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F. Muktari, A. Bouabdallah, M. Zizi, S. Hanchi, A. Alemany. Forced convection and thermocapillarity in silicon Czochralski crystal growth in hemispherical crucible. Journal of Physics: Conference Series, 2009, The Third International Symposium on Bifurcations and Instabilities in Fluid Dynamics 10–13 August 2009, Nottingham, UK, 216, pp.012014. <10.1088/1742-6596/216/1/012014>. <hal-00454071>

HAL Id: hal-00454071

<https://hal.science/hal-00454071v1>

Submitted on 2 Apr 2020

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Forced and thermocapillary convection in silicon Czochralski crystal growth in semispherical crucible

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Abstract. In order to understand the influence of a semispherical crucible geometry combined with different convection modes as a thermocapillary convection, natural convection and forced convection, induced by crystal rotation, on melt flow pattern in silicon Czochralski crystal growth process, a set of numerical simulations are conducted using Fluent Software. We solve the system of equations of heat and momentum transfer in classical geometry as cylindrical and modified crystal growth process geometry as cylindro-spherical. In addition, we adopt hypothesis adapted to boundary conditions near the interface and calculations are executed to determine temperature, pressure and velocity fields versus Grashof and Reynolds numbers. The analysis of the obtained results led to conclude that there is advantage to modify geometry in comparison with the traditional one. The absence of the stagnation regions of fluid in the hemispherical crucible corner and the possibility to control the melt flow using the crystal rotation enhances the quality of the process comparatively to the cylindrical one. The pressure field is strongly related to the swirl velocity.

1. Introduction

The silicon crystals used in the technology of semiconductors are mainly produced by the Czochralski technique. During the Czochralski process three modes of heat transfer are present, namely, conduction in melt and crystal, natural convection due to the temperature gradient between the crucible wall and the melt/crystal interface. In addition, forced convection caused by crystal and/or crucible rotation combined to thermocapillary convection which is known as Marangoni convection induced by surface tension gradients on the free surface of the melt. In some cases the heat transfer by radiation between various surfaces of the furnace is also present.

The effect of natural convection on the growth in the traditional system (cylindrical crucible) was studied by several researchers; Kobayashi N [1] and Langlois W E [2] were the first to do it. Turbulent natural convection in large melt volumes have been studied in recent works [3-6]. According to Kumar V et al. [7] the effect of surface tension gradients was neglected in several studies on silicon growth because of the weakness of Prandtl number ($Pr=0.0113$). They showed that the Marangoni convection influences the thermal and dynamic field in the melt. K.Kakimoto and

H.Ozoe [8] carried by using three-dimensional numerical simulations and found that the surface tensions influence the flow and the oxygen transport in melt. By simulating the three-dimensional flow in an oxide Czochralski melt, Basu B et al. [9] have found that the crystal rotation affects the quality and the stability of the growing crystal, the centrifugal force due to the rotating crystal leads to a stabilising effect on the three dimensional flow caused by natural convection. They have found a critical value of Richardson number ($Ri=Gr/Re^2=235$) beyond it the flow become periodic. According to D.T.J.Hurle [10] the rotating crystal acts as a centrifugal fan sucking up fluid axially spinning it up in an Ekman layer and ejecting it tangentially. The radially outward flow due to the rotating crystal meets the radially inflowing fluid driven by the hot crucible wall and a down flow occurs at some radial distance which depends upon the relative strength of the crystal flow and the buoyancy-driven convection. Z.Galazka and H.Wilke [11] have showed that during the growth of YAG, the interface convexity decreases when the crystal rotation rate increases, it becomes almost flat for high Reynolds number values. Crochet M J et al [12] studied numerically the effect of forced convection induced by rotation on the Czochralski crystal growth of GaAs in a cylindrical crucible for different Reynolds number values. Liu L and Kakimoto K [13] analysed the effect of crystal rotation on the growth interface shape in the presence of a transverse magnetic field, they found that the melt-crystal interface changes from an obvious 3D shape when the crystal is not rotating to an almost 2D shape with increase in rotation rate.

Our work consists in studying numerically the combined effects of crucible geometry, thermocapillary convection and crystal rotation on silicon Czochralski crystal growth. We show the advantage of the modified geometry, and we establish the relation between the swirl velocity and the corresponding pressure field for different Reynolds number values.

2. Modelling

Anselmo A P et al. [14] and Tavakoli M H et al. [15] showed that a cylindrical crucible with a curved bottom has advantages for crystallization. Therefore, it is proposed recently by Mokhtari F et al. [16,17,18] to modify the device as a cylindro-spherical system (figure 1).

