25.0 $\psi_{max}$ = 1.98*10$^{-3}$ $\psi_{min}$ = −7.21*10$^{-4}$

(b) t = 50.0 $\psi_{max}$ = 1.22*10$^{-3}$ $\psi_{min}$ = −1.07*10$^{-5}$

(c) t = 100.0 $\psi_{max}$ = 5.40*10$^{-4}$ $\psi_{min}$ = −4.34*10$^{-4}$

(d) t = 150.0 $\psi_{max}$ = 3.43*10$^{-3}$ $\psi_{min}$ = −3.02*10$^{-5}$

(e) t = 200.0 $\psi_{max}$ = 2.77*10$^{-3}$ $\psi_{min}$ = −2.12*10$^{-4}$

(f) t = 300.0 $\psi_{max}$ = 1.94*10$^{-4}$ $\psi_{min}$ = −4.11*10$^{-6}$

Fig. 3. Transient temperature fields in the crystal and flow field in the melt for the standard case ($\Delta T$=250°C, $\Delta\psi_{max}=2.0\times10^{-3}$ and $\Delta\psi_{min}=2.0\times10^{-5}$)

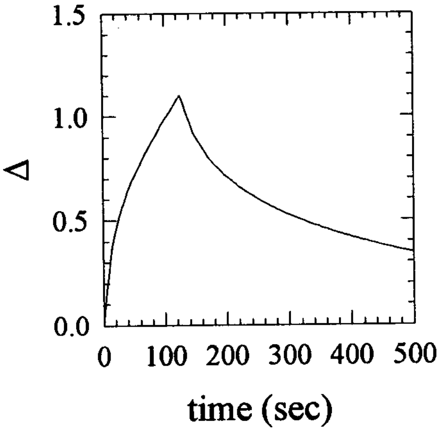


Fig. 4. The melt/solid interface deflection in the standard case.