Investigation of defects formation on different stages of sapphire crystals growth

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Abstract. The mathematical simulations and calculations allow to estimate the large number of factors influence on crystals growth and quality and to determine the main cause of the defect structures formation in the solid phase. The research results allow making the quantity estimation of defects formation in sapphire crystals. The simulation results can help to define defect formation zones and can be used for recommendations about crystals quality improvement.

1. Introduction
The SOI technology is one of the most progressive manufacturing techniques of inertial MEMS-sensors [1]. The various poly- and monocrystal materials can be applied as a dielectric. The sapphire crystal is one of perspective materials especially for MEMS in which sensitive elements work in the gigahertz ranges. The quality of all MEMS-sensor operation depends on quality and a minimum of such crystal superficial defects. So the problem of defects formation on different stages of sapphire crystals growth is actual one now.

The big size sapphire crystals growth with improved structural characteristics became a priority of modern science and equipment. In the last decades the Czochralsky’s method, Kiropoulos’s method, the horizontal directed crystallization (HDC) method and Stepanov’s method were applied for big size monocrystals growth. These crystals were widely used in electronic technique [2].

The sapphire crystal quality can be characterized by the orientation, density and the nature of dot defects distribution, density of single dislocations, extension of blocks borders and their disorientation, the value of residual tension, chemical purity and impurity heterogeneity [2-4].

2. Mathematical simulation of defects formation during sapphire crystals growth
The HDC method has some advantages in comparison with other methods. First of all it is because the melt height and its square are constant during all process. The large melt volume allows to provide the effective impurities evaporation. The fuse process and the border of phase division can be controlled visually or by means of optical devices.

The bubbles formation in the melt and their capture by the crystallization front are one of the problems during sapphire crystals growth by the HDC method. These bubbles can be the cause of pores emergence in crystal which being tension concentrators are an additional source of dislocations and, as a result, a source of crystal defects in the form of blocks.
It is noticed [5,6] that at the different stages of crystal growth the position and form of bubbles congestions are various. The largest problem for bubbles congestions removal is made by the bubbles located in the benthonic zone of sapphire crystal growth furnace SZVN 155.320 crucible beginning, approximately, at the distance 250 mm from the fuse with the container length 320 mm. It is shown schematically in Figure 1. It is also shown in the photographs of crystals taken at lighting, presented in Figure 2.

**Figure 1.** The scheme of position of the crystallization front and bubbles congestion at various stages of growth (the arrow showed the direction of crucible movement)

**Figure 2.** Inclusions in sapphire crystals: a) at the distance 60 mm from the fuse, b) at the distance 250 mm from the fuse

In the case of horizontal directed crystallization, the projection of Archimedean velocity on the direction of front movement is zero, and the direction of the temperature gradient in the melt $\nabla T$ coincides with the direction of growth front velocity.

Considering that the thermocapillary force in the stationary mode [7,8]

$$F_k = -2\pi \cdot r^2 \cdot \frac{\partial \alpha}{\partial T} \cdot \nabla T,$$

where $\nabla T$ is the direction of the temperature gradient in the melt; $r$ is the radius of gas bubble; $\alpha$ is the surface tension between gas and liquid boundary must be equal to the force of the medium viscous resistance [9]

$$F_c = 6\pi \cdot r \cdot \mu \cdot V_k.$$

The rate of bubble thermocapillary movement relatively to the liquid can be determined by the formula

$$V_k = \frac{r}{3 \cdot \mu} \cdot \frac{\partial \alpha}{\partial T} \cdot \nabla T,$$
where $\mu$ is the dynamic viscosity of liquid. It is also considered that for the majority of liquids the derivative $\partial \alpha / \partial T < 0$. At the rate $V_a = 0$ the velocity of the bubble relatively the growth front is defined by equality

$$V_p = r \frac{\partial k}{\partial T} \cdot \nabla T - V_f.$$  \hspace{1cm} (4)

As we can see from (4) in the bubbles ensemble in the melt there are bubbles of critical size $r_k$ at which the bubble motion direction relatively the crystal growth surface is changed. Gas bubbles with size

$$r > r^* = \frac{V_f}{\sqrt{T}} \frac{\partial \alpha / \partial T}{r}$$  \hspace{1cm} (5)

will move in the direction from the front, and smaller bubbles $r < r^*$ will move in the opposite direction.

Bubbles with radius $r < r^*$ appeared in surface of phase transformation during crystallization will be grasped by the growth front. So they create system of steady defects in material volume.

