Journal of Ceramic Processing Research. Vol. 17, No. 11, pp. 1175~1180 (2016)

J O U R N A L O F
Ceramic Processing Research

Effect of heat shield on the thermal fields during sapphire crystal growth by Kyropoulos method

Yuqing Guo, Nannan Ge, Kai Wang, Meng Shen and Ming Wang*
Key Laboratory on Opto-Electronic Technology of Jiangsu Province, School of Physics Science and Technology, Nanjing Normal University, Nanjing 210023, China

In this paper, the numerical computation was performed to investigate the influence of the heat shields of sapphire crystal furnace on temperature distributions for the Kyropoulos sapphire crystal growth process. The heat shields with different shield radius, shield layers and the shield materials were considered, the Kyropoulos growth process was simulated by the finite-element method. It is found that radial temperature gradient of crystal would increase along with the increase of the shield radius and the decrease of the layers of top shield. If properly increasing the shield radius and the layers of the top shield, axial temperature gradient in crystal would decrease, but the layers of side shield have little influence on the temperature in crystal. The heat shields made of different materials were compared by analyzing the crystal temperature distribution. Simulation results showed that temperature distribution in crystal by W-Mo shield is much closer to that of ZrO_2 shield, both of temperature distribution are higher than that of graphite heat shield.

Key words: Kyropoulos technique, Sapphire, Numerical simulation, Heat shield, Thermal field analysis.

Introduction

Sapphire crystal has been widely used in the applications of substrate of optoelectronic device and optical windows [1-2]. Kyropoulos (KY) technique has become an important technique for growing large sized high-quality sapphire single crystals, which can be achieved by optimizing and improving the crystal growth process inside the furnace. Such improvements of furnace structure can be done with an efficient support of numerical simulation [3-5]. The furnace structure is very important for the production of high quality sapphire single crystals. Geometrical parameters of crucible (size, location and shape) would make direct influence on the crystal growth. Chen and Lu [6] studied the influence of crucible shape on the shape of melt/crystal interface during crystal growth by heat exchanger methods (HEM). They found that curved crucible bottoms decrease d the convexity of the melt/ crystal interface during crystal growth. Lee et al. [7] investigated the crucible geometry on temperature distribution, melt convection behavior and shape of the melt/crystal interface during the Kyropoulos sapphire crystal growth process. C. Chen et al. [8] studies the effect of crucible shape on heat transport and melt-crystal interface during the Kyropoulos sapphire crystal growth and finds that the growth system with proposed crucible shape can result in more suitable thermal and flow fields. Wu, Zhao and Liu [9] investigated the melt–crystal interface shape and the thermal stress distribution in the growing crystals and reveal the effects of the crucible cover on the crystal growth. Some researchers [10-11] analyzed the effect of heat shield on the shape of solid-liquid interface and temperature field during the process of Czochralski (CZ) silicon crystal growth. Jin and Fang [12] summarized several thermophysical properties of sapphire in a wide temperature range, and adopt them in the simulation of sapphire growth process.

However, the effect of the heat shield on the KY sapphire crystal growth has not been studied so far. In this study, we investigate the influence of the heat shields of sapphire crystal furnace on the thermal fields for the Kyropoulos crystal growth process. The performance of heat shields with different shield radius, shield layers and the shield materials were analyzed, and the thermal field of sapphire crystal furnace was simulated and optimized by the finite-element method.

*Corresponding author:
Tel : +86-189-518-91-385
E-mail: wangming@njnu.edu.cn

Model

Model Building

Schematic illustrations of the industrial KY furnace with a heat shield utilized in this study are shown in Fig. 1, simplified model is assumed that the system is axisymmetric and all materials are physically isotropic. The finite element method is adopted during calculation, the computations is made using the CGSim package [13]. STR's CGSim is widely used for simulating calculation of crystal growth, including sapphire crystal growth. It not only can be used in Czochralski methods

