Journal of Ceramic Processing Research. Vol. 17, No. 3, pp. 205~212 (2016)

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

Hot zone design optimization for YAG single crystal growth

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The YAG crystal growth industry will follow the same path as the semiconductor industry with cost becoming the main driver. Thus, it become imperative that models be developed that can be used for scaling purposes for the design of large YAG crystal growth systems. Analysis of thermal phenomena in the crystal growing by induction heating hot zone and resistance heating carbon hot zone has been addressed. To grow a good quality, productivity improvement, and large size of YAG crystal growing is shortfalls of current induction heating Czochralski method. The simulation includes the temperature gradient in melt and melt-crystal interface, growing zone. Resistance heating has stable temperature gradient in melt and higher gradient in melt-crystal interface which can be controlled by hot zone design change. Power consumption also improved by using lower thermal conductivity carbon insulation materials.

Key words: Hot zone, YAG, Single crystal growth, Uniformity.

Introducton

$Y_3Al_5O_{12}$ is commonly called yttrium aluminum garnet (YAG). YAG is advnaced ceramic with interesting optical and mechanical properties [1, 2]. In 1965, Union Carbide produced the first commercial Nd : YAG laser crystal. When doped with a lanthanide element, YAG can be widely used as solid-state laser material such as frequency-doubled continuous wave, high-energy Q-swithed, and so forth in applicaions to the medical, industrial, military and scientific research. While the field of optics has advanced at a rapid pace to develop and improve such lasers it would seem that the crystal growth of garnet has not advanced quite at the same pace. The main YAG growth techniques are the conventional Czochralski (CZ) [3, 4]. And other techniques has been developed such as temperature gradient technique (TGT) [5], horizontal directional solidification (HDS) [6], hydrothermal growth [7], EFG/Stepanov technique [8], heat exchange method (HEM) [9].

To achieve this progress, numerous advances to the understanding of the role that liquid and crystal composition, interface shape/fluid dynamics, thermal geometry and hot zone design have on the quality of the resulting crystal had to be determined. In this paper the influence of the hot zone design on the quality and yield of YAG crystals with 2 inches is numercially studied using software package CGSim ver.15.1(STR).

Since the Czochralski (CZ) technique is the conventional method for growing YAG single crystal, commercially available size of ingot diamaeter is limited with 4 inches due to lack of control temperature gradient. As the demand for YAG and their uses increases, the need for the growth of larger crystals with improved qualty will also increase, the YAG crystal growth industry will follow the same path as the semiconductor industry with cost becoming the main driver. This has already happened for Sapphire(Al_2O_3) material. However, unlike the semiconductor industry, adequate models for the design of these large YAG crystal growth furnaces did not exist and their ultimate configuration was based on trial-and-error methodology that, for large system, can become costly. For example, the investment in one large Iridum crucible could be as much as $100,000. Thus, it bcomes imperative that models be developed that can be used for scaling purposes for the design of large YAG crystal systems. Today, the lack of understanding are many and such models do not exist. There are two groups, who deals with YAG materials itself and deals the physical design of the furnace and the mothod of heating.

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Induction heating vs. resistance heating

The CZ method can be categorized depending on whether it is heated by induction heating or resistance heating. Each method has its pros and cons. Induction heating requires less energy input and is therefore easier to heat to high temperatures, but its reproducibility is low and it does not allow for crucible rotation. In contrast, resistance heating involves high energy input, but it allows for crucible rotation and offer relatively good reproducibility. Reproducibility is of critical importance in mass production, indicating that YAG crystal growth by the resistance heating CZ method has the potential to be the leading technique in the YAG single crystal market.

