Thermal lag of furnace when growing on a seed large alkali halide crystals from melt

O.Ts. Sidletskiy◊, V.I. Goriletsky, M.M. Tymoshenko, V.I. Sumin, O.V. Sizov, B.V. Grinyov
Institute for Scintillation Materials of NAS of Ukraine
60, Lenin Pr., Kharkiv, 61001, Ukraine
E-mail: sidletskiy@isc.kharkov.com

Abstract
Influence of the bottom and side heaters on thermal field of a “ROST” type growth furnace and its thermal lag has been studied experimentally using an infrared thermometer. Contribution of the bottom heater is decisive for temperature field formation in whole growth furnace excluding periphery circular vessel of the crucible where the side heater plays the main role. It has been determined that in response to heater temperature variation the whole crystal surface heats (cools) almost uniformly and simultaneously: for 13 – 16 min when bottom heater temperature \( t_{bot} \) is being changed and 8 – 11 min in the case of side heater temperature \( t_{side} \) change. Possibility of automated growth control system improvement accounting for the data obtained is discussed. It is shown that the preferred parameter for its utilization in the additional circuit of control system based on the program-controlled \( t_{side} \) correction is periphery circular vessel bottom temperature; such scheme must be sensitive enough to changes in crystallization front shape allowing one to improve speed of response of the control system and to avoid crystallization front deformations due to untimely \( t_{side} \) corrections.

Key Words: Alkali halide crystal growth, furnace heaters, IR-pyrometry, temperature field, thermal lag.

◊ Author for correspondence
1. Introduction

Temperature conditions inside the furnace are the decisive factor at growing on a seed large alkali halide crystals from melt. Deviation of temperature from the preset values manifests in disturbances of crystallization front (CF) shape that worsen structure perfection and physical properties of growing ingots and, in particular, can lead to non-uniform activator distribution in crystal and worsening of scintillation characteristics of alkali halide crystals.

Thermal field in “ROST” type setups is formed by the bottom and side heaters, and water-cooled furnace walls (Fig. 1). Existing automated growth control system is based on bottom heater temperature \( t_{\text{bot}} \) programmed correction using signals from the melt level probe. Automated semicontinuous method (ASC) [1] provides a constancy of melt level in the crucible due to the feeding by a raw material from the hopper. A raw material melts in peripheral circular vessel and then run off into the crucible. Thus, side heater in current scheme is used only for melting of a raw material (melting temperature of CsI is 621°C). Substantial lack of the existing control system using the melt level probe is a big response time to occurring changes of temperature conditions, so, bottom heater often receives control signal too late to react on changes adequately.

![Fig. 1. Scheme of the “ROST” type setup: 1 – removable quartz windows; 2 – pyrometer; 3 – growing crystal; 4 – melt; 5 – periphery circular vessel; 6 – seed; 7 – bottom heater; 8 – side heater. Measurement points: A – side crystal surface (30 mm above the melt), B – side crystal surface (100 mm above the melt); C – crystal upper butt (on diameter 130 mm), D – crucible bottom; E – wall of the periphery circular vessel (5 mm from the upper edge); F – bottom of the periphery circular vessel.](image-url)
At the same time, it was established [2] that at growth of crystals with maximal dimensions, of diameter close to the crucible one, side heater influences not only periphery circular vessel (PCV) temperature, but temperature of surface layer of melt and side surface of the ingot too. In return, growing crystal gradually overheats the PCV requiring compensation by a $t_{\text{side}}$ decrease. Thus, untimely program-controlled $t_{\text{side}}$ corrections lead to disturbances in CF shape and worsening of ingot structural properties. To avoid such undesirable effects, a conception of two-circuit growth control system based on $t_{\text{side}}$ adjustment based on signals from the IR-sensor measuring PCV bottom temperature was proposed [3].

To realize the two-circuit system on practice, it is necessary to study temperature field on the whole crystal surface and crucible, and determine the contribution of $t_{\text{bot}}$ and $t_{\text{side}}$ into thermal field inside the furnace.

This work aimed at studies of thermal conditions both on the side crystal surface of growing ingot and crucible surface, as well as determination of thermal lag of the furnace to changes of bottom and side heater temperatures. Dynamical temperature characteristics of the side crystal surface and crucible elements on the different growth stages have been measured, as well as response in different points of the system to heater temperature changes have been determined using IR-pyrometry.