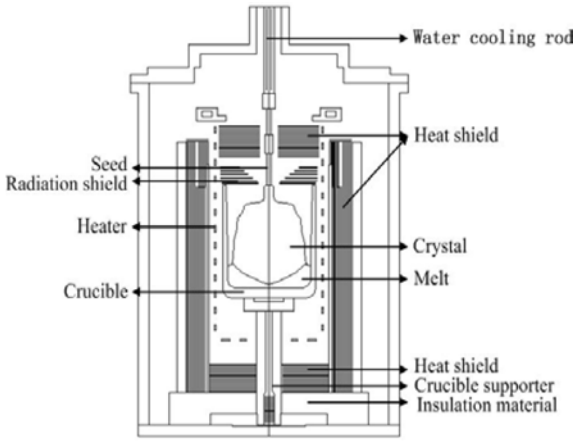

Fig. 1. A schematic of the KY furnace and crystal growth models.

but also used in Kyropoulos methods. This paper used CGSim to simulate and optimize the growth process of Kyropoulos sapphire crystal. The computational model employed in the present study is meshed with triangular and quadrilateral elements about 100000 grids.

KY furnaces mainly consist of crucible, tungsten resistance heater, heat shield, heat exchanger, and insulations. KY furnaces can allow the growth of 80 kg crystal. Three groups of heat shields, namely, the radiation shields, the side shield and the bottom insulation parts, are arranged in furnace satisfying the hot zone requirement. The heat shields to prevent heat loss from the furnace are formed by several layers of highly reflective slices. The crucible located inside the cylindrical heat shields formed by the Mo slices.

Model description

The equations for the energy, continuity, momentum and proper boundary conditions are considered during crystal growth process. The model accounts for heat exchange between various parts, includes heat conduction in the solid parts, convection heat transfer in the melt, radiation heat transfer between the various surfaces as well as latent heat dissipated by crystallization. This study is mainly focused on the influence of heat shield. The heat is transferred to the heat shields by thermal radiation; heat transfer inside the heat shield is heat conduction; the heat is carries away on the outside surface of heat shield by circulating cooling water in the furnace wall.

In order to simplify the simulations, it is assumed that (1) the crystal growth process is quasi-steady state, (2) the melt is incompressible axisymmetric Newtonian fluid, and (3) the kinetic effect of the crystal growth is not considered.

The governing equations for the thermal and flow fields can be expressed as follows:

For the melt: Navier-Stokes equation:

$$\rho_L \upsilon\cdot\nabla\upsilon = -\nabla\cdot p + \mu\nabla^2\upsilon - \rho_L g\beta_t(T-T_m)e_g \quad (1)$$

Continuity equations: $\nabla\cdot\upsilon = 0$ (2)

Heat transfer equation: $\rho_L c_L \upsilon_L\cdot\nabla T = K_L\nabla^2 T - \nabla\cdot q_r$ (3)

where the subscript L, S denotes the melt, crystal. In the above equation, ρ_L is the density, υ is the velocity vector of the melt, p is the pressure, is the dynamic viscosity, g is the gravitational acceleration, e_g is unit vector, β_r is the thermal expansion coefficient of the melt, is the temperature, T_m is melting point, c_L is the heat capacity, K_L is the thermal conductivity in the melt, and q_r is radiative heat flux.

For the crystal : Heat transfer equation of the crystal:

$$K_S\nabla^2 T = \nabla\cdot q_r \quad (4)$$

In the above equation, K_S is the thermal conductivity in the crystal.

(c) The heat-balance equation on the boundary between solid and transparent domains can be written as

$$q^{out} = \varepsilon\sigma T_k^{\ 4} + (1-\varepsilon)q^{in} \quad (5)$$

where q^{out} and q^{in} is the outgoing and incoming radiation flux, respectively, ε is the emissivity of solid material and σ is Stephen Boltzmann constant.

