DISTRIBUTION OF TEMPERATURE IN SEMI-TRANSPARENT SINGLE CRYSTALS DURING THE PROCESS OF THE CZOCHRALSKI PULLING. II. ANALYSIS OF THERMAL FIELDS

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Determination of thermal fields in single crystals pulled by the Czochralski technique is enabled by the utilization of analytical or numerical calculation techniques. Comparison of the results obtained with the help of either of these techniques shows that numerical computations result in far less steep thermal gradients. Semi-transparency of crystals contributes to an increase of drops of temperature. In the paper, some methods of modulating thermal fields in single crystals, as well as some techniques of measuring temperature at side surfaces of crystals and at their geometrical axis are also discussed.

1. Principal methods of analysis of thermal fields of single crystals

The problem of determining the distribution of the thermal field of a single crystal growing by the Czochralski method has been studied by many scientists. The initial research was performed by Billig already in 1956, i.e. shortly after the first successful production application of the Czochralski technique. In the late sixties this research became especially intensive, which gives evidence of the importance of the problem. Indeed with the temperature gradients in a crystal, thermoelastic stresses appear and also other defects, i.e. dislocation, block structure etc.

This leads to deterioration of the quality of a crystal and in extreme cases may result in its destruction.

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The so far existing approaches to this problem made by several authors may be roughly divided into two groups: the first group wouldc over the attempts to determine the temperature distribution in the crystal analytically, the other — numerical computations. In analytic solutions the boundary condition on the side surface of the crystal is assumed linear in form; it is the only possibility of making it independent of the z coordinate directed perpendicularly to the front of crystallization. In the calculations of the author [1] convection in the protective atmosphere is not taken into account, which should not result in a large error for materials showing high melting points. According to Reed [2], convection of gases at the 1 atm pressure, occuring during crystallization of the material with the melting point 2000°C results in a flux of heat comparable to black body emission at the temperature of 800°C. Some analytic solutions allow for convenction by describing it mathematically in the form of an element proportional to the difference of temperatures within the accuracy of the constant E_k. Brice [3] assumed

$$E_k = 0.548 \left\{ \frac{g \varrho c \kappa^3 (T_R - T_0)}{\eta L T_0} \right\}^{1/4}.$$ (1)

Here T_R — temperature of side surface of the crystal, T_0 — temperature of the crystal environment, L — specific heat of solidification, ϱ — density, η — viscosity of the melt, κ — conffient of thermal conductivity, c — specific thermal capacity.

The analytic solutions are approximate since some mathematical formulations are subjected to necessary simplifications and due to the forms of definitions of certain parameters, e.g. T_{av} [1]. The following formulae determining this parameter are possible:

$$T_{av} = \frac{T_t + T_0}{2},$$ (2)

or [4]

$$4T_{av}^3 = (T_t^4 - T_0^4)/(T_t - T_0),$$ (3)

or

$$T_{av}^4 = 5T_0^3 T_t - 3T_0^4,$$ (4)

where T_t is melting point of the material.

Many authors assume that $T = T_t/2$. The heat transfer at the side surface depends on the chosen value of T_{av} and generally does not correspond with the realistic situation: it is smaller than the real value close to the front of crystallization where $T_R > T_{av}$, and greater in the parts of crystal distant from the interface ($T_R < T_{av}$).

The analytical solutions yield results similar conceptually: the axial distributions of temperature are changing as sinh or cosh, whereas the radial distributions are determined by a series of Bessel functions of the first type and of the zeroth order.

A new approach has been presented by Etori [5] who looked for the solutions based on the analogy between the thermal system and corresponding electrical circuit.

