Volume 310 Issues 7–9 April 2008 ISSN 0022-0248

ELSEVIER

JOURNAL OF CRYSTAL GROWTH

Editors: T.F. KUECH (Principal Editor)
M. SCHIEBER (Founding Editor)
R.S. FEIGELSON, R. KERN,
K. NAKAJIMA, G.B. STRINGFELLOW

The Proceedings of the 15th International Conference on Crystal Growth (ICCG-15) in conjunction with the International Conference on Vapor Growth and Epitaxy (ICCG-15) and the US Biennial Workshop on Organometallic Vapor Phase Epitaxy (OMVPE-13)

12 – 17 August, 2007
Salt Lake City, Utah, USA

Guest Editors ICCG-15
Thomas F. Kuech
Susan E. Babcock
and
Guest Editors OMVPE-13
Rajaram Bhat
Catherine Caneau

Available online at
ScienceDirect
www.sciencedirect.com

Available online at www.sciencedirect.com
ScienceDirect
ELSEVIER
Journal of Crystal Growth 310 (2008) 1343–1348
JOURNAL OF CRYSTAL GROWTH
www.elsevier.com/locate/jcrysgro

Experimental and computational study of flow instabilities in a model of Czochralski growth

Maxim Teitel[a], Dietrich Schwabe[b], Alexander Yu. Gelfgat[a,*]

[a]Faculty of Engineering, School of Mechanical Engineering, Tel-Aviv University, Ramat Aviv, 69978 Tel Aviv, Israel
[b]Physikalisches Institut, University of Giessen, Giessen, Germany

Available online 3 December 2007

Dedicated to Prof. Dr. A. Scharmann on the occasion of his 80th birthday

Abstract

The results of experimental modeling of Czochralski flow of large-Prandtl-number oxide melts are reported. An instability mechanism resulting in cold plumes detaching from the cold crystal and then descending towards the crucible bottom is observed. With the increase of the temperature difference or crystal rotation the baroclinic instability mechanism, characterized by three-dimensional oscillations of a cold jet descending along the symmetry axis, becomes dominant. The experiments are carried out for different silicone oils characterized by different Prandtl numbers and for varying crystal/crucible radii ratio. Results of computational modeling results qualitatively agree with the experimental observations. Possible reasons for the quantitative disagreement are discussed.

PACS: 81.10.Fq; 44.25; 47.20; 47.32

Keywords: A1. Convection; A1. Fluid flows; A1. Heat transfer; A2. Czochralski method

1. Introduction

Melt flow instabilities arising during Czochralski growth of bulk oxide crystals can possibly initiate the spiraling shape of the growing crystal [1]. Recent growth experiments [2] reported that a change of direction of crystal rotation changes the twist of the crystal spiral from clockwise to counterclockwise. These observations show that the spiral-growth instability is connected with the melt flow and its instabilities. The goal of the experiments and calculations described below was to detect at which critical conditions the instability of the initially steady and axisymmetric melt flow sets in, and to which three-dimensional (3D) flow pattern the instability develops.

Oxide melts are characterized by Prandtl numbers in the range $5<Pr<20$. This allows us to model their flows experimentally using different silicone oils in the slightly modified experimental setup of Czochralski melt flow previously used in Refs. [3,4]. The experimental visualization and measurements are done by thermocouples, shadowgraph technique and PIV, and are focused on the determination of critical parameters corresponding to a transition from an axisymmetric to a 3D flow state. The experimental setup allows us to vary the crystal rotation rate and the temperature difference between the cold crystal dummy and the hot crucible wall in wide ranges. Special care is taken to define all velocity and thermal boundary conditions with high precision, so that they can be adequately reproduced in a computational model.

Varying the silicone oils and crystal dummy diameter we observed an instability, which can be described as formation of cold plumes below the crystal with their consequent detachment from the crystal dummy and descending along the symmetry axis. Such instability was mentioned in an earlier review of Carruthers [5] as hot thermal plumes, rising from the crucible bottom. Cold plumes detaching from the crystal surface were observed in

*Corresponding author. Tel.: +972 3 6407207; fax: +972 3 6407334.
E-mail address: gelfgat@eng.tau.ac.il (A.Y. Gelfgat).