(d) The balance equation of crystallization:

$$-K_s n_i\nabla T|_S - K_L n_i \nabla T|_L = \rho_s VH \quad T = T_m \quad (6)$$

In the above equation, $\nabla T|_S$ and $\nabla T|_L$ is the temperature gradient of crystal and melt on solid-liquid interface, ρ_s is the crystal density, H is the latent heat of crystallization, V is the crystallization rate and n_i is the direction vector. The crystallization rate is proportional to the difference between heat fluxes in the crystal and melt in the melt/crystal interface. To increase the crystallization rate, one should raise the crystal temperature gradient, and reduce the melt temperature gradient. The melt temperature

Table 1. Physical properties of sapphire used in calculations. [14]

Description/ Unit	Value
Thermal conductivity of melt/(W/m/K)	2.05
Thermal conductivity of crystal/(W/m/K)	5.8
Density of melt/(kg/m^3)	3030
Density of crystal/(kg/m^3)	3970
Heat capacity of melt/(J/kg)	1260
Heat capacity of crystal/(J/kg)	1430
Emissivity of melt	0.33
Emissivity of crystal	0.4
Melting point/K	2313
Dynamic viscosity of melt/(Pa*s)	0.057
Poisson's ratio	0.29
Young modulus/Pa	44e10

gradient can be reduced by reducing the heater power, but it is necessary to keep the melt temperature above the melting temperature.

Physical properties of sapphire used in simulated calculation are shown as Table 1.

Simulating calculation

Temperature field calculation inside the crystal growth furnace is rather complex, which accounts for thermal radiation, heat conduction, heat convection as well as the latent heat dissipated by crystallization. Temperature gradient can be divided into axial thermal gradient and radial thermal gradient.

Fig. 2 demonstrates the calculation results of the temperature distribution in the iso-diameter growth process. The computations show that temperature is low at center part while high at the border part; besides, temperature is low at the upper part and high at the lower part. The isothermal is convex at the bottom and is dense at neck part. The convexity of the solid-liquid interface considering internal radiation is larger than that without internal radiation [15].

As shown in Fig. 2, the radial and axial temperature distribution in the crystal was analyzed. The L1 and L2 directions have been specified. L1 represents the direction from the seed at the center line to the crystal from the crystal-melt interface; L2 represents the direction along crystal radius direction.

Fig. 3 shows the temperature distribution on the axis

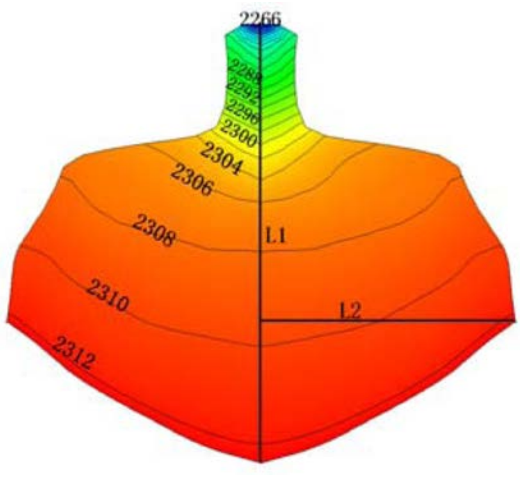

Fig. 2. Temperature distribution in the crystal.

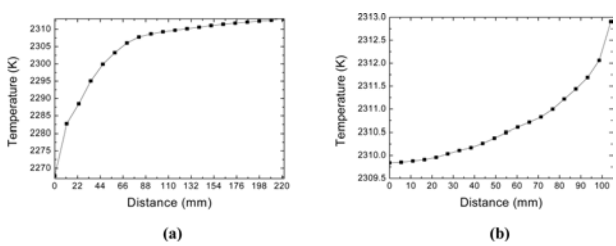

Fig. 3. Temperature distribution in crystal: (a) on the axis and (b) on the radial.

(a) and radial (b) of crystal during the iso-diameter growth process. Along the crystal axis direction L1, the temperature increase with the distance; however, the temperature variation gradually becomes small, which means axial temperature gradient decreases. While along the crystal radial direction L2, the temperature increase with the distance; and the temperature variation becomes larger, which means radial temperature gradient increases.