The high operating temperature required for the growth of YAG materials generally requires that the crucible becomes the active source for heat generation within the furnace, i.e. the crucible is heated by RF energy. The coupling of the crucible to the RF field is strongly dependent upon the diameter and length of the RF coil as well as the shape (diameter and height) of the crucible. Also, the position of the crucible within the RF coil strongly influences the thermal gradients generated within the liquid [10]. One of the most difficult tasks in designing these large systems is the establishment of a suitable radial thermal gradient in the liquid that allows good seeding and control led growth during the "shouldering" phase when the crystal is being brought out to the required diameter. In general, as the size of the system increases, the radial liquid thermal gradient decreases that making control of start more difficult [11].

Hot zone design for simulation

Based on 2 inches ingot growing, resistance heating system is compared with induction heating system by using same size of crucible in Fig. 1. Induction heating crucible is Iridum crucible with metal and ceramic materials, and Resistance heating crucible is low cost Mo/W with carbon insulation matierials that consist of graphite felt and graphite shield. Fig. 2 shows temperature distribution of resistance heating at hot zone area, crucuble and YAG melt. The center of heater area has highest temperature because graphite resistance is high. In the same way, crucible also has higher temperature at the center. However, bottom keeps low temperature and less convection all the way of growing process, which reflect introducing 2 zone heaters with bottom heater. For the YAG melt, key thermophysical properties of YAG are used in the Table 1 because YAG melt properties have measured the density, surface tension and viscosity of from 1970 to 2070 °C by V.J. Fratello *et al*. [12]. And the non-dimensional quantities have been evaluated using published values of thermo-physical properties of YAG [13]. In addition, simualtion parameters are taken by heating method in Table 2. In terms of insualation materials, induction heating is considered ZrO_2 and resistanance heating is based on graphite insulation from Morgan Advanced Materials [14].

It may have many hot zone design varities that considered previous research. The mirror like shield and water cooled heat shield are introduced for the purposed of latent heat is transported more effectively away [15]. Double crucible mehtod helps to prevent asymmetric crystal grwoth by omptimization of temperature gradient and seed centering [16]. Thermal shock casued by water flow rate, which influence on the growth process and optical quality and makes process instable [17]. To find the hot zone optimiation, five different cases of resistance

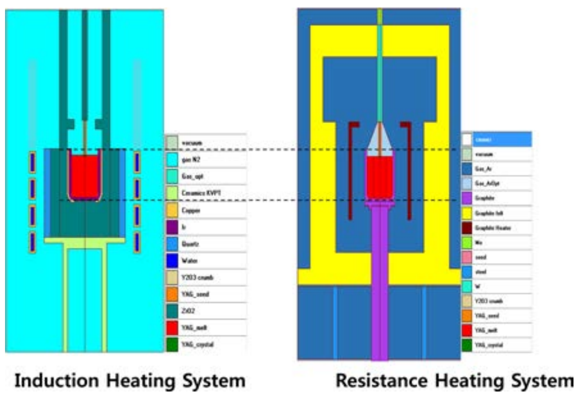


Fig. 1. Hot zone design for simulation: (a) induction heating system (b) resistance heating system.

Table 1. Key thermo physical properties of YAG used in the simulation.

Description	Unit	Values
Heat conductivity	W/(m*K)	4
Emissivity		0.3
Density	kg/m^3	3600
Heat capacity	J/kg*K	800
Melting temperature	K	2243
Latent heat	J/kg	4.55E + 05
Dynamic viscosity	Pa*s	0.0468
Marangoni coefficient	N/m*k	3.50E-05