0022-0248/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.jcrysgro.2007.11.164

model experiments, where the working liquid was silicone oil with the Prandtl number 1000 and larger [6,7]. In these experiments crystal or crucible always rotated. To the best of our knowledge such instability was never observed before in Czochralski experiments for $Pr<50$ and for the stationary crystal and crucible. Based on previous experiments [3,4] with less viscous silicone oil we assume that this type of instability can be characteristic for large-Prandtl-number melts, or for the systems having significant concentration gradients and large Schmidt numbers, e.g., in case of growth from a solution or in melts of heavily doped semiconductors.

Recent numerical studies [8–10] report a steep decrease of the critical temperature difference, corresponding to the primary melt flow instability, with the increase of the crystal rotation. One of the goals of the present study was to check these predictions experimentally. Another goal was to generate the experimental data that can be used later for validation of the codes dedicated to a direct analysis of 3D instability of Czochralski melt flow [9,10]. The convergence studies made for a certain Czochralski configuration [10], as well as more detailed convergence studies made for a series of model problems [11,12], showed that the reliable stability results are extremely demanding to the numerical accuracy. This makes it necessary to validate the codes dealing with such a complicated system as Czochralski melt flow against reliable experimental data.

2. Experimental setup

The experimental setup is the same as that used in Refs. [3,4] and is shown schematically in Fig. 1. The system consists of seven main parts: (1) a middle heating chamber; (2) bottom heating chamber; (3) an upper cooling chamber; (4) a sapphire crucible; (5) a pulling rod; (6) a crystal dummy; and (7) thermal baths (not shown) that support constant heating and cooling of water temperatures for (1)–(3). Details can be found in Refs. [3,4].

The bulk of the experimental results was obtained by thermocouple measurements and shadowgraph observations. Properties of different silicone oils used in the experiment are summarized in Table 1. Note that the physical properties of the silicone oils are close, except their viscosities. This allows us to interpret changes of the experimental liquid as a change of viscosity or, assuming almost unchanging heat diffusivity, as a change of only the Prandtl number. At the same time we must keep in mind that other governing parameters, i.e., Grashof, Marangoni and Reynolds numbers also depend on viscosity. The crystal radius was varied using different dummies that had diameters 14, 15.76, 18, 20 and 22 mm. Their lateral surface was shaped in a way that preserved the area, thus ensuring that the heat transfer with the air convecting in the upper chamber remains approximately the same. The crucible inner radius was 20 mm, and the height of the melt was 20 mm in all the experiments.

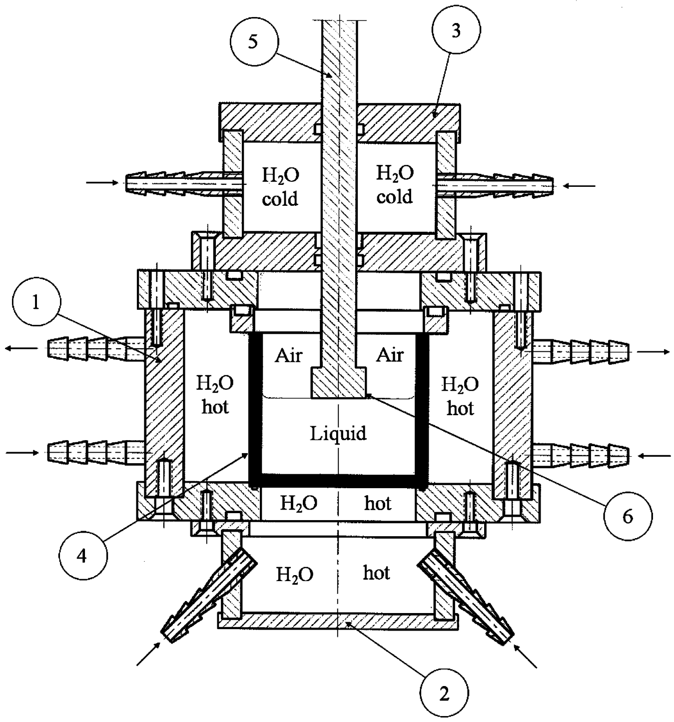

Fig. 1. Sketch of the experimental setup.