Because the cooling water leads to large heat dissipation near the neck in the crystal, axial temperature variation is much larger on there. The crystal can absorb more heat from the crucible sidewall towards the crystal, the maximum radial temperature variation in the crystal occurs near the sidewall of the crucible.

The temperature gradients inside the crystal are very low with higher temperature gradients in the melt occurring near the crystal-melt interface due to high effective thermal conductivity in the crystal and strong convection in the melt. The temperature gradient will affect the crystal quality. The large axial temperature gradient result in large thermal stress inside the crystal, and cause the defects and fracture in crystal. The small radial temperature gradient would cause some quality problems at the crystal shouldering and iso-diameter growth stages, such as twin crystal, scatter particle and bubble. Because the large diameter crucible and resistance heater are used in kyropoulos furnace, it may easily lead to the problem that the radial temperature gradient is too small and the axial temperature gradient is too big. In order to solve the problem, radial temperature gradient should be increased and axial temperature gradient should be decreased appropriately. In this paper, the adjustment of the temperature gradient is realized by changing the heat shield, including shield radius, shield layers and the shield materials.

Results and Analysis

In this study, three different factors of heat shield are considered in numerical simulations, which are carried out to investigate the effect of the heat shield on the temperature field in the crystal during the growth process.

Influence of radiation shield aperture on the crystal temperature

The radiation shield can reflects the radiation heat back into the furnace and reduce the heat loss. In order to study the influence of radiation shield aperture on the crystal temperature and the temperature gradient, we chooses aperture radius of 22.5 mm, 32.5 mm and 42.5 mm respectively, the calculation results are shown as Fig. 4.

Heat loss from the crucible increases with the increase of the radiation shield aperture. The axial temperature and radial temperature in crystal decrease as shown in Fig. 4(a-b). When the radius of radiation shield aperture increases from 22.5 mm to 42.5 mm, the

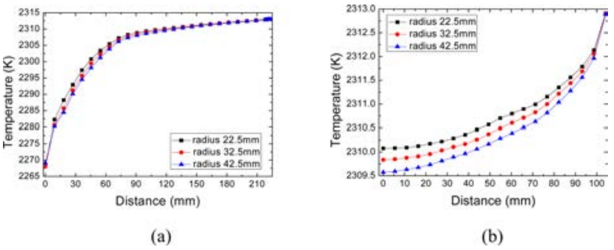

Fig. 4. Temperature distribution in crystal by different aperture radius of radiation shield: (a) on the axis and (b) on the radial

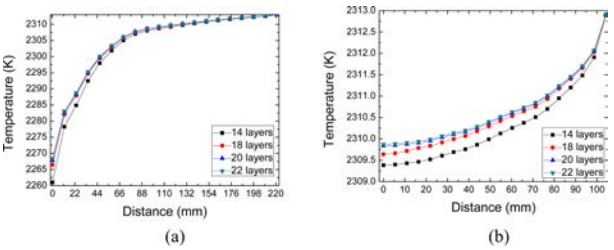

Fig. 5. Temperature distribution in crystal by different layers of top heat insulation: (a) on the axis and (b) on the radial.

axial temperature gradient in the crystal decrease from 2.056 K/cm to 1.962 K/cm, and radial temperature gradient in the crystal increase from 0.272 K/cm to 0.318 K/cm, respectively. That means the suitable aperture radius can increase radial temperature gradient and reduce the axial temperature gradient effectively. However, too big aperture radius will cause the heat energy loss from the crucible much, which is unfavorable for the crystal growing.

Influence of heat shield layers on the crystal temperature

The heat shields to prevent heat loss from the furnace are formed by several layers of highly reflective slices. Therefore, the thickness and layers are the important influencing parameters on the thermal insulation. The effect of the heat shield layers on the crystal temperature is discussed.