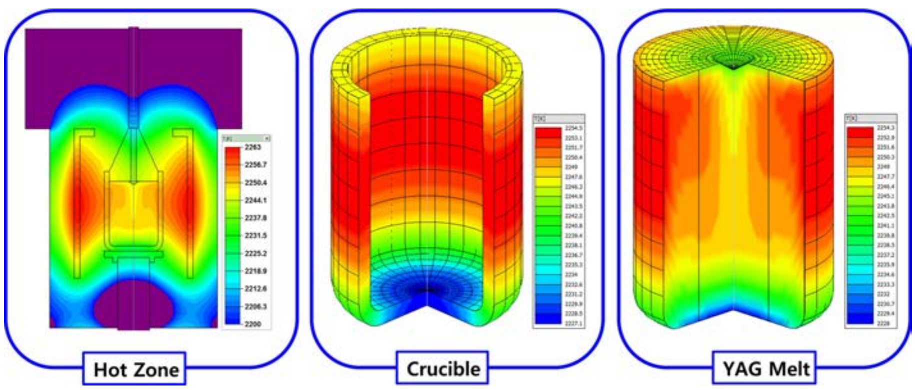


Fig. 2. Temperature distribution of resistance heating.

Table 2. Simulation parameters by heating method.

Induction Heating	Resistance Heating
- Inductor Frequency 8000 Hz	- Argon Atmosphere
- Nitrogen Atmosphere	- Mo Crucible
- Ir Crucible	→ Heat conductivity [W/(m*K)] : 105
→ Heat conductivity [W/(m*K)] : 147	→ Emissivity : 0.4
→ Electric conductivity [S/m] : 1.72e6	- Graphite insulation
- ZrO2 Insulation	→ Heat conductivity [W/(m*K)] : 0.76 (constant)
→ Heat conductivity [W/(m*K)] : 2.13	→ Emissivity : 0.66 (constant)
→ Emissivity : 0.8	→ Heat capacity [J/kg*K) : 2100 (constant)

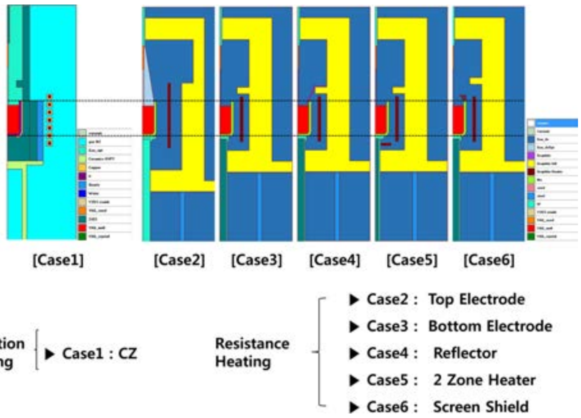


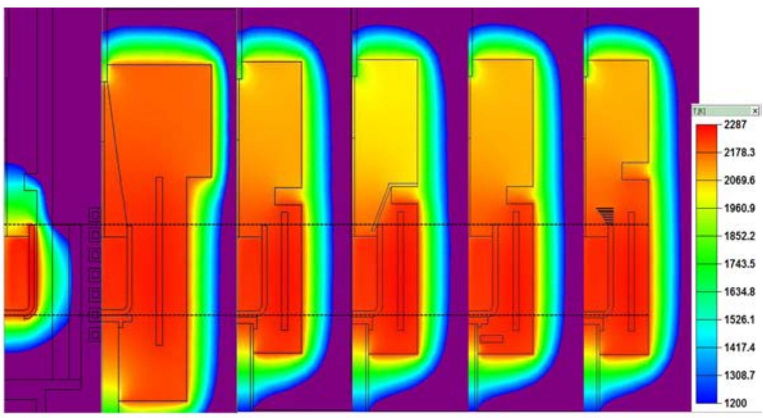
Fig. 3. Hot zone design variation.

heating is considered to compare with induction heating as a case of 1 in Fig. 3. Case 2 has top electrode that heater need to be longer to cover the hot zone, case 3 has bottom electrode that could be shorter, case 4 introduce shield that expect to reduce the heat dissipation, case 5 has two heaters that expect to improve temperature uniformity, case 6 has screen shield that expect to reduce the temperature fluctation.

Results and Discussion

Induction heating shows confined temperature distribution which increase the effectiveness of power consumption in Fig. 4. Top electrode, case 2, has wide and broad temperature distribution that consumes power highly. Bottom electode, case 3 to case 6, looks similiar confined temperature distribution and power consumption. However, it has difference at bottom and top of the hot zone by different factors. Power consumption in resistance heating can be optimized by two zone heaters.