Table 1
Properties of silicone oils

Oil	1	2	3	4	5
Kinematic viscosity ν (10^{-6} m^2/s)	0.66	2	5	10	50
Density ρ (kg/m^2)	760	860	910	930	950
Thermal expansion coefficient β (10^{-3} K^{-1})	1.34	1.2	1.1	1.0	1.0
Heat capacity c_p (10^3 J/(kg K))	1.38	1.39	1.4	1.43	1.45
Thermal diffusivity χ (10^{-8} m^2/s)	9.5	8.4	8.6	9.77	10
Thermal conductivity λ (W/(K m))	0.1	0.1	0.11	0.134	0.15
Surface tension σ (10^{-3} N/m)	15.9	19.5	19.7	19.9	19.9
$\partial\sigma/\partial T$ (10^{-5} N/(m K))	−6.4	−6.4	−6.4	−6.4	−5.9
Prandtl number (Pr)	6.8	28	58	102	500

3. Results

3.1. Experimental results

The most unexpected observation of the present experiments was an instability illustrated by the shadowgraph snapshots in Fig. 2. The instability can be described as follows. Hot fluid ascends along the crucible wall and then is driven along the free surface from the crucible wall towards the crystal. Note that the buoyant and the thermocapillary force drive the convective circulation in the same direction (see also Section 3.2). Passing along the free surface the hot liquid loses some heat to the air convecting above the interface. The convective circulation drives this slightly cooled fluid below the dummy–oil interface where the fluid is being cooled down even more

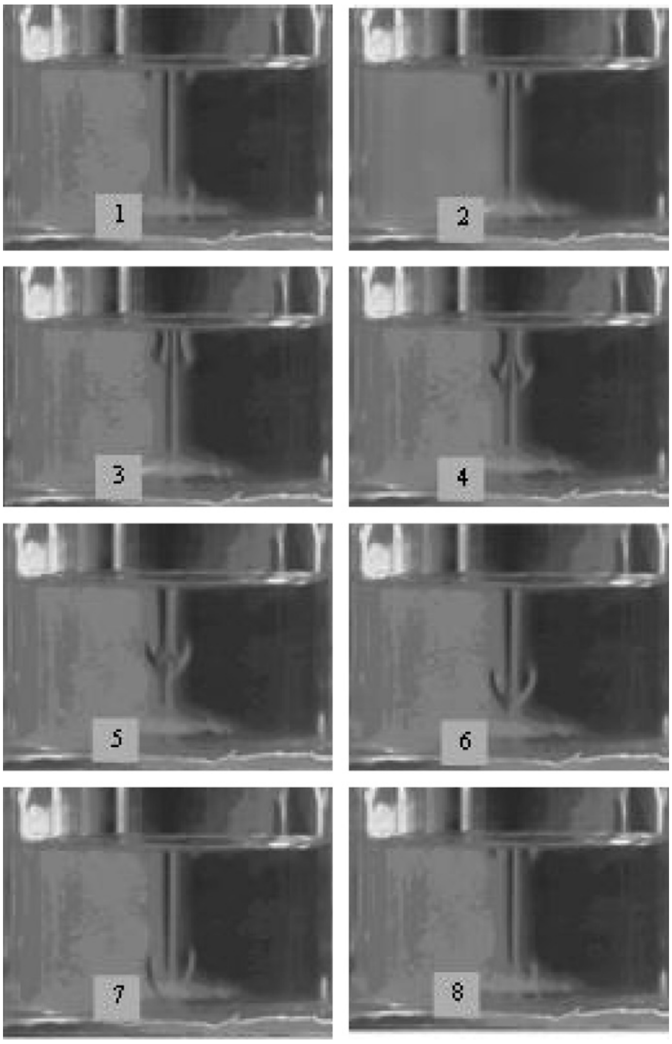

Fig. 2. Shadowgraph snapshots of "cold plume" instability. Oil 2, temperature difference is 20 K, no rotation. The radii ratio is 0.35.