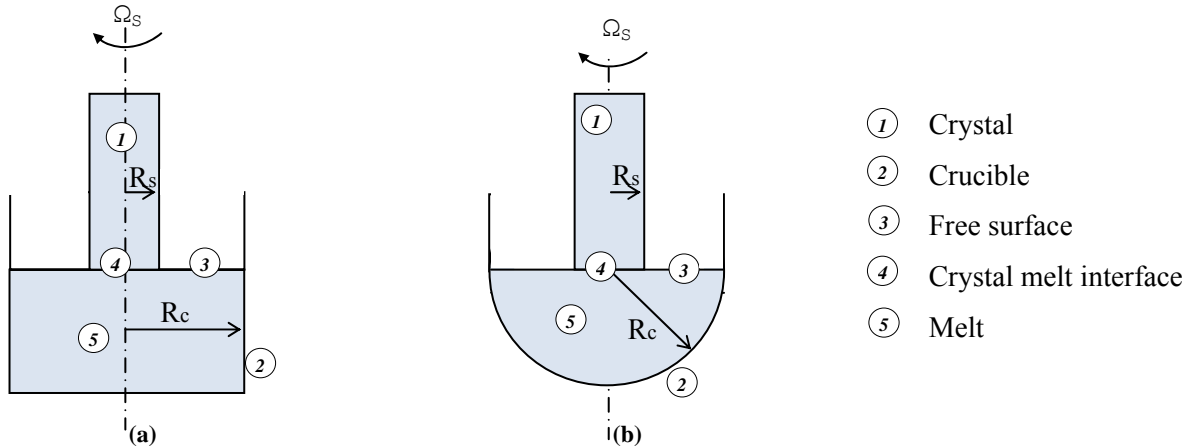


Figure 1. Traditional (a) and modified (b) Czochralski crystal growth systems

To facilitate the procedure of modelling related to the considered problem, we admit some assumptions; the molten silicon is assumed to be a viscous, Newtonian and incompressible fluid satisfying the Boussinesq assumption. We assume the solid liquid interface and the free surface to be flat initially. The thermo physical properties of the fluid are constant except for the density variation in the buoyancy force term. The flow is symmetric in the axial direction. Fixed temperatures are imposed on the walls of the melt crucible and the crystal melt interface. Considering the preceding assumptions, the resolution of the problem will be done in two-dimensional geometries, limited to axisymmetric sections of the crucible represented by the schematic diagrams (figure 2).

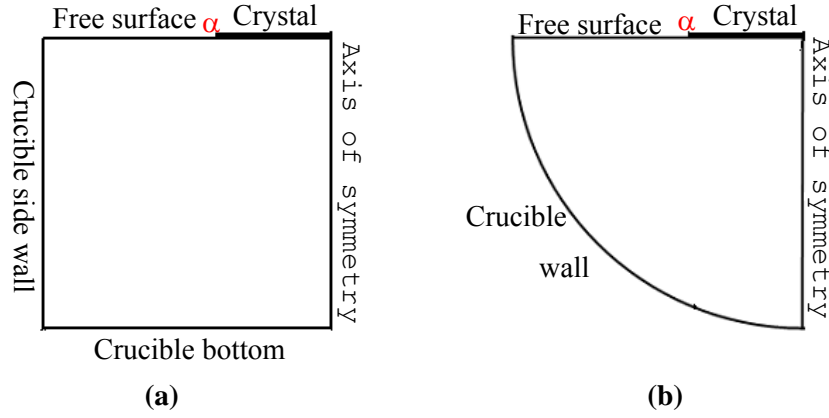


Figure 2. Computational domains for mathematical model: (a) traditional (b) modified

The dimensionless governing equations for the fluid motion and temperature field derived under the above assumptions are

Continuity

$$\frac{\partial V_r}{\partial r} + \frac{V_r}{r} + \frac{\partial V_z}{\partial z} = 0$$

Momentum

$$\begin{aligned} \frac{\partial V_r}{\partial t} + V_r \frac{\partial V_r}{\partial r} + V_z \frac{\partial V_r}{\partial z} &= -\frac{\partial P}{\partial r} + \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial V_r}{\partial r} \right) - \frac{V_r}{r^2} + \frac{\partial^2 V_r}{\partial z^2} \\ \frac{\partial V_\phi}{\partial t} + V_r \frac{\partial V_\phi}{\partial r} + V_z \frac{\partial V_\phi}{\partial z} + \frac{V_r V_\phi}{r} &= \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial V_\phi}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 V_\phi}{\partial \phi^2} + \frac{2}{r^2} \frac{\partial V_r}{\partial \phi} + \frac{\partial^2 V_\phi}{\partial z^2} \\ \frac{\partial V_z}{\partial t} + V_r \frac{\partial V_z}{\partial r} + V_z \frac{\partial V_z}{\partial z} &= -\frac{\partial P}{\partial z} + \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial V_z}{\partial r} \right) + \frac{\partial^2 V_z}{\partial z^2} - \text{Gr} \cdot T \end{aligned}$$

Energy

$$\frac{\partial T}{\partial t} + V_r \frac{\partial T}{\partial r} + V_z \frac{\partial T}{\partial z} = \frac{1}{\text{Pr}} \left\{ \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T}{\partial r} \right) + \frac{\partial^2 T}{\partial z^2} \right\}$$

Where Grashof number $\text{Gr} = \frac{g \beta R_c^3 (T_c - T_f)}{\nu^2}$ characterises natural convection effect. Prandtl number $\text{Pr} = \frac{\nu}{\alpha}$

measures the relative importance of heat transfer by conduction and convection.