It is necessary to calculate the temperature distribution in sapphire crystals growth equipment by the HDC method for defects diagnostic. The temperature distribution determination in crystal-melt-crystal powder system consists of heat conductivity equations solution [10]:

$$\frac{\partial T_i}{\partial \tau} = \left( \frac{\partial}{\partial x} a_i \frac{\partial T_i}{\partial x} + \frac{\partial}{\partial y} a_i \frac{\partial T_i}{\partial y} + \frac{\partial}{\partial z} a_i \frac{\partial T_i}{\partial z} \right) - W \frac{\partial T_i}{\partial x},$$  \hspace{1cm} (6)

where $i = 1, 2, 3$ is the crystal, melt and crystal powder accordingly; $a_i$ is the heat diffusivity coefficients ($a_i = \frac{\lambda_i}{\rho_i C_i}$), where $\lambda_i$ is the thermal conductivity coefficient; $\rho_i$ is the material density; $C_i$ is the specific heat); $W$ is the container movement speed.

The container movement speed is small enough, so we can conclude that the process is in quasisteady state and the temperature distribution can be found by the following formula [11]

$$\text{div} (\lambda_i \text{grad} T_i(x, y, z)) = 0.$$  \hspace{1cm} (7)

The boundary conditions for system of the equations (7), reflecting continuity of thermal fields and streams on limits of environments section, can be found by the following ratios:

$$\lambda_1 \frac{\partial T_1(x_T, y, z)}{\partial x} = \lambda_2 \frac{\partial T_2(x_T, y, z)}{\partial x},$$  \hspace{1cm} (8)

$$\lambda_2 \frac{\partial T_2(x_T + \Delta x, y, z)}{\partial x} = \lambda_3 \frac{\partial T_3(x_T + \Delta x, y, z)}{\partial x},$$  \hspace{1cm} (9)

$$q_{s1} = q_{s2} = q_{s3} = \sigma \beta (T^4 - T_{hot}^4),$$  \hspace{1cm} (10)

where $\sigma$ is the Stephane-Boltsmana constant; $\beta$ is the radiation coefficient; $T_{hot}$ is the function with the heat temperature distribution.

The final volume method on the unstructured grid was used for simulation. The temperature fields in crystal-liquid-powder system are showed in Figure 2, a. The temperature fields in vertical crystal cut are presented in Figure 2, b.
The temperatures gradients allow to estimate the sizes of forming vapour bubbles. So it is known that the crystal growth rate influence on the capture of impurity and foreign inclusions by the front of crystallization. At bubble capture the ratio of crystal growth and bubble rates defines a final form of inclusion. The received dependences of the bubbles average size from the growth speed can be explained [10]. At the set crystallization speed the bubbles with certain radius can be characterized by the greatest probability of capture. The increase in growth rate will lead to increase the probability of larger bubbles capture.

The dependence of segregation effective coefficient on the interface of phases from growth rate can be determined [2,5,10] as

$$K_{\text{eff}} = \frac{K_0}{K_0 + (1 - K_0) e^{\frac{V \delta}{D}}}, \quad (11)$$

where $K_0$ is the equilibrium segregation coefficient; $\delta$ is the depth of the area with raised or lowered concentration of impurity at the crystallization front; $D$ is the diffusion coefficient of impurity in melt; $V$ is the crystal growth rate. At the intensive interfusion $\delta \to 0$ or at the small crystallization speed $V \to 0$, $K_{\text{eff}} \to K_0$ the impurities and bubbles are taking away in melt. When the growth speed $V \gg \frac{D}{\delta} (e^{\frac{V \delta}{D}} \to 0)$, $K_{\text{eff}} = 1$ it means that all bubbles and impurity are taken by crystal.

The growth rate depends on position of the container relatively the heater and increases throughout the process of sapphire crystals growth in the conditions of large temperature gradients [5]. The effective coefficient dependence at the various stages of growth is shown in Figure 3.

Figure 2. Temperature distribution in crystal-liquid-powder system
Figure 3. The effective coefficient of segregation depending on the crucible position: 1 – the gradient in melt is 1000 K/m, 2 – the gradient in melt is 500K/m

From Figure 3 it is visible that only at the initial stage (x=0.1m) the effective coefficient of segregation is close to equilibrium one. It is defined by small growth rate. Thus the crystal grows without foreign inclusions. As you can see from the figure the effective coefficient of segregation doesn’t depend on temperature gradient in melt practically.

3. The investigation results
The results showed that the melt width increased with heat temperature increase on 300 K and as a result the crystallization front location changed. So we can conclude that we have possibility to control the crystallization front location by heat temperature increase and it can lead to defects redistribution. The suggested model describes adequately the processes of bubbles formation in sapphire and can be used at the decision of various scientific and practical problems for crystals defects investigation.

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