Three-dimensional modelling of thermal stress in floating zone silicon crystal growth

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Abstract.

During the growth of large diameter silicon single crystals with the industrial floating zone method, undesirable level of thermal stress in the crystal is easily reached due to the inhomogeneous expansion as the crystal cools down. Shapes of the phase boundaries, temperature field and elastic material properties determine the thermal stress distribution in the solid mono crystalline silicon during cylindrical growth. Excessive stress can lead to fracture, generation of dislocations and altered distribution of intrinsic point defects. Although appearance of ridges on the crystal surface is the decisive factor of a dislocation-free growth, the influence of these ridges on the stress field is not completely clear. Here we present the results of thermal stress analysis for 4" and 5" diameter crystals using a quasi-stationary three dimensional mathematical model including the material anisotropy and the presence of experimentally observed ridges which cannot be addressed with axis-symmetric models. The ridge has a local but relatively strong influence on thermal stress therefore its relation to the origin of fracture is hypothesized. In addition, thermal stresses at the crystal rim are found to increase for a particular position of the crystal radiation reflector.

1. Introduction

The industrial floating zone (FZ) method for the growth of silicon single crystals provides semiconductor material of lowest impurity concentration which is mainly used for high power devices. The main parts of the process inside the growth chamber are the cylindrical poly crystalline silicon feed rod being pushed downwards, the single turn high-frequency induction coil to melt the feed rod through its needle-eye opening, the molten zone just below the inductor and the growing crystal which supports the molten zone. The crystal is axially rotated therefore cylindrically shaped ingot is obtained and crystallization interface is normally concave because the latent heat at the centre can not be removed as easily as at the periphery close to the radiating crystal surface. The non-flat crystallization interface and the thermal radiation from the surface gives a radial component of temperature gradient resulting in an inhomogeneous expansion and possibly high levels of thermal stress. The consequences of the stress with its considerable influence on the process and product quality can be divided into three categories:

- Fracture of the crystal when the stress is above some critical level. The sudden and unpredictable fracture leads to a complete material loss and a damage to the growth equipment due to the spillage of the melt. Risk of the fracture increases with crystal diameter.
• Dislocation generation, the probability of which correlates to the stress level and also leads to a process failure in a less destructive manner. Large diameter single crystals can not be grown with dislocations [1].

• Altered incorporation of the intrinsic point defects as thermal stress influences the Voronkov’s criterion, the critical ratio \( v/G \) or \( \Gamma_{0_{\text{crit}}} \). Under compressive stress the \( \Gamma_{0_{\text{crit}}} \) tends to decrease and vice versa [2].

With the needle-eye technique, the capillary limitation of a cylindrical zone is surmounted. However, currently the ingot diameter is limited to 200 mm with the two main impediments being the crystal fracture due to thermal stress with related dislocation generation and electrical breakdown at the inductor main slit [3].

One of the first reported calculations of thermal stress in the silicon crystal during FZ growth was part of a global axis-symmetric quasi-steady model including electromagnetic field and heat transfer for a 4” crystal diameter [4]. Analytic approach for the stress was used and various inductor designs were compared with the aid of the model. Maximum compressive stress at the centre of the crystallization interface and a smaller maximum of tensile stress on the crystal surface at or close to the triple point was found and the scalar von Mises stress was also used as a measure.

3D anisotropic model of thermal stress for 6” and 8” crystal diameters using the Czochralski (Cz) method predicted dislocation generation in all of the considered cases [5]. Using the knowledge of the discrepancy between predictions by existing models and experimental observations, a concept of a metastable state of the crystal where the energy necessary to generate dislocations is small compared to the total energy of the crystal has been introduced [6]. The higher total energy of the crystal the smaller is the energy barrier for dislocation generation and random perturbations can lead to dislocation generation more easily. The idea supported the results of axis-symmetric finite element simulations for 4” FZ crystals where exceed stress was used to predict dislocations in a crystal region spanning from the crystallization interface until approximately a radius below. Reduction of the thermal stresses with the introduction of a crystal radiation reflector was shown as well. Global model of FZ system which includes axis-symmetric stress field using Comsol Multiphysics has been developed at the Leibniz Institute for Crystal Growth [7]. The case study revealed that higher pull rate leads to an increase of thermal stress, and presence of the reflector decreases the stress. The above-mentioned studies all consider a perfectly round crystal in contrast to a numerical study of sinusoidal surface variations for a Cz crystal based on experimental observations [8]. Thermal stress was shown to locally increase in any of the considered configurations of the periodical surface undulations. The problem of thermal stress in the Cz crystal has been further addressed by development of a three-dimensional (3D) model including the uneven surface and material anisotropy [9]. The \( \langle 1 1 1 \rangle \) crystal growth orientation was found to be of the lowest stress level.

However, experimentally observed ridges on the crystal surface [10] which can not be studied by axis-symmetric models are the main indication for a dislocation free growth. Morphology of the ridges for FZ crystals of \( \langle 1 1 1 \rangle \) orientation up to 5” diameter has been described and a loss of particular ridge shapes when the crystal dislocates is suggested [11]. Size of the ridge has been theoretically calculated for a \( \langle 1 0 0 \rangle \) 4” Cz crystal depending strongly on the temperature gradient by the crystallization interface at the triple point [12]. The model predicts larger ridge size for a smaller temperature gradient. Formation of periodic undulations of the ridge – also called bulges – are observed for \( \langle 1 1 1 \rangle \) crystals due to the inhomogeneous growth rate depending on the crystallographic orientation [13]. The necessary undercooling of the melt in the \( \langle 1 1 1 \rangle \) direction is the slowest and will move the triple point lower thus causing the free melt surface to bend outwards and the bulge to grow. Once the bulge has started to grow, the radiation heat losses from the increased surface area will provide enough undercooling for the triple point line to rise and bulge to decrease. For \( \langle 1 0 0 \rangle \) orientation mostly steady state growth of straight
ridges is observed due to the different angle of \{1 1 1\} planes. The period of these oscillations depends on the physical conditions at the triple point line during the process which determine the temperature gradient.