intensively due to the large heat conductivity of the copper dummy. This creates an unstable stratification in which the cold fluid is positioned above the warmer one. It is seen in Fig. 1 that at the moment when the stratified layer becomes unstable a plume of cold liquid detaches from the cold surface of the crystal dummy and descends along the axis towards the crucible bottom. The whole phenomenon is strictly periodic.[1]

The critical temperature difference at which the instability sets in was derived from thermocouple measurements. An example of that is shown in Fig. 3. Here, starting from a certain supercritical temperature difference, e.g., $\Delta T = 7\,\mathrm{K}$, we observe periodic oscillations with a finite amplitude of approximately 0.9 °C (Fig. 3a). Gradually reducing ΔT we observe a decrease of the amplitude until it reaches the noise level. In the example shown in Fig. 3a this happens at $\Delta T_{cr} = 4\,°\mathrm{C}$. This temperature difference and the corresponding oscillations frequency we call critical. In Fig. 3b $f_{cr} = 0.19\,\mathrm{Hz}$.

The dependence of the ΔT_{cr} and f_{cr} on the crystal dummy diameter is shown in Fig. 4. With the increase of the crystal diameter the net heat transfer through the melt increases, so that the "cold plume" instability is being observed at lower temperature differences (Fig. 4a). The critical frequency also decreases with the increase of the diameter (Fig. 4b). We observed that at larger cold surface of the crystal dummy larger plumes form. The formation of a larger plume needs more time, which increases the time period of the phenomenon and consequently decreases the frequency.

The ΔT_{cr} and f_{cr} for different silicone oils are shown in Fig. 5. As discussed above, the variation of silicone oils can be interpreted as a variation of the Prandtl number. In Fig. 5a we show the critical temperature difference in logarithmic scale, which leads to the conclusion that it depends on the Prandtl number exponentially. At the same time the dependence of the critical frequency on the Prandtl number is not monotonic.

A slow rotation of the crystal dummy leads to a strong decrease of ΔT_{cr}. For example, experiments with the oil 2 showed that when the 20-mm-diameter crystal dummy rotates with the frequency of 0.07 Hz, the critical temperature difference drops from 4 K at zero rotation to 2 K. This observation qualitatively agrees with the numerical predictions of Refs. [2,8,10]. Further increase of the crystal dummy rotation leads to a replacement of the instability mechanism, so that a so-called "oscillatory jet" instability is observed first. This instability was observed and described in Ref. [4] and is characterized by a precession of a cold descending jet around the symmetry axis. In the present experiments we observed the "oscillatory jet" instability at zero rotation in the more viscous oil 4 (Table 1) with kinematic viscosity of 10 cSt. These observations are illustrated by shadowgraph snapshots in Fig. 6.

A combination of heating and rotation can lead to the simultaneous development of both "cold plumes" and "oscillatory jet" modes. This was observed for the case of Fig. 2, when the crystal dummy rotates with the angular velocity of 0.168 rps. The shadowgraph snapshots for this case are shown in Fig. 7. One can distinguish on these snapshots almost parallel lines that belong to the jet and the lines crossing them, which are traces of the cold plumes.

3.2. Numerical results

Several attempts to calculate the non-isothermal silicone oil flow only and then to study its stability did not succeed to reproduce the observed "cold plume" instability. We concluded that the heat transfer between the melt surface and the air convecting above it cannot be described by a constant convection coefficient, which is a usual assumption of many computational studies. To model this part of the heat transfer problem properly we considered the melt flow along with the convection of air, thus solving the

[1]Some shadowgraph videos are available at http://www.eng.tau.ac.il/~gelfgat/CZ_2007.htm.