The numerical method provides better accuracy. First of all it enables us to take into account the dependence of crystals on temperature, which is specially important

for the heat conduction coefficient [6]. Also, linearization of the boundary condition at the side surface of a crystal for the entire specimen is not necessary. Sakharov and his coworkers [7] divided a crystal conceptually into thin slices perpendicular to the axis of a boule; for every slice a uniform temperature along the z axis was assumed and separate boundary conditions formulated. However, neither they nor Akiyama et al. [6] have taken convection into account: their calculations regarded only the process performed in vacuum; this improvement has been made by Arizumi and Kobayashi [8]. In Fig. 1 and 2 the temprature distributions taken from the paper od Sakharov and others are
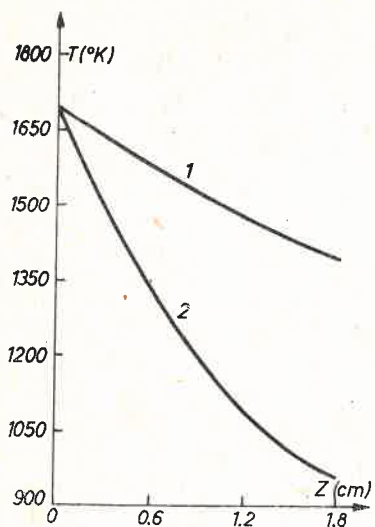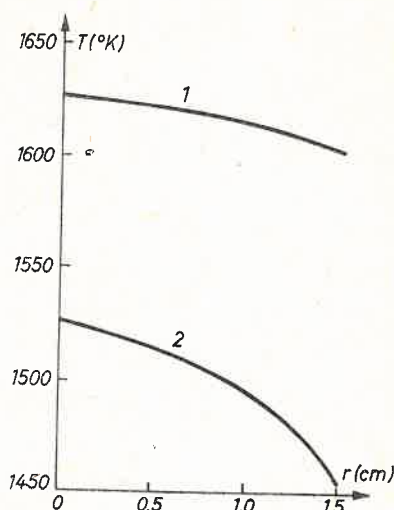

Fig. 1

Fig. 2

Fig. 1. Comparison of axial temperature distributions in silicon single crystal computed numerically (curve 1) by Sakharov and others, and analytically (curve 2) by Billig
Fig. 2. Comparison of radial temperature distributions in silicon single crystal computed numerically (curve 1) by Sakharov and others, and analytically (curve 2) by Billig

presented. These distributions are compared with the ones calculated by Billig [9]. Both works consider silicon single crystals and do not allow for volumetric emission of this material. We see that in the result of numerical calculations much smaller thermal gradients are obtained.

At present it would be difficult to judge which one of the computational methods presented is better. These doubts are probably common, which is proved by the increase of number of works taking into account both computational techniques.

2. Thermal fields of semi-transparent materials

Making use of the formula for temperature distribution in a semi-transparent material [1], the thermal field for crystals of yttrium-aluminum garnet $Y_3Al^5O_{12}$ and lithium niobate $LiNbO_3$ has been calculated for three cases:

(a) boule of 2 cm diameter and 5 cm length pulling with afterheating up ($T_0 = 1270°$K in case of YAG and $1070°$K in case of $LiNbO_3$);

(b) boule of 1 cm diameter and 5 cm length pulling with afterheating ($T_0 = 1270°$K in case of YAG and 1070 for $LiNbO_3$);

(c) crystal of 2 cm diameter and 5 cm length pulling without afterheater ($T_0 = 300°$K for both YAG and $LiNbO_3$).

The results are given in Tables I and II. Simultaneously the temperature distributions for the same materials are computed without taking into account volumetric radiation of energy Tables I and II. Axial drops in temperatures are calculated along the geometrical axis of the crystal pulling in the direction [100] in the case of YAG and [0001] in the case of $LiNbO_3$.