Fig. 5, Heat flux and temperature distribution in melt, crucible side wall has high temperature due to close contact heat generation. From side to center, temperature changes 30 degrees, in contrast, top to bottom, temperature changes is less. Case 4 and 5 has improved bottom side temperature than case 1,3,6 by helping reflector and bottom heater. At the aspect of heat flux, case 1 and 2 has less dense that 3 to 6. The more amount of heat transferred per unit area per unit time from or to a surface, which effect in convection of melt is presented in Fig. 6. Case 1 and 4 has relatively high of center of convection and others are low center. Case 5 has a



	Case1	Case2	Case3	Case4	Case5	Case6
Power Consumption [kW]	7.91	19.33	9.61	9.43	9.25 Side:8.97 Bttm:028	9.61

Fig. 4. Temperature distribution and power consumption.

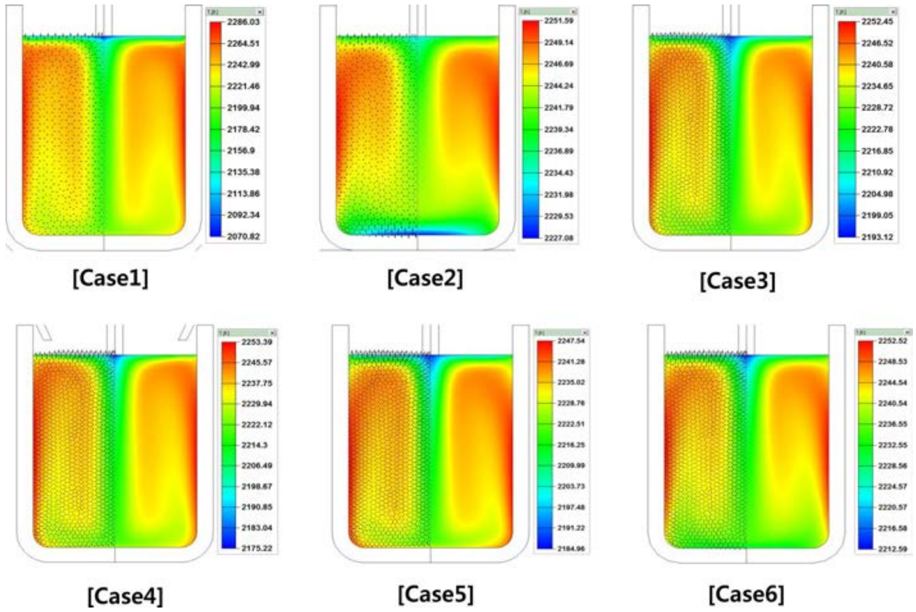


Fig. 5. Heat flux and temperature distribution in melt.