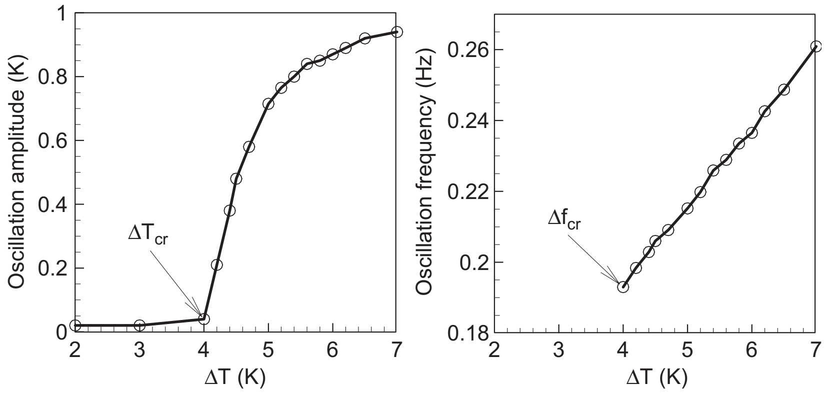

Fig. 3. Dependence of the temperature oscillation amplitude and the frequency of oscillation on the temperature difference between the crystal dummy and the crucible wall. Oil 2, the radii ratio is 0.5.

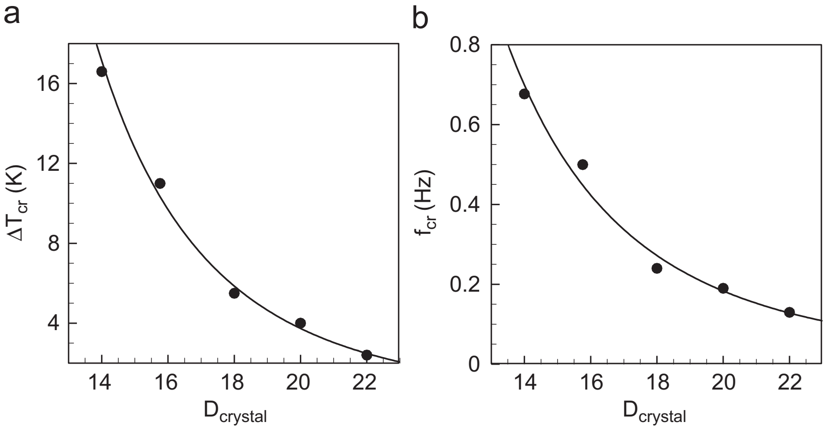

Fig. 4. Dependence of the critical temperature difference (a) and critical frequency (b) on the crystal dummy diameter for oil 2.

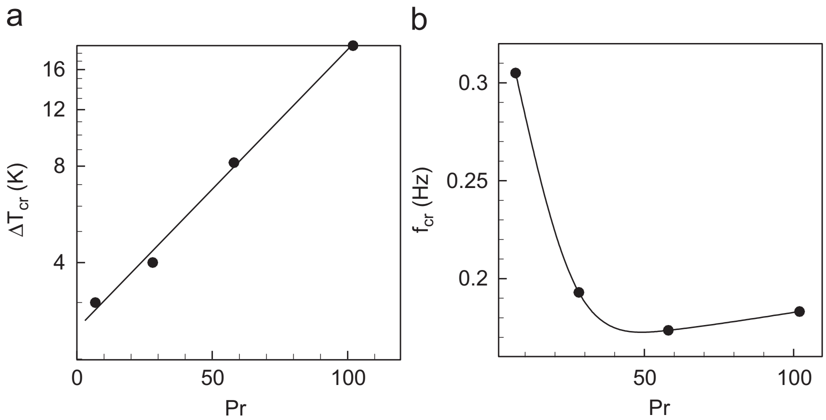

Fig. 5. Dependence of the critical temperature difference (a) and critical frequency (b) on the Prandtl number.

problem for the melt flow, the air convection, and heat conduction inside the crystal dummy simultaneously. Due to the lack of space we do not give here the details of problem formulation and describe only the main features of the flow pattern and some preliminary comparisons with the experiment.