TABLE I

Axial distributions of temperature in single crystals $Y_3Al_5O_{12}$ ($T°$K)

z (cm)	Non-transparent crystal			Semi-transparent crystal			Calculations according to Troitsky		
	a	b	c	a	b	c	a	b	c
0.0	1270	1270	300	1270	1270	300	1270	1270	300
0.5	1298	1309	389	1306	1280	386	1310	1285	382
1.0	1343	1359	499	1330	1302	489	1353	1306	482
1.5	1405	1418	612	1396	1335	608	1404	1333	598
2.0	1480	1490	763	1446	1379	726	1466	1376	733
2.5	1567	1562	914	1523	1439	886	1541	1435	895
3.0	1662	1652	1081	1616	1519	1104	1633	1508	1071
3.5	1765	1746	1367	1731	1629	1202	1745	1614	1289
4.0	1894	1862	1591	1863	1780	1422	1872	1760	1457
4.5	2054	1996	1857	2013	1967	1843	2027	1960	1833
5.0	2242	2262	2242	2242	2250	2241	2246	2242	2250

TABLE II

Axial distributions of temperature in single crystals $LiNbO_3$ ($T°$K)

z (cm)	Non-transparent crystal			Semi-transparent crystal			Calculations according to Troitsky		
	a	b	c	a	b	c	a	b	c
0.0	1070	1070	300	1070	1070	300	1070	1070	300
0.5	1089	1084	363	1084	1082	336	1083	1079	358
1,0	1129	1102	447	1108	1096	425	1103	1090	429
1.5	1134	1124	538	1126	1109	525	1123	1103	510
2.0	1164	1155	640	1152	1120	625	1148	1121	602
2.5	1200	1189	754	1175	1150	737	1181	1146	708
3.0	1244	1228	880	1205	1185	890	1216	1181	829
3.5	1297	1281	1027	1293	1234	984	1275	1232	970
4.0	1362	1349	1100	1357	1304	1122	1343	1303	1134
4.5	1441	1442	1348	1436	1405	1328	1429	1404	1322
5.0	1532	1534	1532	1531	1542	1530	1531	1536	1531

All this calculations are based on the earlier work of the same authors [1]. The radial distributions refer to the surface $z = 4.5$, i.e. such drops in temperature occur in a solid phase distant from the front of crystallization by about 5 mm. These results are presented in Tables III and IV.

TABLE III

Radial distributions of temperature in single crystals $Y_3Al_5O_{12}$ $(T°K)$

r (cm)	Non-transparent crystal			Semi-transparent crystal			Calculations according to Troitsky		
	a	b	c	a	b	c	a	b	c
0.0	2065	2059	2063	2055	1992	1993	2054	1995	1988
0.1	2065	2058	2062	2055	1992	1993	2054	1995	1987
0.2	2065	2056	2061	2054	1989	1992	2053	1993	1986
0.3	2064	2052	2059	2053	1986	1991	2052	1989	1984
0.4	2063	2047	2056	2050	1980	1990	2050	1983	1981
0.5	2061	2041	2053	2048	1873	1987	2047	1973	1978
0.6	2059		2049	2044		1984	2043		1974
0.7	2056		2045	2040		1980	2039		1969
0.8	2053		2039	2034		1976	2033		1963
0.9	2048		2032	2028		1970	2028		1955
1.0	2042		2024	2020		1964	2019		1944

TABLE IV

Radial distributions of temperature in single crystals $LiNbO_3$ $(T°K)$

r (cm)	Non-transparent crystal			Semi-transparent crystal			Calculations according to Troitsky		
	a	b	c	a	b	c	a	b	c
0.0	1436	1405	1368	1408	1405	1389	1432	1406	1344
0.1	1436	1404	1368	1407	1404	1388	1431	1405	1343
0.2	1436	1403	1368	1407	1403	1387	1431	1404	1342
0.3	1435	1401	1367	1406	1401	1386	1429	1401	1340
0.4	1434	1398	1366	1405	1398	1384	1428	1397	1338
0.5	1432	1394	1364	1404	1394	1382	1426	1391	1335
0.6	1431		1362	1402		1379	1423		1332
0,7	1428		1360	1399		1375	1420		1328
0.8	1425		1357	1396		1371	1416		1322
0.9	1422		1353	1392		1366	1411		1317
1.0	1418		1348	1387		1360	1405		1310

The conclusions drawn from these calculations are clear to every worker involved in growing single ctystals by the Czochralski method. They are mentioned here as a confirmation of correctness of final calculation formulae. So:

— heating up of the crystal considerably diminishes axial thermal gradients and partly radial gradients;

— axial drops in temperature decreases with increase in crystal diameter;

— crystal diameter influences also thermal gradients in the radial direction. Namely, in large boules temperature differences are greater close to the edges, and in the inside area, close to the geometrical axis of the crystal, they are subjected to considerable softening.