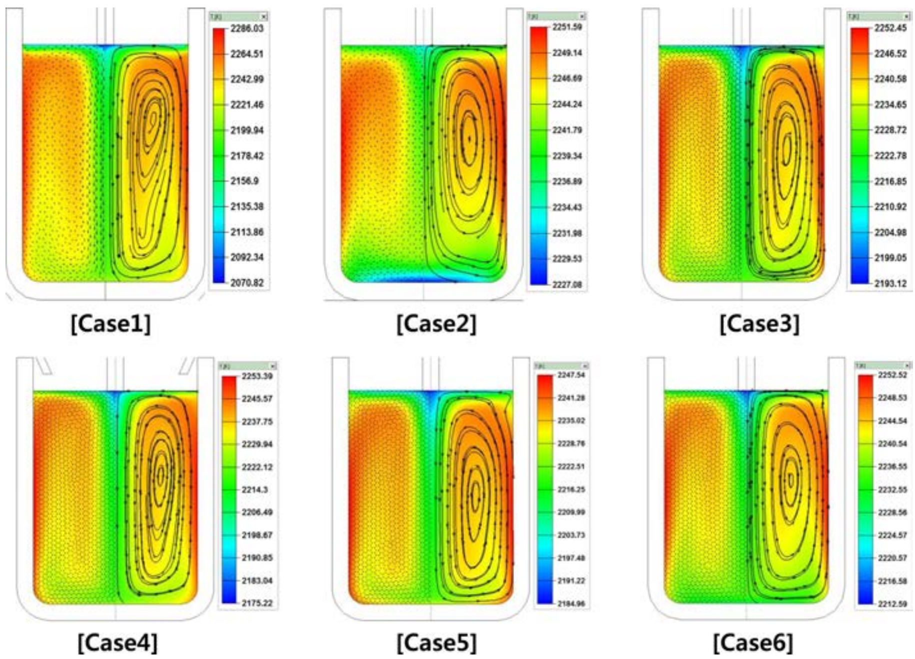


Fig. 6. Convection in melt.

good unifromity in convection. Heat transfer within YAG melt is dominated by convection, and the crystal growth interface shape is strongly influenced by the melt convection. Hence, the crystal growth interface in a YAG melt growth system may be explained in terms of the melt convection. The increased convexity which arises when crystal rotation is added to crucible rotation can also be improve the melt flow. In addition to the convexity, yet another factor which plays a major role indetermining the crystal quality is the thermal gradient at the crystal growth interface [18].

Temperature distribution in melt presented in Fig. 7, induction heating has high temperature distribution, but resistance heating show relatively less temperature gradient expect case 4 and 5. A good thermal gradient is also dependant on the appropriate furnace design. Finally, stable convection flow minimizes the temperature fluctuations of the melt. YAG has a narrow plasticity zone, so that dislocations and residual stresses are formed in the region of the crystal growth interface. As this is governed by the thermal gradient perpendicular to the crystal growth interface [19]. Temperature gradient is analyzed by vertical and horizontal in Fig. 8. In vertical, every case has sharp gradient that provide good convection of the melt, but horizontally smooth in resistance heating compare to induction heating. As bigger in size of

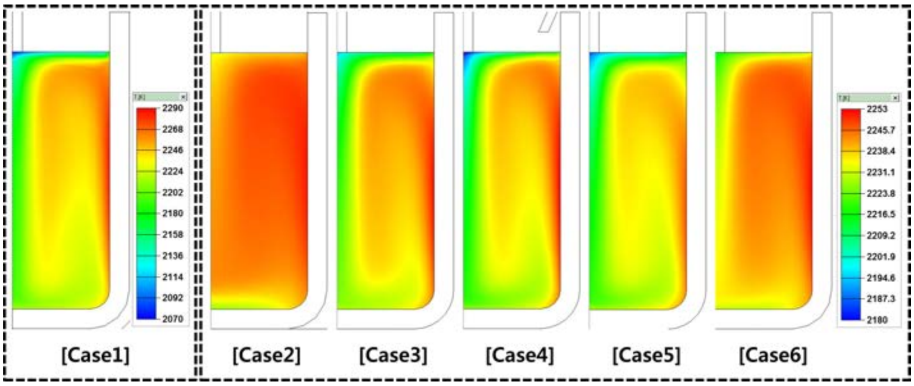


Fig. 7. Temperature distribution in melt.

