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Numerical simulation of the distribution of individual gas bubbles in shaped sapphire crystals

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ABSTRACT

The simulation of the effective density of individual gas bubbles in a two-phase melt, consisting of a liquid and gas bubbles, is performed using the virtual model of the thermal unit. Based on the studies, for the first time the theoretically and experimentally grounded mechanism of individual gas bubbles formation in shaped sapphire is proposed. It is shown that the change of the melt flow pattern in crucible affects greatly the bubble density at the crystallization front, and in the crystal. The obtained results allowed reducing the number of individual gas bubbles in sapphire sheets.

1. Introduction

The non-substrate sapphire market develops slowly but surely. Now non-substrate optical sapphire components are produced mainly using large sapphire ingots grown by the KY method. The Stepanov (EFG) simultaneous crystal growth technology can compete with the KY method, provided that process productivity and crystal quality are improved.

One of the distinctive defects of shaped sapphire crystals is clusters of so-called subsurface gas pores (voids) with a size of 5–15 μm [1,2]. The voids are located at a depth of 50–300 μm from a side surface of a crystal. The mechanism of their formation was thoroughly studied [2–6]. Gas impurities accumulate in front of a melt-solid interface. That violates its morphological stability, and the formation of faceted cells greatly facilitates the nucleation of gas bubbles and their capture by a crystal. Under production, these inevitable defects are removed by grinding of as-grown crystals.

Another type of pores is large gas bubbles of 100–500 μm in size. They are irregularly located in the volume of a crystal. The presence of these bubbles in optical products is not allowed. Normally, the number of these bubbles amounts 4–6 per as-grown sheet, Fig. 1. The problem becomes much more acute when the area of the workpiece is enlarged. Thus, for sapphire ribbons, shown in Fig. 1, the yield of 50 mm × 44 mm blanks amounts 83% of usable surface area, but it drops to 33% for 100 mm × 88 mm blank.

Although, individual gas bubbles are the main cause of surface area losses, the mechanism of their formation is poorly understood. In work [7], the author supposed that large bubbles nucleate heterogeneously on working surface of a die, grow up, absorbing a gaseous impurity in melt meniscus, and then are captured by a crystallization front. In [8,9] it was statistically detected that the number of individual gas bubbles in sapphire ribbons depends on temperature distribution in the crucible, gas pressure in the chamber, and melt holding time before seeding. The data allowed reducing the number of individual gas bubbles in commercially produced crystals by 13%. However, for the competitive production of large sapphire blanks, it is necessary either to completely eliminate these defects, or at least to scale down their number in several times. This required a solid understanding of the mechanism of this defect formation.

2. The mechanism of individual gas bubbles formation

According to [7], a gas bubble, which nucleates and grows on a die top face, becomes a drain for gas impurity, and, consequently, should reduce the density and change the location of subsurface voids. Fig. 2 presents the numerical estimate of bubble growth influence on the impurity distribution in the melt meniscus

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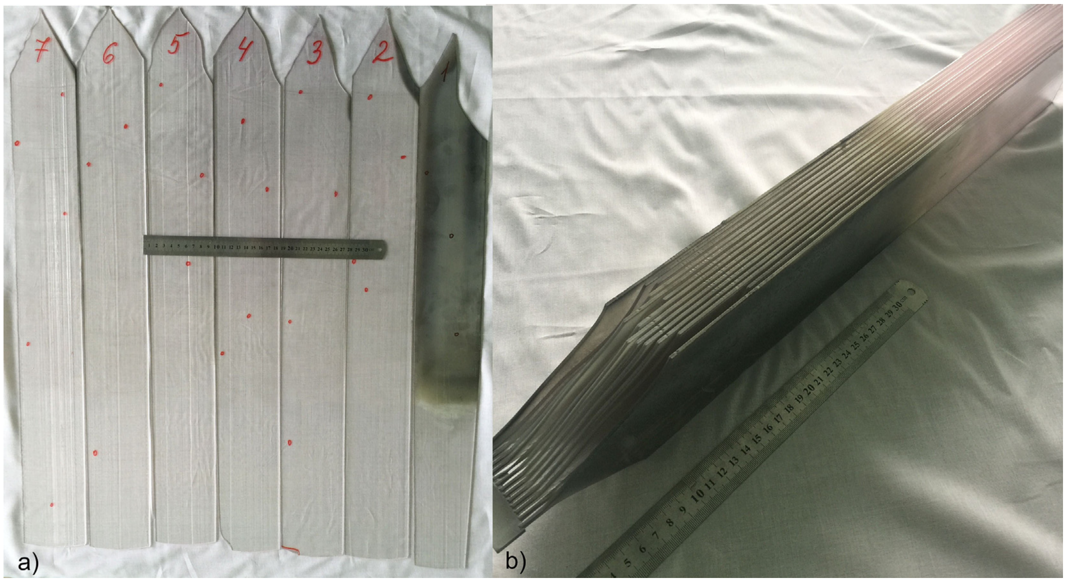

Fig. 1. Typical distribution of individual gas bubbles (point marks) in as-grown sapphire ribbons with the dimensions of 700 mm × 90 mm × 2 mm (half of the package is shown) – (a), the package of 14 crystals – (b).