The non dimensional boundary conditions are as follows

$$\text{Crucible wall and bottom} \quad V_r = V_\phi = V_z = 0 \quad T = 1$$

$$\text{Crystal melt interface} \quad V_r = V_z = 0 \quad V_\phi = r \cdot \text{Re} \quad T = 0$$

Where $\text{Re} = \frac{\Omega_s R_c R_s}{\nu}$ is Reynolds number associated to the rotating crystal.

$$\text{Axis of symmetry} \quad \frac{\partial V_z}{\partial r} = 0 \quad v_r = 0 \quad \frac{\partial T}{\partial r} = 0$$

$$\text{Free surface:} \quad V_z = 0 \quad \frac{\partial V_r}{\partial z} = \frac{\text{Ma}}{\text{Pr}} \frac{\partial T}{\partial r}$$

The last one is the Marangoni condition, it supposes that the flow perpendicular to the free surface is null, and that the shear stresses at the free surface are balanced by the surface tension gradients [19],

$$\mu \frac{\partial V_r}{\partial z} = - \frac{\partial \sigma}{\partial r}$$

Where $Ma = -\frac{\partial \sigma}{\partial T} \frac{R_c (T_c - T_f)}{\mu \alpha}$ is Marangoni number measuring the effect of thermocapillary convection.

The properties used in simulation are dimensionless parameters. The Prandtl number characterizing silicon is 0.0113. We propose to maintain the Grashof number to 10^5 , the Marangoni number value corresponding is $Ma=2000$ [18] and we vary Reynolds number in the range of 1000 to 15000.

The problem is solved using the finite volume package Fluent. The steady segregated axisymmetric swirl solver was used with the first order upwind discretization for convection, SIMPLE algorithm for pressure-velocity coupling and the PRESTO (PREssure STaggering Option) scheme for pressure interpolation. The convergence is handled by monitoring residuals of continuity, momentum and energy equations.

3. Results and discussions

The aim of this work is based on the numerical study of the geometry effect combined with convection effect with its different forms: natural, thermocapillary (Marangoni convection) and forced (induced by crystal rotation) on flow pattern, temperature and pressure field in silicon Czochralski crystal growth process. By carrying out the convergence test of grid and residuals, we find a grid of 200×200 and residuals of 10^{-6} . We maintain the Grashof number value to 10^5 , thus the corresponding Marangoni number value is approximately $Ma=2000$, we modify the crystal rotation rate by increasing Reynolds number value. For the different analyzed cases we represent the corresponding isotherms, isobars, streamlines and azimuthal velocity contours.

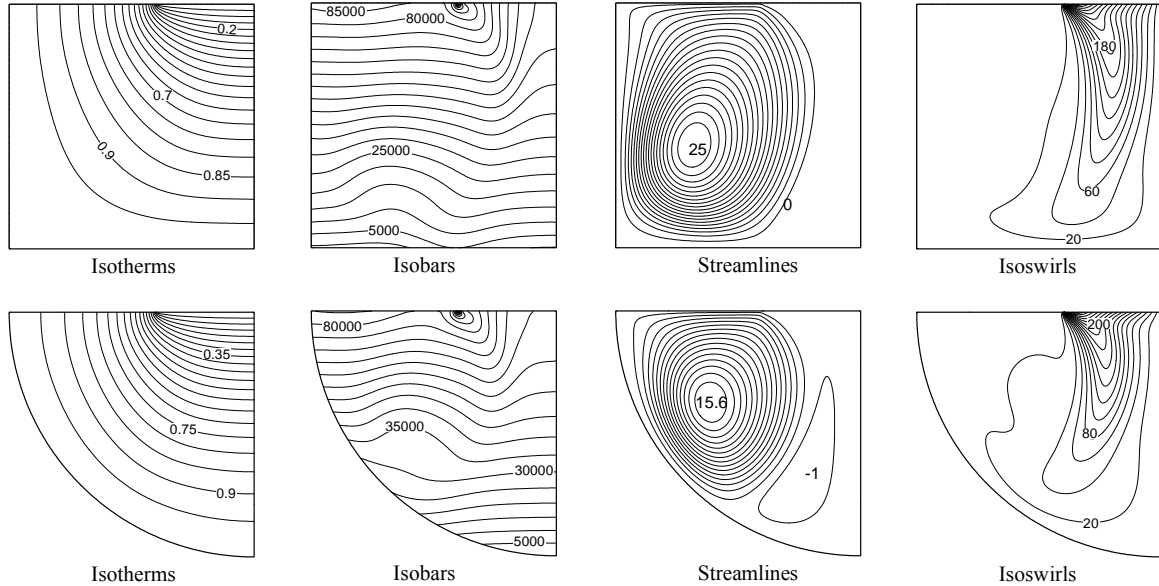


Figure 3. Isotherms, isobars, streamlines, isoswirls for $Gr=10^5$, $Ma=2000$, $Re=1000$