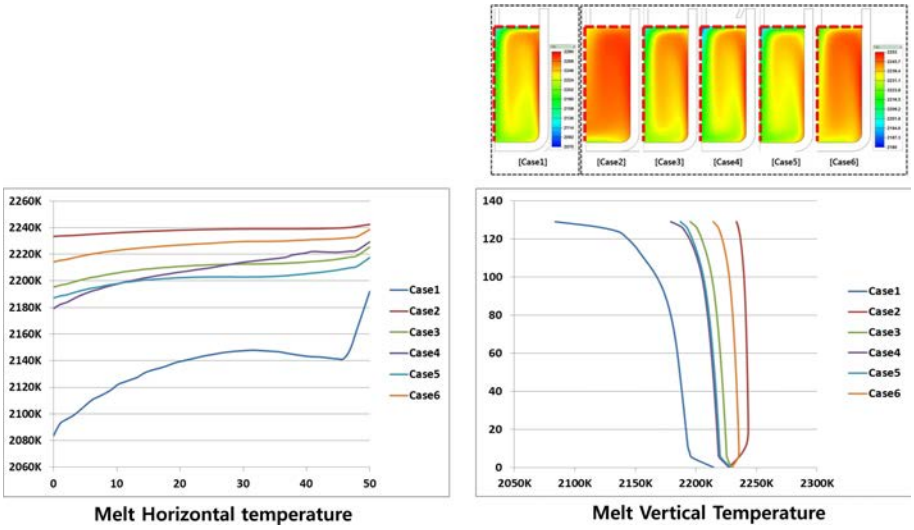


Fig. 8. Vertical and horizontal temperature distribution in melt.

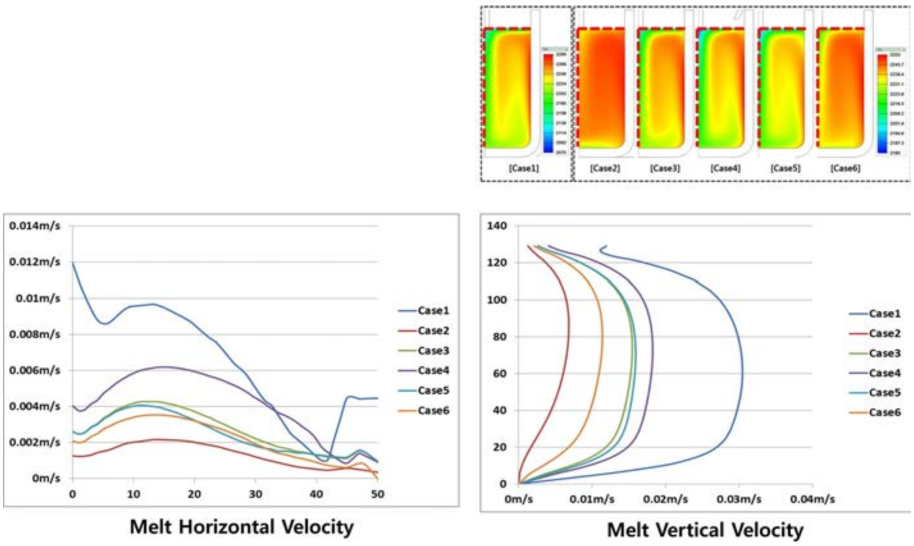


Fig. 9. Horizontal and vertical flow velocity in melt.

furnace, induction heating has difficult in control of temperature gradient which limited to four inches of the ingot diameter today.

Flow velocity in melt is also analyzed in vertical and horizontal to see the convexity, Fig. 9. Induction heating has higher convexity than resistance heating. High vertical velocity in the middle of the melt and getting lower in velocity at top and bottom. Induction heating has two times faster than resistance heating, which show 0.03 m/sec and 0.015 m/sec respectively. Horizontal