An example of a numerically calculated flow and temperature is shown in Fig. 8. The circulation in the crucible is driven by the buoyancy force, which drives the hot fluid upwards along the crucible wall, and by the thermocapillary force driving the fluid along the oil–air surface from the hot crucible wall towards the cold crystal dummy. Therefore the main convective circulation in the silicone oil is counterclockwise in the right half of the crucible. The convection of air is driven mainly by the buoyancy force with an ascending flow along the heated

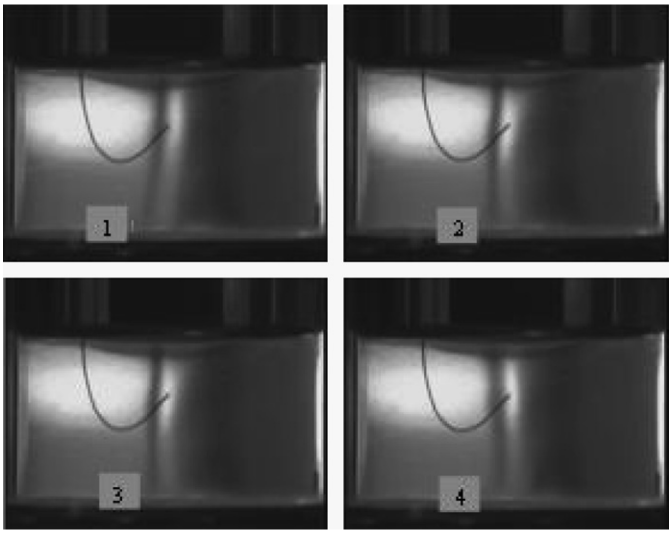

Fig. 6. Shadowgraph snapshots of ‘‘oscillating jet’’ instability. Oil 4, temperature difference is 20.2 K, no crystal rotation. The radii ratio is 0.5.

crucible wall, (above the air–oil interface) and descending flow along the cold pull rod and crystal dummy. Thus, the main convective circulation of air is counterclockwise. These two main circulations are connected by a weak clockwise circulation of air attached to the air–oil interface. At the air–oil interface the air is driven from the hot crucible towards the cold dummy by the thermocapillary liquid motion via viscosity. This leads to a formation of a weak clockwise circulation (Fig. 8).

The weak clockwise circulation of air near the free surface (Fig. 8) complicates the heat transfer between oil and air, thus not allowing us to approximate it by a simplified model. Note also that the examination of isotherms inside the crystal dummy shows that in spite of very high thermal conductivity of copper the crystal dummy is not exactly isothermal. This is a consequence of heat advection by oil convection.

Using the model described we were able to calculate an instability of the flow similar to the experimentally observed ‘‘cold plume’’ one. Corresponding animations of supercritical flows can be found at the WWW page cited above. At the same time we were still unable to compute values of the critical parameters close to the observed ones. Thus, for example, calculating for the parameters of Fig. 3 the calculated ΔT_{cr} is 8.1 K with the f_{cr} of 0.48 Hz. Both numbers are approximately two times larger than those obtained experimentally (Fig. 3). Looking for the reason of this disagreement we considered two main arguments. One is a possible inexactness of physical properties especially that of the temperature coefficient of surface tension coefficient γ. Another one is an evaporation of oil from the interface, which can be enhanced by the air convection. Such evaporation creates a surface heat sink thus affecting additionally the heat transfer between oil and air. We have found that, after reducing γ from -6.4×10^5 to -5.1×10^5 the calculated critical parameters change drastically and become $\Delta T_{cr}=4\,^{\circ}\mathrm{C}$ and $f_{cr}=0.22\,\mathrm{Hz}$, which fits the experimental observation (Fig. 3). We obtain a close result also when, instead of varying γ, we add a surface heat sink of approximately 5.1 W/m^2. It is a striking observation that an addition of such a small surface heat loss, whose effect on the base steady flow is negligible, affects the stability properties so strongly. Unfortunately, we could not fit the results of the experiments with other dummy diameters, using these values of evaporation heat or temperature coefficient of surface tension coefficient. Possibly, the evaporation effect has to be accounted for in a more rigorous way, as it was done in Ref. [13]. It is also still possible that the rather fine grid used here is not sufficient for the complete quantitative resolution of the observed instability, so that detailed convergence studies on finer grids are needed.

