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Abstract

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OSCILLATIONS OF THE CRYSTALLIZATION FRONT DURING THE GROWTH OF SILICON BY THE CZOCHRALSKI METHOD

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Data on temperature oscillations in the melt during the growth of single crystals by the Czochralski method (¹⁻⁴) indicate the possibility of oscillations in the position of the crystallization front (c.f.). This is confirmed by the observed oscillations of the c.f. during horizontal zone melting of bismuth (⁵). However, no direct observations of the behavior of the c.f. in this case have been published. We carried out a study of the behavior of the visible boundary of the c.f. during the growth of single crystals by the Czochralski method using motion-picture and photographic recording.

Silicon was chosen for the experiments, since, owing to its high melting temperature, the need for illumination during filming was eliminated, and the absence of films on the surface of the melt and crystal, together with the noticeable difference in the degree of blackness of solid and molten silicon, makes the c.f. clearly visible. Here and below, by c.f. we mean its visible boundary.

The melts were carried out on two installations of different types. The greater part of the melts was performed on an installation with high-frequency heating in flowing helium, and some of the melts, for comparison, on an installation with a resistance heater in vacuum (10^{-4} mm Hg).

For photography and motion-picture recording, special branch tubes fitted with optical glass were made in the chambers. The axes of the branch tubes coincided, were perpendicular to the pulling axis, and were located at the level of the c.f., which made it possible to record the c.f. simultaneously along its entire length.

In the first series of experiments, in order to determine the nature of the oscillations, motion-picture recording of the c.f. was carried out at filming rates from 10 to 4000 frames per second. Filming was performed on 16-mm motion-picture film with various motion-picture cameras (SKS-1, Pentazet 16 "Applex"). The conditions and optics were selected so that the size of the c.f. image was close to the frame size, which corresponded to a scale from 1 : 1 to 1 : 3. Analysis of the

position of the c.f. was carried out by frame-by-frame viewing of the projected image.

The change in the position of any point along the generatrix of the ingot at the c.f. was measured from a fixed reference point. Comparison of the results obtained when measuring from a reference point taken on the ingot and on the melt surface showed that the result does not change in this case.

The oscillations of the c.f. in the frame amounted to 0.1-0.3 mm, i.e., they were considerably greater than the resolving power of the filming method used.

Analysis of the motion-picture records obtained showed that there are three types of c.f. oscillations.

The first and most strongly expressed type of oscillation is governed by rotation of the crucible. To determine the causes and character of this kind of oscillation, simultaneous recording of the entire perimeter of the c.f. was carried out. It showed that the c.f. has one or two depressions toward the melt at different moments in time. The depression (the difference between the upper and lower positions of the crystallization front) reaches 1 mm.

A typical view of scans of the c.f. is shown in Fig. 1. During rotation of the crucible (Fig. 1a), the depressed part, changing its shape with time, moves

together with the rotation of the crucible, lagging behind it, which indicates a hydrodynamic cause of the difference in the melt temperature near the crystallization front. The lag is most strongly expressed at low crucible rotation rates. For example, at a rate of 2 revolutions per minute, the rotation rate of the front is approximately 1 revolution per minute. This form of the crystallization front during crucible rotation causes alternation of moments of melting of the ingot with moments of growth.

If, in addition to the crucible, the ingot also begins to rotate, the general behavior of the crystallization front does not change; however, the development of the crystallization front becomes flatter (Fig. 1b).

The behavior of the crystallization front remains unchanged during lifting and stopping of the ingot, i.e., the growth process almost does not change the observed picture (1c).

If one considers the oscillations of the crystallization front from the side of the ingot along some generatrix of the ingot, then, during rotation of the latter, the oscillation caused by rotation of the crucible and the rotation of the ingot are superposed on one another, since each generatrix, in one revolution of the crystal, passes through the bend of the crystallization front. This causes periodic melting of a part of the ingot with the frequency of rotation of the seed. It should be noted that oscillation of the crystallization front of the first type occurs also in the absence of rotation of the crucible and seed, but in this case the oscillations are random in character, determined by a spontaneous change in heat removal.

Fig. 1

Figure 1: Fig. 1

The second type of oscillations of the crystallization front has a different character. In this case the front moves upward or downward approximately parallel to itself. The frequency of these oscillations varies within the limits of 1-1.5 oscillations per second. Figure 2 shows the curve of the change in the position of the crystallization front at three points on the crystallization front, displaced in the plane of the crystallization front relative to one another by 60° . The envelope of these curves is the oscillations of the second type, while the higher-frequency oscillations are oscillations of the third type (see below). Their amplitude reaches 0.5 mm. Stopping or growth of the ingot, as well as changing the rotation conditions, do not change the general character of the oscillations.