The thermal radiation is the main heat transfer mode between the heat shields, the radiative heat flux between the heat shields is:

$$q_r = \frac{\sigma(T_1^4 - T_2^4)}{(N+1)\left(\frac{2}{\varepsilon}-1\right)} \tag{9}$$

Where T_1 and T_2 is the temperature of the interior and exterior plane of the heat shield, respectively; ε is the material emissivity of the heat shield, N is the layers of heat shield. The radiative heat flux between the interior and exterior plane of the heat shield decreases with the increases of the layers N. But considering the manufacturing cost of the heat shield, we need to choose the layers of heat shield reasonably.

Influence of layers of top heat shield

The temperature field in crystal is simulated using different layers of top heat shield, including 14, 18, 20 and 22 layers. The axial temperature distribution in crystal and radial temperature crystal distribution in crystal are showed in Fig. 5(a-b) respectively.

Because the kyropoulos furnace is equipped with the water cooling system and its relative open top structure, the top part of crystal would continue to transfer heat to the cooler surroundings. The heat loss from the furnace increases with the decrease of the top shield layers, both the axial temperature and radial temperature in crystal are relatively low. When the heat shield layers increases from 14 to 20, temperature in the crystal increases with the increase of the layers. The temperature rise reaches maximum nearby the seed crystal, about 7 K. It can be found that the axial temperature gradient and the radial temperature gradient decreases with the increases of layers correspondingly. When the shield layers increase from 14th layer to 20th layer, the axial temperature gradient in the crystal decrease from 2.333 K/cm to 2.043 K/cm and radial temperature gradient in the crystal decrease from 0.336 K/cm to 0.295 K/cm, respectively. When the layers exceed 20th layer, temperature variations in crystal are very small. However, too many layers of heat shield will cause the heat energy loss from the furnace much, and increase the cost of equipment and growth.

Influence of layers of side shield

The influence of the layers of side shield on the temperature field in crystal is analyzed by changing the layers of side shield, including 16, 10, 7, and 5 layers. The axial temperature distribution in crystal and radial temperature crystal distribution in crystal are showed in Fig. 6(a-b), respectively.

Fig. 6 shows that temperature distribution in crystal hardly changes with different layers of side shield. Through calculation, it can be obtained that when the side layers decrease from 16th layer to 5th layer, the axial temperature gradient in the crystal decrease from 2.035 K/cm to 2.054 K/cm and radial temperature gradient in the crystal decrease from 0.295 K/cm to 0.297 K/cm, respectively. Results indicate that layers of side shield have little influence on the temperature in crystal. Therefore, we can appropriately increase the side shield layers to reduce the energy consumption cost in the crystal growth process.

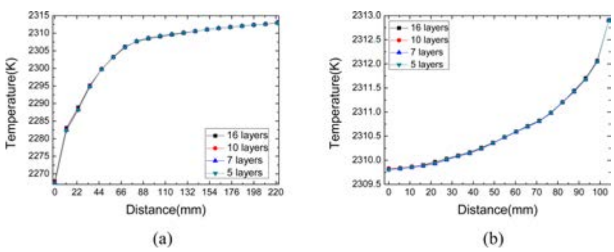

Fig. 6. Temperature distribution in crystal by different layers of side heat insulation. (a) on the axis and (b) on the radial.

Table 2. Physical properties of thermal insulation material.

Materials	Melting point/K	Density/ (kg/m^3)	Emissivity
Tungsten	3653	19350	0.287
Mo	2873	10200	0.25
Graphite	3823	2250	0.8
ZrO_2	2873	6000	0.2

Influence of thermal insulation material on the crystal temperature

The heat shield elements included top multi-layers sheets on the crucible, side-wall insulation layer of foam and bottom thick insulation layer. The material of heat shield is also one of the significant factors which affect the sapphire crystal growth. There are three common thermal insulation material used in the heat shield, including tungsten-molybdenum (W-Mo)[15], ZrO_2 fibers [15], graphite felt [16].

The heat loss depends on the thermal conductivity and emissivity of material. The physical and optical properties of the material used in the analyses are given in Table 2.