2. Experimental

Utilization of infrared thermometers in temperature measurements inside a “ROST” type growth furnace is, probably, the only available tool in the situation when crucible and growing crystals continuously rotate. IR-thermometry (pyrometry) based on monitoring of thermal radiation emitted by heated bodies [4, 5], unlike thermocouple measurements, do not distort temperature field, that is very important in the system where temperature gradients reach tens degrees on centimeter. In [6] we developed a procedure of non-contact pyrometric measurements
of temperature on the crystal surface and crucible elements, which was used in determination of upper crystal butt temperature on the different growth stages.

Measurements were carried out using a Raytek Marathon RAYMA2SC IR-thermometer, allowing one to determine temperature in the range 350 – 2000 °C with reproducibility not worse than 0.01% from the measured value. Pyrometer working wavelength is 1.6 µm. Measured data was being transferred to PC and processed using Raytek Datatemp Multidrop software. Sighting by the pyrometer were conducted through the quartz windows on the upper and bottom walls of the furnace (see Fig. 1).

3. Results and Discussion

Temperature distribution on the side ingot surface is shown in Fig. 2, where dependence of temperature vs. crystal height is presented for Kouropoulos growth of CsI(Na) single crystal of 270 mm in diameter. Temperature distribution on the side surface of crystal, which is completely situated inside the crucible, is linear (curves 1 – 3). As the crystal grows, temperature gradient monotonously decreases from 20 to 8 °C/cm. When the upper butt of the growing ingot leaves the crucible limits, the gradient on the side surface above the crucible upper edge sharply increases up 13 °C/cm due to increase of radiative heat exchange between the side crystal surface and water-cooled furnace walls. Abscissa of the breakpoint between the linear regions of the curve 4 corresponds to the crucible upper edge. It is known that this growth
stage is most dangerous in the context of probability of structure defects formation in crystals. Simultaneous sharp changes in temperature distribution on the upper crystal butt were observed as well in [6]. This thermal field picture is qualitatively similar to the observed temperature distribution at ASC\textsuperscript{1} growth.

Dynamical temperature characteristic of the fragment of the crystal side surface (see Fig.1, point A) is shown in Fig. 3 (curve 1). The temperature increases till the moment when crystal upper butt leaves the crucible limits, and, then, stabilizes. The lack of direct measurements of the crystal surface is a big dispersion of temperature values connected with non-uniformity of condensate layer forming on crystal due to melt evaporation [6]. Variation of temperature on the wall between the crucible and periphery vessel (Fig. 1, point E, and Fig. 3, curve 2) is qualitatively similar to the curve 1, but the observed dispersion of temperature values is substantially smaller. Another advantage of measurements on the wall is that monitoring can be carried out from the seeding moment – both on radial and axial growth stages.

Dynamics of temperature changes on the PCV bottom (Fig.1, point F and Fig. 3, curve 3) differs from the two first cases. The temperature decreases on radial growth stage and stabilizes

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\textsuperscript{1} ASC method is a modification of Kouropoulos growth with elements of Czochralsky method.
on the axial growth – it is even more stable in compare to the curve 2, small fluctuations from the constant value correspond to variations of the crystal diameter from the preset value. Thus, really, holding a temperature of the PCV bottom we can maintain a stable CF shape and, consequently, constant diameter of the growing ingot.

To estimate a speed of response of the existing ($t_{bot}$ correction) and proposed (correction of both $t_{bot}$ and $t_{side}$) automated control systems, an analysis of thermal lag of the growth furnace in response to the heater temperatures variation was carried out. Measurements were conducted in 6 points (see Fig. 1) using a CsI(Na) crystal of 270 mm in dia. and cylindrical part length 120 mm. Temperature was being changed by 2 °C at bottom heater and by 4°C at less powerful side heater. As example, the dependence of temperature vs. time for upper crystal butt (point C in Fig. 1) when varying side heater temperature is shown in fig. 4.