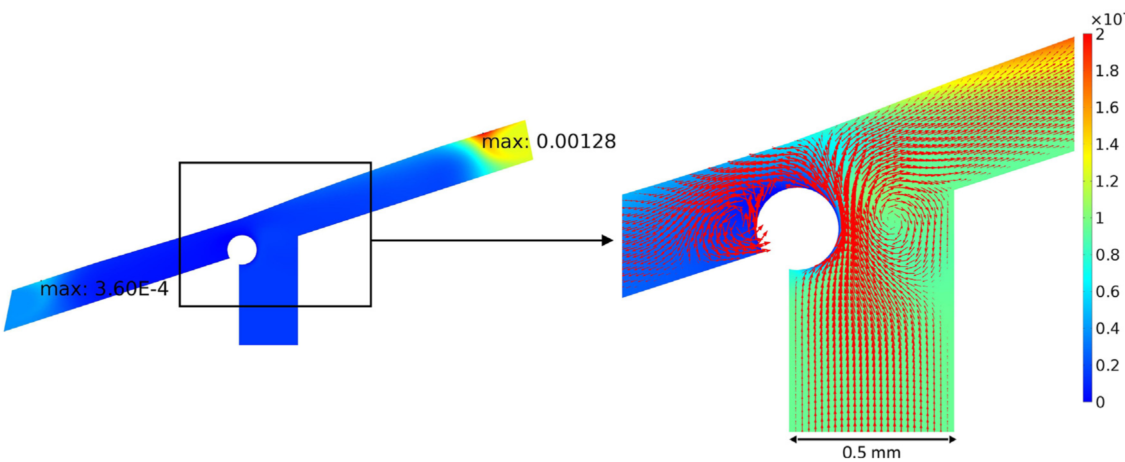

Fig. 2. Impurity concentration (mol/m^3) in the meniscus with the gas bubble growing on the die top face and the melt flow in the region of capillary channel.

(position (5) of Fig. 4).[1] However, our numerous micro-optical studies do not reveal that the individual gas bubble violates an arrangement of subsurface voids, Fig. 3(a).

Moreover, we have experimentally established that the displacement of the capillary cut relative to the center of the die top face causes a similar displacement of individual gas inclusions in the crystal, Fig. 3(b). This result also contradicts the theory stated in [9]. Finally, a micro-optical study of melt samples rapidly crystallized in the crucible revealed the presence of round-shaped gas inclusions with a diameter of 0.1–0.5 mm, Fig. 3(c).

[1] The formulation of the problem of impurity distribution in the meniscus with consideration of thermo capillary convection is well known and presented [5,6]. The Marangoni effect is modeled by using a weak form of the boundary condition on the free surface of the meniscus and of the gas bubble – an action of thermo capillary convection, which causes a melt velocity gradient along the surfaces of both meniscus and gas bubble, is balanced by a viscous shear stress. The change in the coefficient of surface tension of melt was considered linear and equal to 1.9e−4 N/(m K). The initial impurity concentration in the crucible in the region of the entrance to the capillary channel is 10^{-4} mol/m^3, the diffusion coefficient is 2e−9 m^2/s, and the distribution coefficient is 0.1. Impurity drain boundary condition for the bubble consists of two terms – diffusive and convective. The second exceeds the first by orders – a bubble grows mainly due to its motion relative to a gas-enriched liquid.

These results give us grounds to consider the melt as a two-phase medium that includes liquid (molten alumina) and gas bubbles and to assume that nucleation of bubbles occurs heterogeneously on the entire surface of the crucible, instead of the die top face. After detachment from the crucible surface, the bubble moves relative to convectively circulated liquid under the action of buoyant and viscous drag forces. The bubbles, that do not escape the melt free surface, enter the die capillary channel, pass through it to the crystallization front, and penetrate the growing crystal. In order to prove this mechanism, we performed its numerical simulation and compared the obtained dependences with the experimentally observed ones.

3. Formulation of the problem

We consider the typical growth process with the use of NIKA-PROFILE crystal puller [10]. The axial symmetry of the cylindrical chamber of the installation, the inductor and the main elements of the thermal unit allows using the two-dimensional axisymmetric model. The pulling crystal is a sapphire tube with the outer diameter close to 59 mm, the length of 200 mm, and the wall