Fig. 1. Development of the surface of the crystallization front:

a –during rotation of the crucible without growth of the ingot, crucible rotation rate 2 rpm;

b –during rotation of the crucible and crystal without growth of the ingot;

c –during rotation of the crucible and crystal and growth of the ingot, crucible rotation rate 2 rpm, crystal rotation rate 6 rpm, growth rate 1 mm/min.

It should be noted that when the seed or crucible rotates at a rate above 20-30 revolutions per minute, the frequency of oscillations of the first type becomes comparable with the frequency of oscillations of the second type, which makes them difficult to distinguish.

The third type of oscillations has a significantly higher frequency, and the oscillations themselves have the character of short-term fluctuations from the mean value.

Figure 3b shows the pattern of change in the position of the crystallization front in this case, constructed by frame-by-frame analysis of a motion picture taken at a rate of 4000 frames per second. The frequency of the oscillations in this case is 150-160 oscillations per second, and the amplitude of the oscillations reaches 0.2 mm. At a lower filming frequency, because of the lower resolving power and the camera shutter, these oscillations have the form shown in Fig. 2 (modulating oscillations). The pulsed form of the oscillations indicates a high rate of the process, which cannot be recorded when filming at a rate of 4000 frames per second.

Both the second and the third types of oscillations lead, during the growth process, to melting of the grown portions of the ingot.

When viewing films taken at low speed, oscillations of the lateral surface of the melt column near the c.f. are visible; they are analogous to the oscillation of the c.f. and have both a low-frequency and a high-frequency character. The

Fig. 2 and Fig. 3: oscillation plots

Figure 2: Fig. 2 and Fig. 3: oscillation plots

low-frequency oscillations have an amplitude of 0.2 mm and a frequency of ~ 2 oscillations per second. The oscillations are asymmetric. The high-frequency oscillations (Fig. 3a

Fig. 2. Oscillations measured at three points on the crystallization front. Envelopes—oscillations of the 2nd type. Filming at a rate of 300 frames/sec

Fig. 3. Form of oscillations of the crystallization front (Δ), and of the right (\square) and left (\circ) sides of the melt column when filmed at a rate of 4000 frames/sec

and \square) have an amplitude of 0.1 mm and a frequency of 150 oscillations per second. These oscillations are symmetric in character.

As was noted, the first type of oscillations is associated with asymmetry in the mixing of the melt.

The causes of the second and third types of oscillations should be sought in temperature oscillations set up by convective flows caused by the presence of temperature gradients.

The short-time character of the processes occurring in the third type of oscillations indicates a direct motion of convective flows near the c.f. (i.e., in the melt column), or a surface character of the melting associated with oscillations of the melt.

The second type of oscillations may be connected with processes occurring in the melt itself, and with the filtering out of temperature oscillations during transfer through the melt column.

Our recording of thermocouple readings on a high-speed recorder confirms the presence of temperature oscillations in the melt with a frequency of several oscillations per second and an amplitude of up to 10° . However, the thermocouple-recorder system must filter out high-frequency temperature oscillations, and therefore the possibility of direct compa-

of changes in the thermocouple readings and analysis of the cine records requires special study.

As was shown in paper (6), direct comparison of cine records of crystallization-front oscillations with impurity bands in the grown ingots shows that melting causes the formation of transverse impurity bands in the grown ingots. However, the relation between crystallization-front oscillations and the appearance of impurity bands is more complicated, since the largest oscillation causes melting of part of the grown ingot with already fixed growth bands, and therefore not every crystallization-front oscillation is reflected in the impurity structure of the ingot.

In view of the foregoing, the method we used of cine filming the crystallization front directly has certain advantages over the study of temperature oscillations in the melt and the study of the finished ingot.

The data we have obtained indicate the need to control crystallization-front oscillations during crystal growth in order to improve the impurity structure of ingots.

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REFERENCES

1. Mühlbauer, *Zs. Naturforsch.*, **21a**, No. 4, 490 (1966).
2. A. Muller, M. Wilhelm, *Zs. Naturforsch.*, **19b**, 254 (1964).
3. W. R. Wilcox, L. D. Fullmer, *J. Appl. Phys.*, **36**, No. 7, 2201 (1965).
4. D. T. J. Hurle, *Phil. Mag.*, **13**, No. 122, 305 (1966).
5. G. V. Komarov, A. R. Regel, *FTT*, **5**, issue 3, 773 (1963).
6. Yu. M. Shashkov, N. Ya. Shushlebina, *DAN*, **178**, No. 1 (1968).

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