Results of the obtained curves processing are summarized in the Table I. One can see that under $t_{bot}$ changes the crucible heats (cools) first (6 – 7 min), and, then, temperature stabilizes almost simultaneously on the whole crystal surface (13 – 15 min). This effect is caused by the fact that alkali halide crystals possess not only rather large thermal conductivity ($1.1 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$), but a very low thermal radiation absorption coefficient as well ($\alpha = 10^{-3} – 10^{-4} \text{ cm}^{-1}$) [8], ensuring fast warming-up (cooling down) of the whole crystal due to radiative heat transfer.

In response to $t_{side}$ variation, like it was predicted earlier, the PCV bottom (Fig.1, point F) warms-up first (4 min.) and, then, temperature stabilizes on the crystal surface (9 – 11 min.) and
crucible bottom (point D) – in fact, it means a melt temperature stabilization. It is worth to note that response time of the crystal surface (points A – C) to the bottom heater changes (13 – 16 min) is rather big in compare to the side heater (8 – 11 min).

Table I. Thermal lag of crystal and crucible elements at variation of heater temperatures: the first values – temperature change in the point (°C), the second values – time of temperature stabilization (minutes).

<table>
<thead>
<tr>
<th>Points on the Fig. 1</th>
<th>At $t_{\text{bot}}$ variation (±2°C)</th>
<th>At $t_{\text{side}}$ variation (±4°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>1.1; 13.5</td>
<td>1.1; 8.8</td>
</tr>
<tr>
<td>B</td>
<td>0.9; 16.5</td>
<td>1.3; 9.0</td>
</tr>
<tr>
<td>C</td>
<td>0.9; 14.0</td>
<td>1.1; 10.5</td>
</tr>
<tr>
<td>D</td>
<td>1.0; 6.0</td>
<td>1.1; 11.3</td>
</tr>
<tr>
<td>E</td>
<td>0.8; 16.0</td>
<td>2.3; 9.0</td>
</tr>
<tr>
<td>F</td>
<td>0.4; 7.0</td>
<td>2.5; 4.0</td>
</tr>
</tbody>
</table>

Concerning changes in temperature, one can see (Table I) that the system, like it was provided by the construction, responds more intensively to changes in temperature of the more powerful bottom heater, however, $t_{\text{side}}$ contribution is rather big on the PCV wall (point E) and side ingot surface (points A, B). The exclusion is the PCV bottom (point F), where $t_{\text{side}}$ contribution is weightier, and $t_{\text{bot}}$ influence is weak.

Thus, the acceptable objects for temperature monitoring with IR-sensor aimed at $t_{\text{side}}$ controlled regulation in the additional circuit of the automated growth system is the PCV bottom and the wall between it and the crucible. Accounting for the fact that the PCV wall, like crystal surface, is covered with condensate layer due to the melt evaporation, the scheme utilizing the PCV bottom temperature seems more reliable providing a high sensitivity to changes of temperature conditions near the crystallization front.

Conclusions

1. It has been shown experimentally that contribution of the bottom heater is decisive for temperature field formation in whole growth furnace excluding PCV of the crucible. Also, it has
been confirmed that side heater not only plays the main role in formation of temperature field in
the PCV, but substantially influences temperature of the PCV wall and crystal surface.

2. The whole crystal surface at heaters temperature variation heats up (cools down) almost
uniformly and simultaneously: for 13 – 16 min when \( t_{bot} \) is being changed and 8 – 11 min in the
case of \( t_{side} \) change.

3. Direct measurements of crystal surface temperature aimed at modernization of automated
growth control system is not a reliable way because of non-uniformity of condensate layer
thickness deposited on the crystal leading to drop of temperature measurements precision. The
preferred parameter for its utilization in the additional circuit of \( t_{side} \) program-controlled
regulation is temperature on the PCV bottom: the proposed scheme will be sensitive enough to
changes in CF shape allowing one to improve speed of response of the control system and avoid
CF deformations due to untimely program-controlled \( t_{side} \) corrections.

References

2. V.I. Goriletsky, B.V. Grinyov, V.S. Suzdal et al. // Proc. of XIV Intern. Conf. on Crystal
3. V.I. Goriletsky, B.V. Grinyov, V.S. Suzdal et al. Patent of Ukraine No. 71835A. Published
15.12.04, bull. № 12.
4. Transactions in measurement and control. Vol. 1. Non-contact temperature measurements
press).
(editor), Marcel Dekker, 1991).