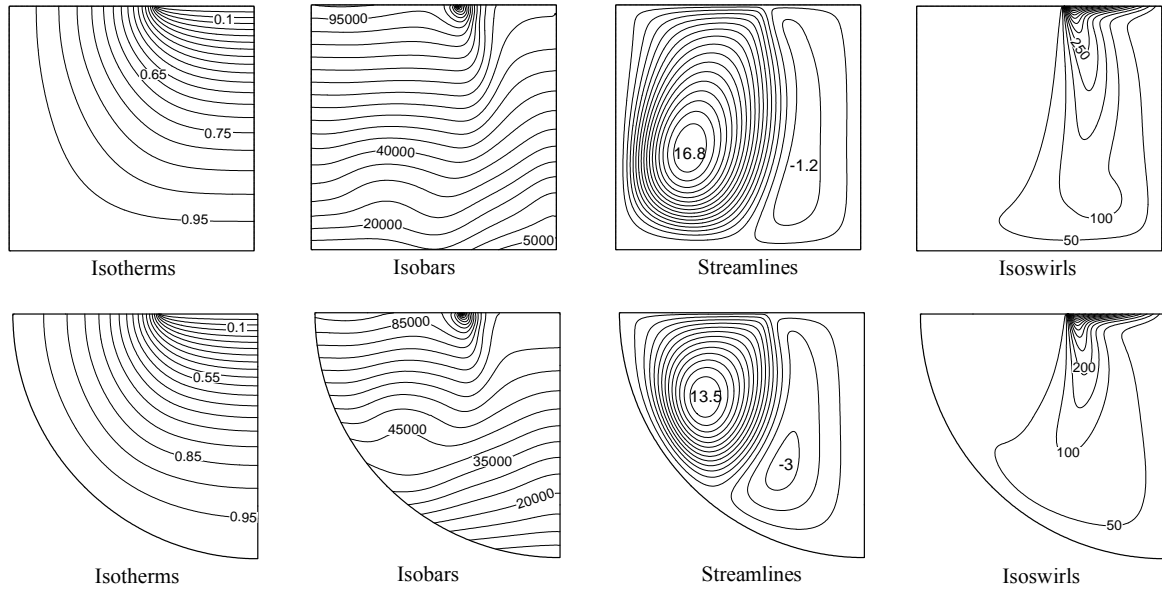


Figure 4. Isotherms, isobars, streamlines, isoswirls for $Gr=10^5$, $Ma=2000$, $Re=2000$

For low Reynolds number values ($Re < 1000$), the melt flow in the crucible is induced by buoyancy and thermocapillary effects so that the convection modes present are natural and Marangoni convection, one anticlockwise vortex is present in the melt. Beyond $Re=1000$ a second vortex, induced by crystal rotation, begins to form in the melt, it is clockwise. Its intensity and volume become higher by increasing the crystal rotation rate.

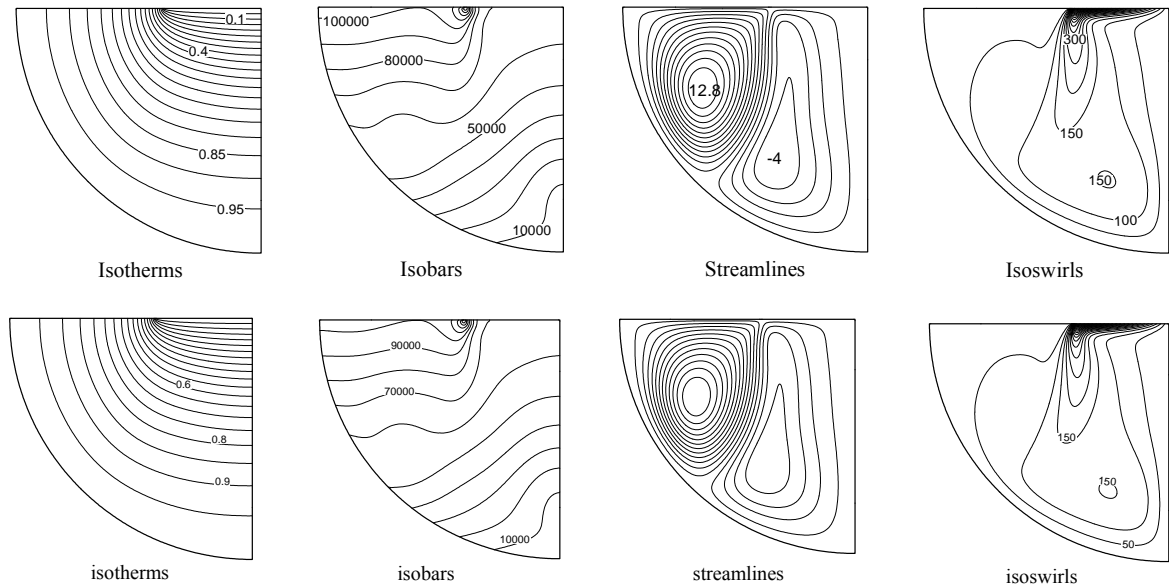


Figure 5. Isotherms, isobars, streamlines, isoswirls for $Gr=10^5$, $Ma=2000$, $Re=3000$