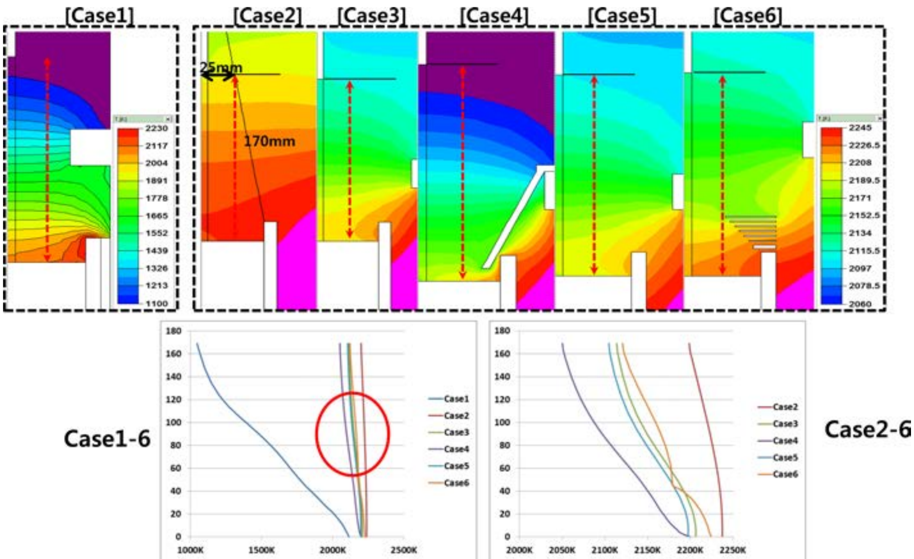


Fig. 10. Melt-crystal interface: vertical temperature gradient.

velocity is relatively lower than vertical, induction heating is 0.008 m/sec and resistance heating is 0.004 m/sec.

In the CZ process,the crystal–melt interface shape is dictated by crystal thermal history. When the interface shape is curved, the thermal field becomes non-uniform along the radial direction, which may result in non-uniformities in mechanical, optical, and electrical properties. Therefore, it is important to keep the interface shape as flat as possible during the single crystal growth. To achieve a good growth interface shape, it is necessary to optimize the temperature gradient. The temperature gradient has to be large enough to prevent both faceting at the interface and constitutional supercooling of the fluid close to the interface. However, too large of a temperature gradient must also be avoided since this can lead to large thermal stress which induces dislocation multiplication and sub-grain boundary formation in the hot crystal region [20]. Stabilizing the gradient is also an important factor. With stable gradients (or low temperature fluctuations in the melt), the growth rate will remain more constant and fluctuations in the dopant concentration will be minimized [21]. Fig. 10 show the melt crystal interface as vertical temperature gradient. Case 4 has large temperature gradient than the other cases expect case 1, however, case has too large gardient and difficult to control with high sensitivity of larger dimension of vertical and horizontal ingot. Case 4 looks able to achieve a good growth interface shape with stable gradients or low temperature fluctuations in the melt, the growth rate will remain more constant and fluctuations in the dopant concentration will be minimized.

Table 3. Power consumption changes by changing carbon heat conductivity.

	Case1	Case2	Case3
Heat conductivity W/(m*K)	0.5	0.76	0.9
Emissivity	0.66	0.66	0.66
Heat capacity J/(kg*K)	2100	2100	2100
Power consumption (kW)	7.08	9.61	10.89

The carbon insulation materials has a wide range of products that are available in the market, which can big impact in saving power consumption. Heat conductivity changes in Table 3, from 0.5 W/(m*K) to 0.9 W/(m*K), power consumption changes from 7.08 W to 10.89W,

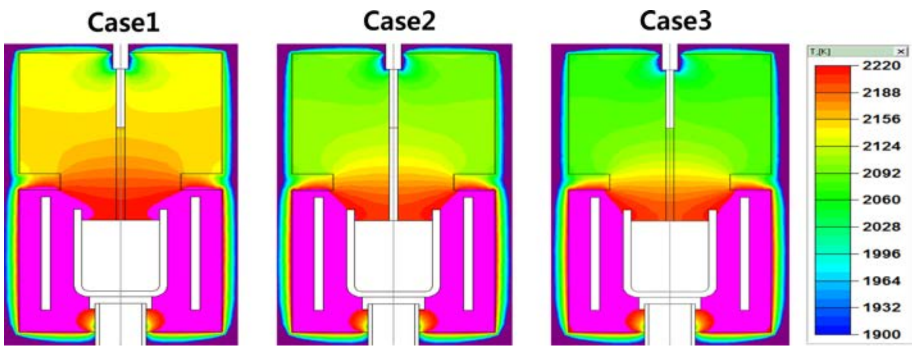


Fig. 11. Sensitivity analysis: thermal conductivity changes of carbon insulation.