From the Table 2, the decreasing order of the thermal conductivity of thermal insulation material is summarized as follows: tungsten-molybdenum; graphite; zirconia (The physical property of high-temperature zirconia is hard to obtain by experiment and it can be approximately considered as a constant in calculation). The order of the emissivity of the thermal insulation material may be shown as follows: graphite; tungsten-molybdenum; zirconia.

Tungsten-molybdenum has the characteristics of high temperature resistance, high hardness, difficult to melt and volatile. It also has good lateral conductivity, so that heat can transfer from the metal sheet to the surrounding environment.

Graphite has excellent performance of heat insulation with an extremely high melting point. At present, porous graphite foam or graphite felt are adopted as heat insulation material.

Zirconia is a good insulating material characterized by high-temperature resistance, oxidation resistance, corrosion resistance and low thermal conductivity.

In sapphire crystal growth furnace, heat insulation principle of tungsten-molybdenum sheets, graphite felt and zirconia oxide is different, tungsten-molybdenum screens insulate heat by strong reflection, while graphite felt and zirconium oxide insulate heat by low thermal conductivity of materials. Different thermal insulation materials will affect the crystal growth temperature. Fig. 7 and Fig. 8 show the axial temperature distribution and radial temperature distribution in crystal at various growth processes respectively when using different heat shield materials.

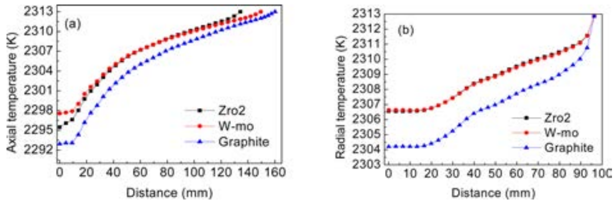

Fig. 7. Temperature distribution in crystal using different thermal insulation material at the shouldering growth stage (3.6 kg) (a) on the axis and (b) on the radial.

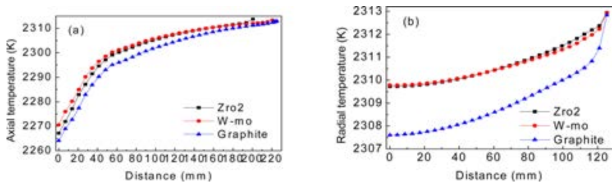

Fig. 8. Temperature distribution in crystal using different thermal insulation material at iso-diameter growth stage (21 kg): (a) on the axis and (b) on the radial.

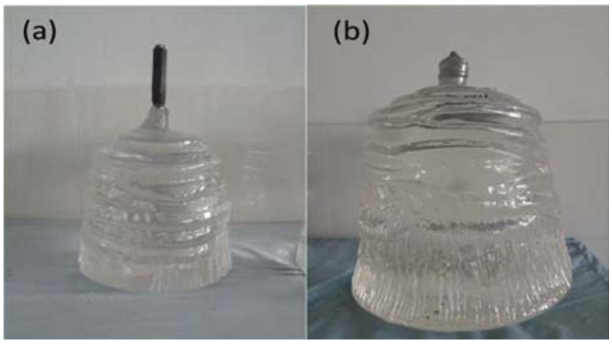

Fig. 9. Sapphire boule with weight (a) 30 kg and (b) 80 kg.

The temperature distribution is compared for different heat shields made of W-Mo, Graphite and ZrO_2 in different growth process, and the results show that temperature distribution in crystal by W-Mo heat shield and ZrO_2 heat shield get close in the same growth stage. At the same time, both the axial temperature and radial temperature in crystal by W-Mo heat shield and ZrO_2 heat shield are high than that by using graphite heat shield.

Heat in the furnace will be mainly transferred to thermal insulation material through thermal radiation. Comparing three kinds of heat insulation materials for radiation heat absorption, the lower emissivity of opaque object surface, the higher the reflectivity, the less heat loss. The heat loss of graphite heat shield is far greater than that of tungsten-molybdenum and zirconia. Therefore, the temperature in crystal is lower when using thermal insulation material of graphite.