In the computations performed in [1], only the first five elements of the series (15) are taken into account. Precisely, this is only an approximation though relatively good. In Table V distribution of temperature at the front of cristallization $z = z_0$ is given.

TABLE V

Distributions of temperature at the interface in $Y_3Al_5O_{12}$ and $LiNbO_3$ single crystals ($T°K$)

r (cm)	$Y_3Al_5O_{12}$			$LiNbO_3$		
	a	b	c	a	b	c
0.0	2242	2242	2242	1531	1542	1530
0.1	2241	2241	2241	1530	1541	1530
0.2	2238	2241	2240	1529	1541	1529
0.3	2239	2241	2240	1529	1541	1529
0.4	2240	2241	2241	1530	1542	1529
0.5	2240	2238	2241	1530	1540	1529
0.6	2239		2239	1529		1529
0.7	2238		2240	1529		1529
0.8	2241		2241	1530		1529
0.9	2241		2242	1531		1530
1.0	2235		2237	1527		1527

Deviations from the melting point are here not greater than 0.5% of the nominal value. When the axial gradients of temperature occuring close to the interface is about 350°C/cm, this gives a maximum deviation in the shape of crystallization front from the assumed flat surface of about 0.3 mm, which can be neglected in practice.

3. Practical techniques of measurement and control of heat fluxes

According to the above calculations the problem of control of heat fluxes in a growing single crystals is especially important.

One of the basic ways of modulating these fluxes is achieved by means of afterheaters. Some technical designs are shown in Fig. 3. The important thing that is to be remembered is fitting the maximum crystal diameter to the diameter of a crucible, since the dimensions of the latter considerably influence the thermal field in the vicinity of the solid physe.

Temperature measurement in a growing single crystal is rather cumbersome. Several techniques have been applied (Fig. 4). One is the placement of a thermocouple in the crystal near the axis of the boule [10]; in order to perform this, a cavity is made. This technique enables temperature measurements close to the front of crystallization, it provides, however, some inconveniences and limitations: it gives the distribution of the temperature in the