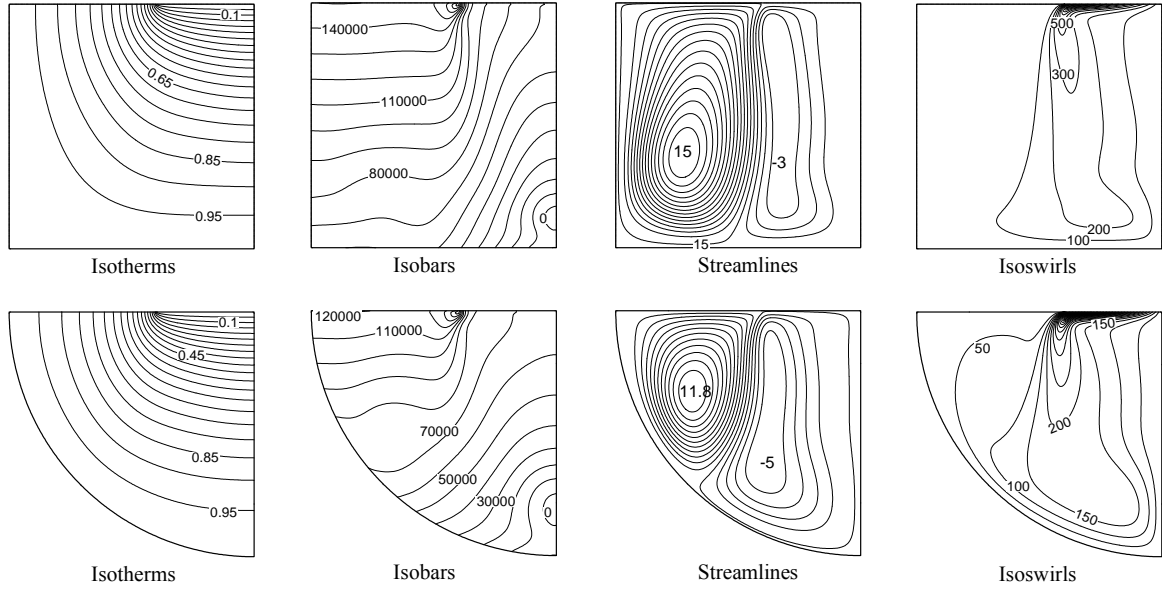


Figure 6. Isotherms, isobars, streamlines, isoswirls for $Gr=10^5$, $Ma=2000$, $Re=4000$

For Reynolds number values $Re \in [1000, 5000]$ corresponding to figures. 3-7 the natural and Marangoni convection dominate, the anticlockwise vortex becomes bigger and more intense. For $Re \approx 5500$ the two vortices have almost the same intensity and occupy the same volume in the melt, this Reynolds number value is corresponding to the transition from the natural and thermocapillary mode toward the forced mode. Beyond this critical value, that's the forced vortex (clockwise vortex) which is more intense, it takes the upper part of the crucible whereas the anticlockwise vortex goes down to the crucible bottom.

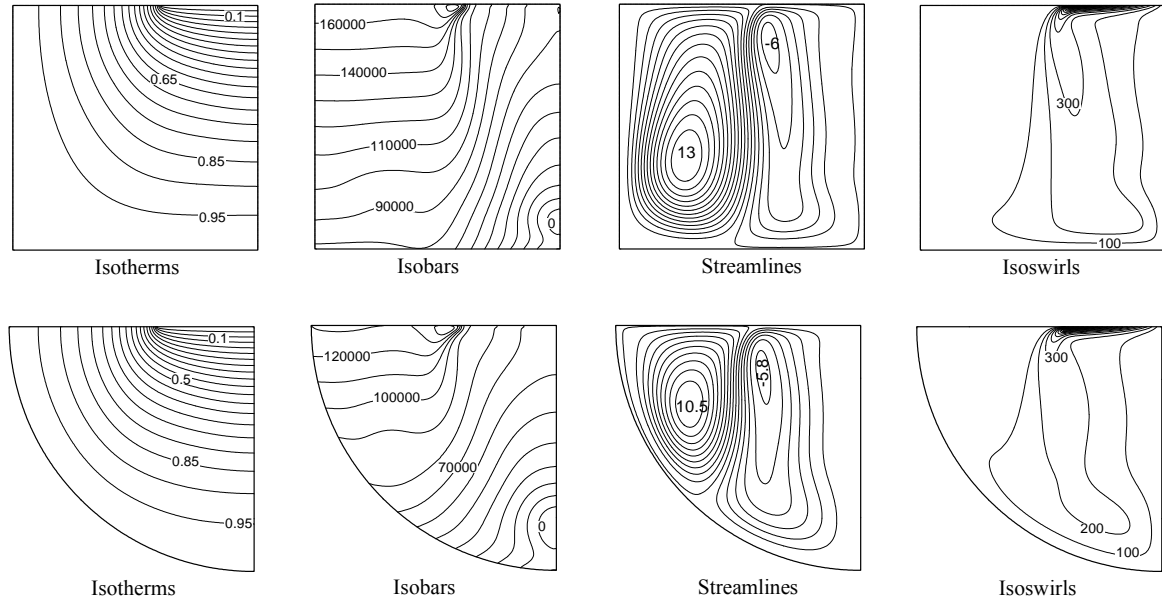


Figure 7. Isotherms, isobars, streamlines, isoswirls for $Gr=10^5$, $Ma=2000$, $Re=5000$