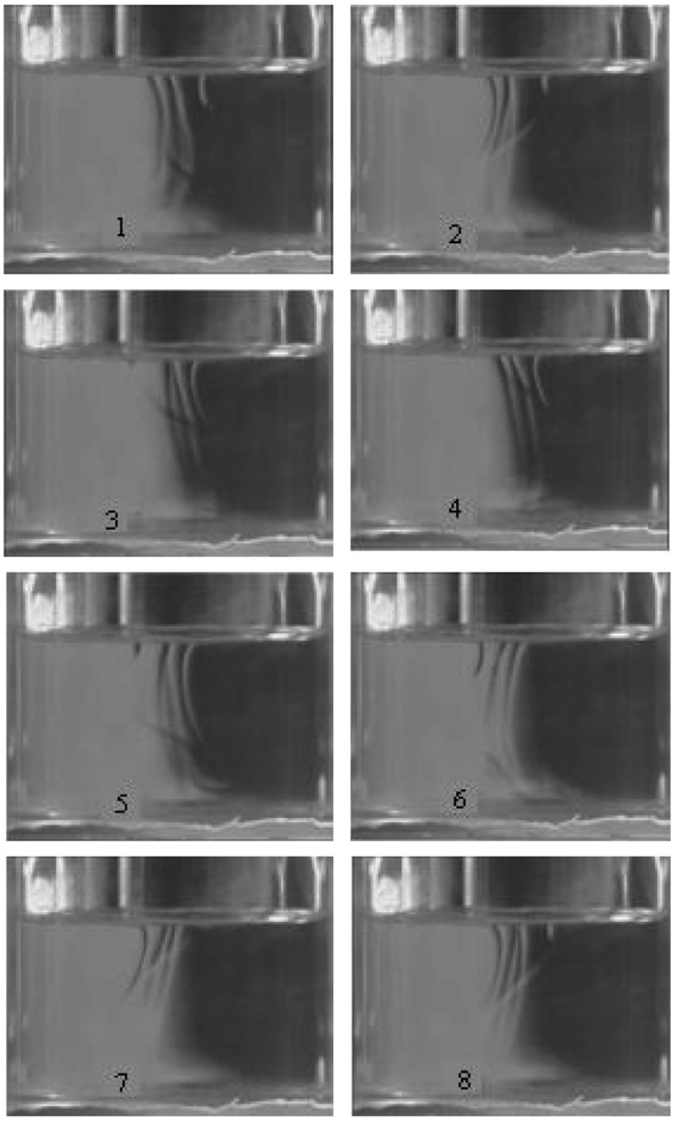

Fig. 7. Shadowgraph snapshots of ‘‘cold plume’’ interacting with an ‘‘oscillating jet’’. Oil 2, temperature difference is 20 K, crystal dummy rotates with 0.168 rps. The radii ratio is 0.35.