axis of the boule only. Finally it causes difficulty in turning the crystal and introduces considerable perturbations to the thermal field. The measurement of temperature of the crystal side surface by means of a pyrometer is less accurate. The measurement should be performed discretely as is shown in Fig. 4b. For semi-transparent crystals these two
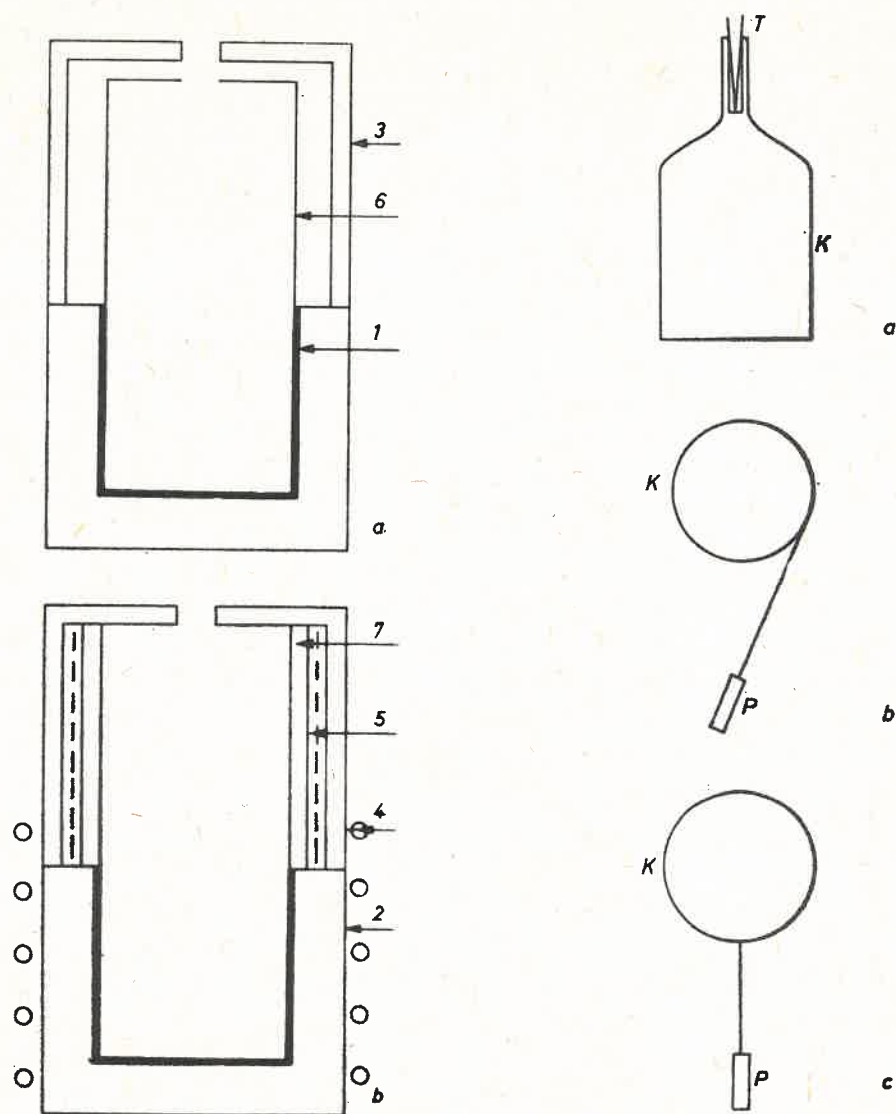

Fig. 3

Fig. 4

Fig. 3. Types of afterheaters used for pulling single crystals of oxide materials: inductive afterheater (a) and resistance one (b). Notation: *1* — crucible, *2* — ceramic "nest" of crucible, *3* — ceramic shield, *4* — r.f. coil, *5* — resistance afterheater, *6* — induction afterheater, *7* — internal ceramic shield

Fig. 4. Techniques of temperature measurement of a growing single crystal. Notation: K — Crystal, T — thermocouple, P — pyrometer

techniques may contribute to give the desired effect. The technique of measurement presented in Fig. 4c is not reliable since the result refers to the value of temperature averaged with respect to the crystal radius.

4. Temperature distributions in semi-transparent crystals according to Troitsky

Temperature distributions in semi-transparent single crystals have been studied by Troitsky [11]. He analyzed the exchange of thermal energy between adjoining volumetric elements of a crystal. He showed that in a semi-transparent medium characterized by an absorption coefficient μ and a refraction coefficient n, the radiational heat flux is proportional to $T_R - T_0$ with the proportionality coefficient K equal to

$$K = \frac{16 n^2 \sigma T^3}{3\mu}, \tag{5}$$

where T_R — temperature of crystal side surface; T_0 — ambient temperature; T_{av} — average temperature of crystal side surface; σ — Stefan–Boltzmann constant equals $5672 \cdot 10^{-5}$ erg/cm^{2}deg^4.

In Tables I—IV the thermal fields for single crystals of yttrium-aluminum garnet and lithium niobate calculated from the formula developed by Troitsky are given. The results do not differ considerably from the results of this paper.

5. Conclusions

(a) Thermal field in semi-transparent single crystals is characterized by much greater temperature differences that in the case of non-transparent crystals. It is due to the additional flux of thermal energy radiated by the internal regions of crystals to the environment.

(b) Temperature distribution in a crystal depends considerably on such factors as afterheating of the solid phase, radius of a crystal and its absorption coefficient.

(c) Calculations of temperature distribution performed in the present work are consistent (within the limits of error) with calculations made by other authors.

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