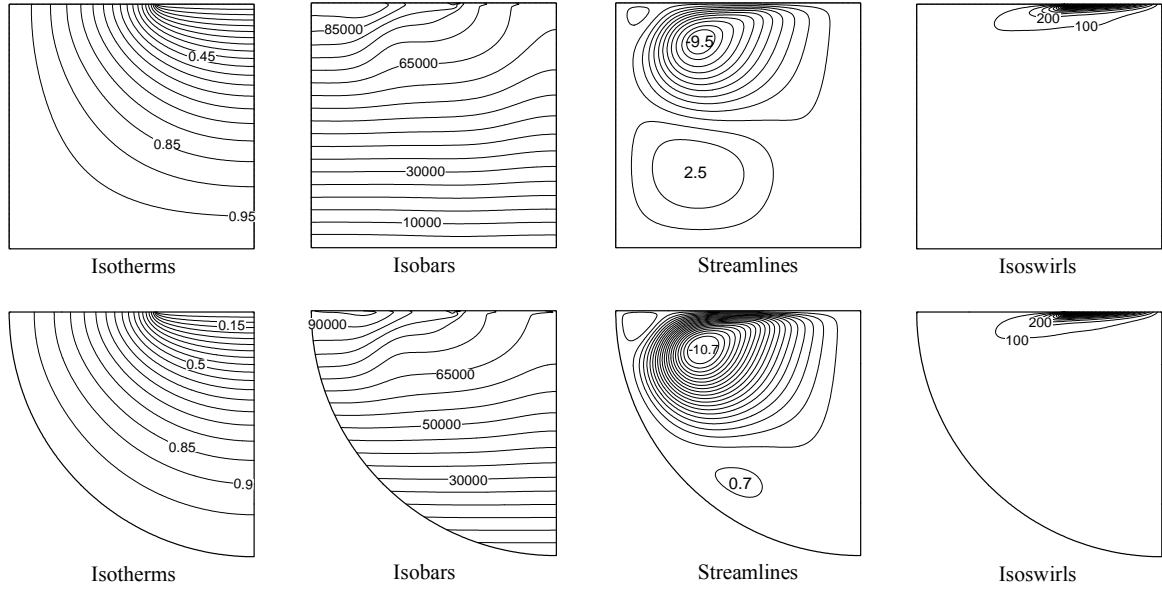


Figure 8. Isotherms, isobars, streamlines, isoswirls for $Gr=10^5$, $Ma=2000$, $Re=6000$

By following the center location of both vortices, we note that for low rotation velocities ($Re < 3000$) the center of natural and Marangoni vortex is above. For Reynolds number $Re=4000$ (figure 6), they are almost in the same level, but beyond $Re=5000$ (figure 7), the center of forced vortex moves upward whereas the other remains in the lower part of the crucible.

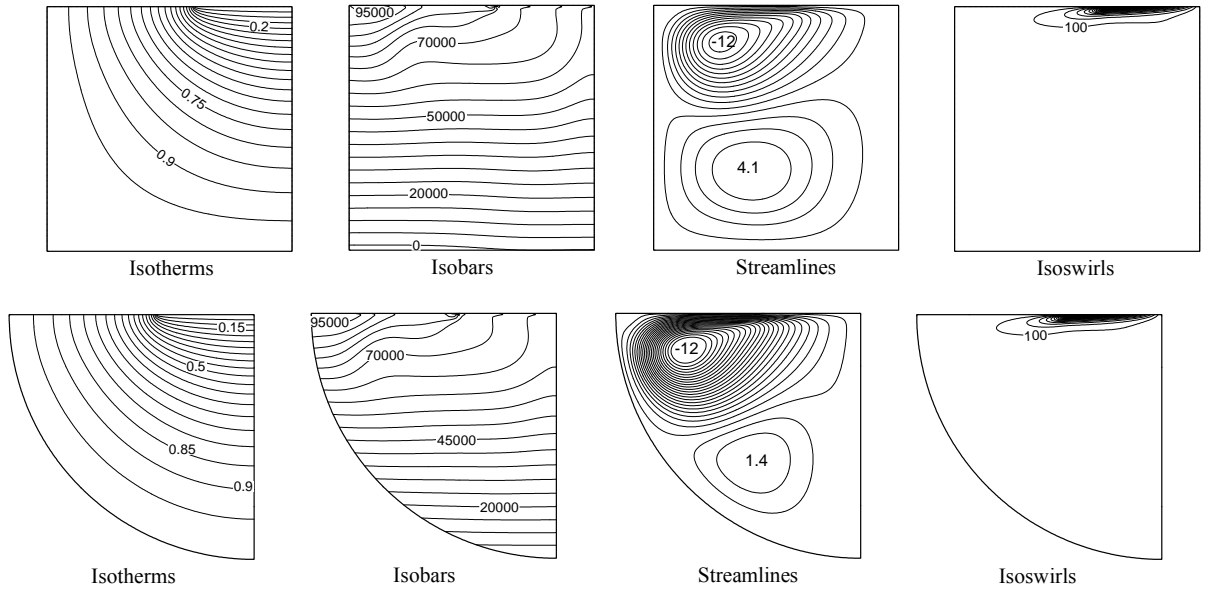


Figure 9. Isotherms, isobars, streamlines, isoswirls for $Gr=10^5$, $Ma=2000$, $Re=7000$

Unlike isotherms, the isobars undergo significant distortions under the rotation effect and their intensity increases. For $Re=3000$, 4000 and 5000 corresponding respectively to figures. 5, 6 and 7, the incline of isobars in the melt zone under the crystal is in the same phase with the presence of a second vortex under the crystal (this vortex is induced by forced convection), when this vortex moves to the free surface ($Re=6000$, figure 8) the isobars become parallel to the free surface and perpendicular to the symmetry axis.

In all studied cases we note the existence of a node in isobars at the point α (figure 2), where three phases are present simultaneously: gas (argon), liquid (silicon melt) and solid (crystal). This node, which is induced by Marangoni convection, has been noted in a precedent work [18]. By comparing streamlines and isotherms obtained for both considered geometries, we note the absence of stagnation region or region of died fluid in the hemispherical crucible and the isotherms follow the symmetrical geometry (spherical). The intensity of the natural and thermocapillary vortex is weaker comparing with the cylinder.