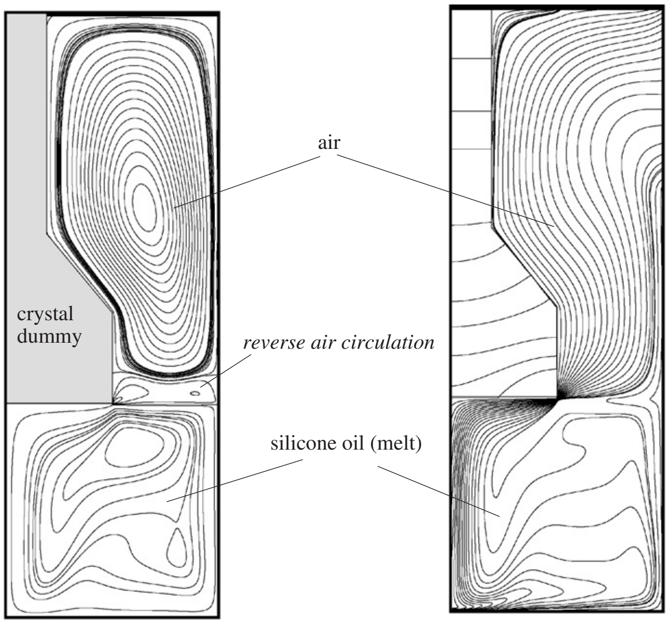

Fig. 8. Streamlines of the silicone oil (melt) flow and air convection (left) and isotherms (right). Oil 2, $\Delta T = 3\,\mathrm{K}$, no crystal rotation. Calculation using 300×600 stretched finite volume grid.

4. Conclusions and discussion

In this study we started a series of experiments whose purpose was to study the possible modes of 3D instability of Czochralski melt flows driven by buoyancy, thermocapillarity and rotation. These experiments are important not only as a tool for better basic understanding of the instabilities and their physical mechanisms, but also for accumulation of reliable experimental data needed for the validation of computational codes.

A series of preliminary experiments already allowed us to observe the "cold plume" instabilities, which were not observed in previous studies of Czochralski melts for working liquids with the Prandtl number below 50, and for non-rotating crystal and crucible it is too early to judge whether this instability can be responsible for the spiraling growth of some refractories. This instability seems to appear only at sufficiently large Prandtl numbers, which explains why it was not observed for experimental liquids with lower viscosity. The decrease of ΔT_{cr} with increasing diameter, with decreasing Prandtl number and under the influence of crystal rotation indicates some likeliness for this instability in real growth. For compound materials, e.g., alloys, solutions or heavily doped semiconductors, the role of a large Prandtl number can be attained by the Schmidt number, which can lead to a similar instability.

The computational modeling of such a sensitive feature as flow instability should exactly reproduce the experimental conditions. A good comparison between the experimental and numerical modeling can be expected only when the physical properties of working materials and boundary conditions are well known. This is fulfilled for our model experiment to a much higher degree than for real growth. Performing the computational simulation of these experiments we could not completely satisfy to both the above conditions, which did not allow us to obtain a quantitative agreement. The qualitative agreement, however, was successfully obtained.

Acknowledgments

This study was supported by the German-Israeli Foundation, Grant no. 1-794-145.10/2004 and by Israel Science Foundation, Grant no. 156/05.

References

[1] R. Uecker, H. Wilke, D.G. Schlom, B. Velickov, P. Reiche, A. Polity, M. Bernhagen, M. Rossberg, J. Crystal Growth 295 (2006) 84.
[2] A.Yu. Gelfgat, E. Kit, K.A. Cliffe, N. Crnogorac, H. Wilke, J. Crystal Growth, submitted for publication.
[3] P. Hintz, D. Schwabe, H. Wilke, J. Crystal Growth 222 (2001) 343.
[4] P. Hintz, D. Schwabe, J. Crystal Growth 222 (2001) 356.
[5] J.R. Carruthers, J. Crystal Growth 32 (1976) 13.
[6] T. Munakata, I. Tanasawa, J. Crystal Growth 106 (1990) 566.
[7] H. Ozoe, K. Toh, T. Inoue, J. Crystal Growth 110 (1991) 472.
[8] A.Yu. Gelfgat, J. Crystal Growth 303 (2007) 226.
[9] H. Wilke, N. Crnogorac, K.A. Cliffe, J. Crystal Growth 303 (2007) 246.
[10] A. Yu. Gelfgat, in: A. Gelfgat (Ed.), Studies on Flow Instabilities in Bulk Crystal Growth, Transworld Research Network, 2007.
[11] A. Yu. Gelfgat, Int. J. Numer. Methods Fluids 53 (2006) 485.
[12] A. Yu. Gelfgat, Int. J. Numer. Methods Fluids 54 (2007) 485.
[13] K.S. Das, C.A. Ward, Phys. Rev. E 75 (2007) 065303.