Based on the result and analysis, we can optimize actual heat shield parameters such as increasing the shield radius, changing the layers of top heat shield, and replacing the graphite shield with the W-Mo or ZrO_2 shield.

Fig. 9 shows the grown sapphire boule with weight (a) 30 kg and (b) 80 kg produced by improved kyropoulos furnace. The crystal has no obvious defects such as bubbles, inclusions and other defects in the crystal, no cracking on the shoulder and the diameter of crystal, its utilization rate is very high.

Conclusions

In the present study, the numerical simulation has been performed to investigate the effect of heat shield on the flow and temperature field of the system by Kyropoulos method.

Comparing the simulation results for different shield radius, shield layers and the shield materials, and analyzing the influence of heat shield on the crystal temperature, we can make the following conclusions:

1) The heat shield can influence the thermal field in crystal. The shield radius, shield layers and the shield materials are important factors which influence the crystal temperature distribution.

2) The axial/radial temperature in crystal decrease with the increase of the radiation shield aperture. The suitable aperture radius can increase radial temperature gradient and reduce the axial temperature gradient effectively.

3) When the layers of top heat shield increases, temperature in the crystal increases and the axial/radial temperature gradient decreases correspondingly. The layers of side shield have little influence on the temperature in crystal.

4) The temperature distribution in crystal by W-Mo heat shield and ZrO_2 heat shield get close in the same growth stage. At the same time, both the axial temperature and radial temperature in crystal by W-Mo heat shield and ZrO_2 heat shield are high than that by using graphite heat shield.

Acknowledgments

This work was supported by the Jiangsu Province Prospective Joint Research Project under Grant BY2012005.

References

1. M.S. Akselrod, F.J. Bruni, J. Cryst. Growth 360 (2012)134-145.
2. G. Wang, H. Zuo, H. Zhang, Q. Wu, M. Zhang, X. He, Z. Hu, Z. Lin, Materials and Design 31 (2010) 706-711.
3. S.E. Demina, E.N. Bystrova, M.A. Lukanina, et al. Optical Materials 30 (2007) 62-65.
4. C.H Chen, J.C. Chen, C.W. Lu et al., J. Cryst. Growth 318[1] (2011) 162-167.
5. C.H Chen, J.C. Chen, C.W. Lu et al., J. Cryst. Growth 352[1] (2012) 9-15.
6. J.C. Chen, C.W. Lu, J. Cryst. Growth 266 (2004) 239.
7. W.J. Lee, Y.C. Lee, H.H. Jo, et al., J. Cryst. Growth 324[1] (2012) 248-254.
8. C. Chen, H.J. Chen, W.B. Yan, et al., J. Cryst. Growth 388[1] (2013) 29-34.
9. M. Wu, W.H. Zhao, L.J. Liu, et al., J. Cryst. Growth 404[1] (2014) 130-135.
10. M.G. Vasiliev, O.N. Budenkova. J. Cryst. Growth 275[1-2] (2005) 745-750.
11. W. Su, R. Zuo, K. Mazaev, V. Kalaev, J. Cryst. Growth 312[1] (2010) 495-501.
12. Z.L. Jin, H.S. Fang, N. Yang, et al., J. Cryst. Growth 405[1] (2014) 52-58.
13. S.E. Demina, E.N. Bystrova, V.S. Postolov, et al., J. Cryst. Growth 310[7] (2008) 1443-1447.
14. STR Group, CGSim Material Data base: v.9.3[DB/CD], St. Petersburg, Russia: Richmond VA, 2010 [2009-7-17]. http://www. semitech. Us.
15. H.S. Fang, Z.L. Jin, M.J. Zhang, Z. Zhang, C.J. Zhao, International Journal of Heat and Mass Transfer 67 (2013) 967-973.
16. W. von Ammon, E. Dornberger, H. Oelkrug, H. Weidner. J. Cryst Growth 151[3] (1995) 273-277.