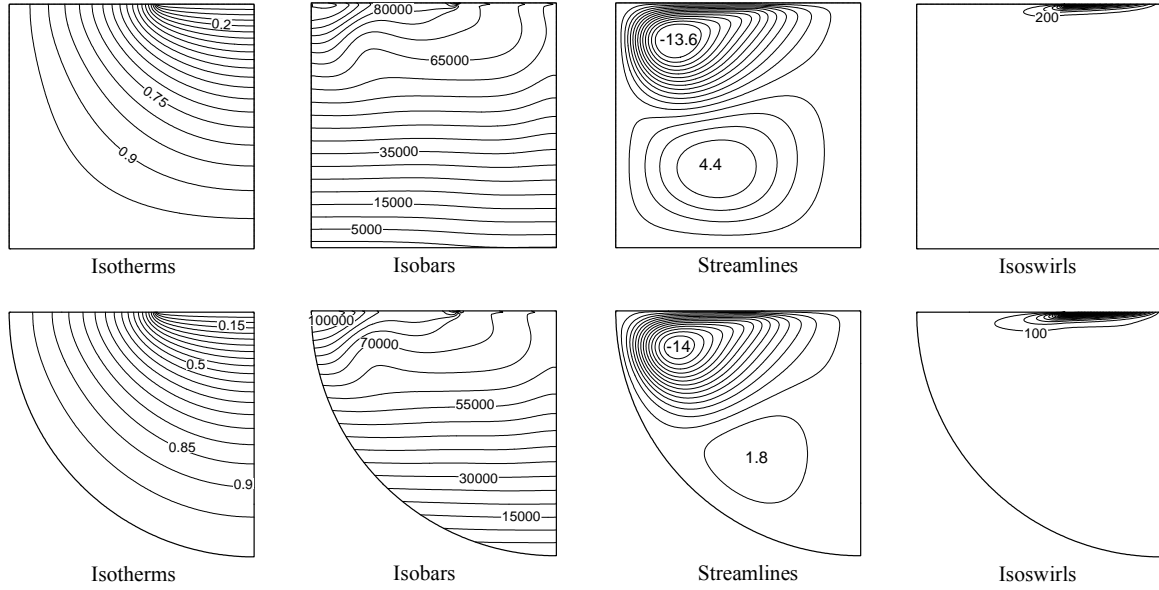


Figure 10. Isotherms, isobars, streamlines, isoswirls for $Gr=10^5$, $Ma=2000$, $Re=8000$

For the Reynolds number value $Re=1000$ corresponding to figure 3 the forced vortex appears in the hemispherical crucible but it is absent in the cylinder. We have showed in a precedent work [18] that the Richardson number $Ri = Gr/Re^2$ corresponding to the transition from natural mode to forced mode is more important for the cylinder when the Marangoni effect is not considered. The presence of the forced vortex is also accompanied with the deformation of isobars especially in the part of melt under the crystal with increasing the pressure everywhere in the melt.

The contour lines of azimuthal velocity occupy more space in the hemispherical crucible than in the cylindrical one. In figure 5 corresponding to Reynolds number value $Re=3000$, a small node is formed in the contours of azimuthal velocity in the lower part of the melt, beyond this Reynolds number value a dynamic boundary layer forms at the crystal bottom in both geometries, it is the Ekman layer [10]. But the interaction of the Ekman layer on the melt flow is still unknown.

For $Re=6000$ (figure 8), the forced vortex of the hemispherical case is bigger than the one in the cylindrical crucible, it remains small traces of the natural and thermocapillary vortex, thus we can conclude that the hemispherical geometry helps us to better control the melt flow in the traditional one even with thermocapillary convection.

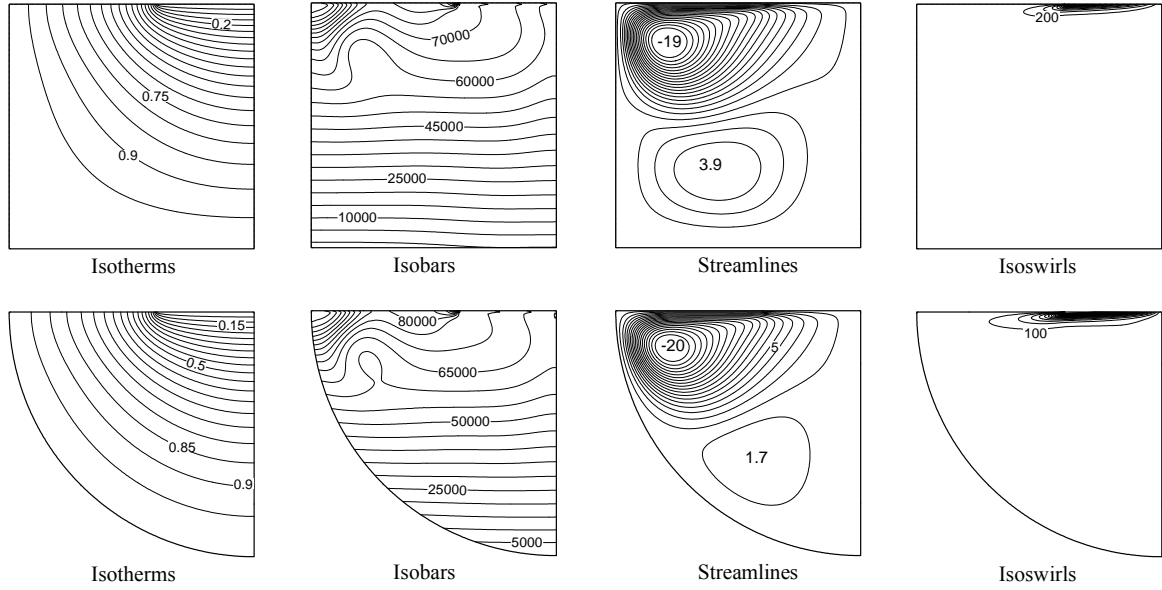


Figure 11. Isotherms, isobars, streamlines, isoswirls for $Gr=10^5$, $Ma=2000$, $Re=10000$

Beyond $Re=6000$, the center of the forced vortex begins to move toward the crucible wall, its intensity increases more and more, the natural and thermocapillary vortex begins to rebuild but remaining always in the lower part of the melt. The movement of the center of forced vortex is accompanied with the deformation of isobars in the upper part of the melt under the free surface.