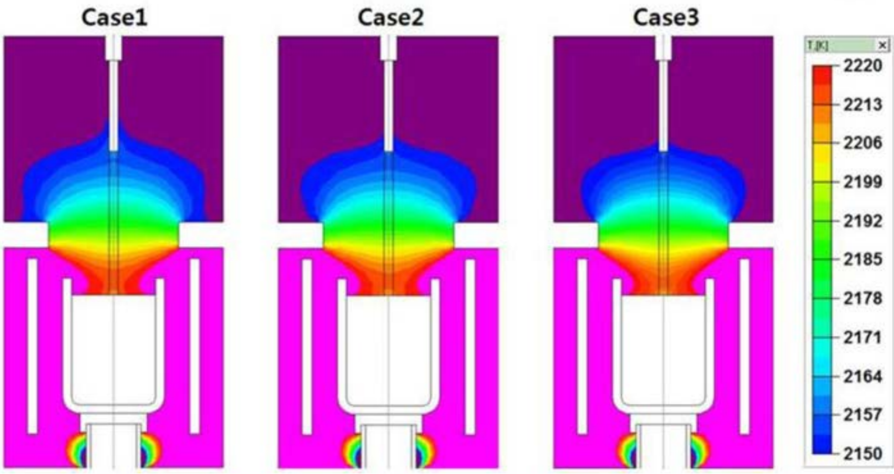


Fig. 12. Sensitivity analysis: thermal emissivity changes of carbon insulation.

Table 4. Power consumption changes by changing carbon emissivity.

	Case1	Case2	Case3
Heat conductivity W/(m*K)	0.5	0.5	0.5
Emissivity	0.42	0.66	0.8
Heat capacity J/(kg*K)	2100	2100	2100
Power consumption (kW)	6.91	7.08	7.18

power consumption has high sensisivity in heat coductivity. Fig. 11 show that less energy loss and more broad temperature distribution in low heat conductivity carbon insulation. By comparing with induction heating 7.91 W and resistance heating 7.08 W, resistance heating is not inferior and advantages in large growing systems. In addition to heat conductivity, thermal emissivity is another key parameter to reduce power consumption. Table 4. Show the change of thermal emissivity from 0.42 to 0.8, and the power comsumption results are changed from 6.91W to 7.18 W. As less change in power consumption, temperature distribution in each cases has hard to distingusih in Fig. 12. It means that power consumption has more dependency in heat conductivity rather than thermal emissivity.

Future work

We are in the process of continuing YAG single crystal growth studies to follow the same path as silicon and sapphire industry with cost becoming the main driver. Sensitivity analysis for key parameters that help to understand and control of the single crystal growth. Narrow the gap between simulation and actual growth by optimizing hot zone design and its parameters, and find the scaling factors and apply in the large size YAG growth.

Conclusions

The hot zone design is crucial for the good yield and quality, productivity improvement, and large size YAG crystals, which are shortfalls of current induction heating Czochralski method. We also have findings of follows. Understanding the importance of temperature gradient in Melt and Melt-Crystal Interface, growing zone. Resistance heating has stable temperature gradient in Melt and higher temperature gradient in Melt-Crystal Interface, which can be controlled by hot zone design change. Power consumption also be improved by using lower thermal conductivity carbon insulation material

The present paper investigates five cases of design variation by numerical simulation with respect to temperature gradient, convexity, and melt velocity. Case 4 and 5 are found to fulfill the requirments of these objectivs best. This can be achieved by combination of reflector and two zone heaters.

Acknowlegments

This work was supported by the Korea Research Foundation (KRF) grant funded by Korea Goverment (MEST) (No. 10047571).

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