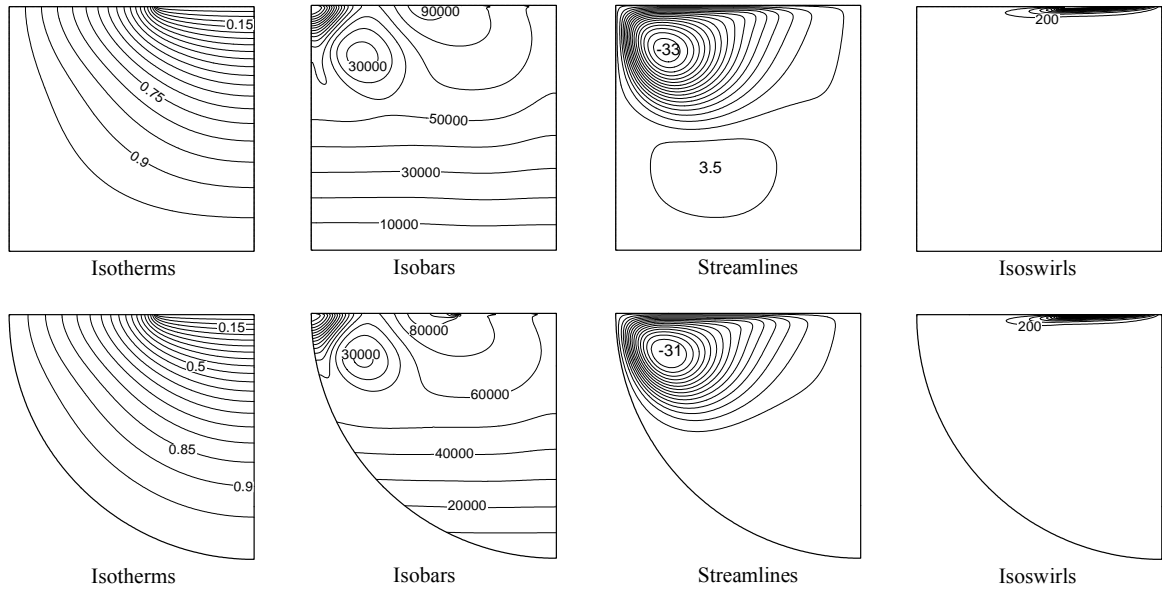


Figure 12. Isotherms, isobars, streamlines, isoswirls for $Gr=10^5$, $Ma=2000$, $Re=15000$

According to Kobayashi S [20] oxygen is the major impurity in Czochralski silicon crystals, the oxygen comes from a continuous corrosion of the walls of the silica crucible by molten silicon [10]. This oxygen is transported by the natural vortex, one part is evaporated at the free surface if there is a contact between this vortex and the free surface, the remaining part is transferred to the forced vortex. The latter assigns it to the crystal by intermediate of the growth interface. Thus the contact area between both vortices has an important role and can provide some information about the quality of the grown crystal.

For high Reynolds number values, $Re=7000$, 8000 and 10000 (figures. 9, 10, 11), the natural vortex in the cylindrical crucible is more intense and more voluminous than the one in the hemispherical crucible. For $Re=15000$ (figure 12), the natural vortex disappears in the hemispherical crucible but it remains in the cylindrical one. Thus we establish that the contact area between the two vortices is greater in the cylindrical system compared to the hemispherical one, this difference between the two geometries gives more advantage for the modified geometry because the more the contact area is smaller less impurities can be transferred to the forced vortex and then to the crystal.

4. Conclusion

Under the effect of natural, thermocapillary and forced convection, we tried to show the effect of crucible geometry on temperature, velocity and pressure fields in silicon melt of a Czochralski crystal growth system. The analysis of the obtained results enables us to conclude the advantage of the modified geometry in comparison with the traditional one. The absence of the stagnation region or region of died fluid in the hemispherical crucible corner and the facility to control the melt flow, by using the crystal rotation, and then the pressure field favors the hemispherical crucible shape to the cylindrical one.

The pressure field is strongly related to the swirl velocity, a clockwise vortex induced by forced convection is present in the melt. The distortion of isobars follows the location, the shape and the center position of this vortex. For low Reynolds values ($Re < 6000$), the forced vortex is situated under the crystal, the isobars deformation or their inclination is also situated under the crystal. For high Reynolds number values ($Re > 6000$) the forced vortex is under the free surface and the isobars deform in the same region i.e. under the free surface and their deformation follows the movement of the forced vortex center. The obtained results for high rotation rate values enable us to note that the contact area between the natural and forced vortexes is small in the hemispherical crucible in comparison with the cylindrical one. Consequently this helps us to minimize impurities in the grown crystal.

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