Numerical simulation of the crystal ridge was first carried out by [14] for a 4” FZ \langle 1 1 1\rangle crystal. However, the standard mesh was used without refining the ridge area which can have a considerable influence of the precision of the numerical results. The mathematical model developed and shown in [14] is used for the present calculations. The same numerical procedures apply except for mesh refinement by the ridge. Smooth crystal shape is compared to surface with a ridge for 4” and 5” crystal diameters using two different pull rates and \langle 1 0 0\rangle and \langle 1 1 1\rangle crystal orientations and an undulated feature of the ridge is also introduced as reported in [15]. The crystal radiation reflector is considered for 5” crystal with 2.5 mm/min pull rate. The given modelling approach is suitable for quasi-steady conditions which correspond to the cylindrical growth phase.

2. Numerical model

2.1. Boundary conditions

To have realistic interface shapes and temperature field in the crystal, initially a converged state of quasi-steady calculations using the program FZone [16] with inductor shape from [7] is obtained for the given crystal diameter and the pull rate. Shape of the crystallization interface is found by solving global heat transfer problem which couples the temperature field in the silicon with radiation on the surfaces using view factors, and the heat sources are taken from 3D high-frequency electromagnetic field calculation [17]. The inductor current is adjusted to provide the target zone height, i.e., the vertical distance between the triple point and rim of the melting front. With crystal radiation reflector present, temperature in the reflector is calculated using emissivity and heat conductivity of the specified material.

2.2. Mesh generation and 3D geometry

A quasi-structured mesh of second order elements is used to solve the heat transfer and linear elasticity problems. First, a unit circle is meshed symmetrically with radial layers of triangular or quadrilateral elements. Second, the converged boundary shapes from the axis-symmetric FZone calculation are rotated to obtain the initial crystal domain in 3D. Then, the 2D mesh is mapped onto the crystallization interface and successive layers of the respective prism and hexahedron elements are added along the vertical and almost parallel to the crystallization interface with the desired discretization level.

Mesh for the ridge is refined in the area spanning four quadrilateral elements in the azimuthal direction and two – in the radial direction to have twice the elements on the crystal surface covering the ridge to capture any rapid changes of the temperature and the strain field, see Fig. 1. Shape of the ridge on the triple point line is approximated by analytical formula:

\[ R(\phi) = R_0 + A e^{- \left( \frac{\phi}{\phi_0} \right)^2} \]  \hspace{1cm} (1)

where \( \phi \) is the azimuthal angle, \( R_0 \) is the initial crystal radius, \( A = 1 \) mm is the amplitude of the ridge and \( \phi_0 = 5^\circ \) is the effective width of the ridge.

Periodic vertical undulations are introduced to the ridge by

\[ R(z) = R_0 + \frac{R - R_0}{2} \left( 1 - \sin \frac{z - z_0}{l} 2\pi \right) \]  \hspace{1cm} (2)

where \( l = 15 \) mm is the period of the undulations and \( z_0 \) is vertical shift. The size parameters are taken from the crystal surface photographs in [15], see Fig. 2 for the corresponding numerical model with two ridge shapes considered.
2.3. Temperature field

Heat conduction equation with temperature dependent heat conductivity $\lambda$ is solved in the crystal:

$$\nabla \left( \lambda(T) \nabla T \right) = 0$$

(3)

Constant temperature boundary condition is applied to the crystallization interface and distribution of the apparent ambient temperature for the radiation boundary condition is applied on the crystal surface. This temperature is found from the initial results by FZone using formula:

$$T_{amb} = 4\sqrt{T^4 - \frac{q_{rad}}{\epsilon\sigma_{SB}}}$$

(4)

where $T = T(z)$ is the temperature on the crystal surface, $q_{rad}$ is the net radiation heat flux, $\epsilon = \epsilon(T)$ is the emissivity and $\sigma_{SB}$ is the Stefan–Boltzmann constant.

2.4. Thermal stress field

Linear elasticity problem based on the stress-strain relationship (Hooke’s law) is solved:

$$\sigma_i = C_{ij} (\epsilon_j - \epsilon_{Tj})$$

(5)

where $\epsilon_{T}$ is isotropic thermal strain and $C$ is the elasticity matrix. For $\langle 100 \rangle$ crystal growth orientation, the elasticity matrix can be expressed as:

$$C = \begin{bmatrix}
C_{11} & C_{12} & C_{12} & 0 & 0 & 0 \\
C_{12} & C_{11} & C_{12} & 0 & 0 & 0 \\
C_{12} & C_{12} & C_{11} & 0 & 0 & 0 \\
0 & 0 & 0 & \text{Sym.} & C_{44} & 0 \\
0 & 0 & 0 & C_{44} & 0 & \text{Sym.} \\
0 & 0 & 0 & 0 & C_{44} & 0
\end{bmatrix}$$

(6)

And the respective expression for $\langle 111 \rangle$ orientation:

$$C = \begin{bmatrix}
C_{11} - K/2 & C_{12} + K/6 & C_{12} + K/3 & -\sqrt{2}K/6 & 0 & 0 \\
C_{11} - K/2 & C_{12} + K/3 & \sqrt{2}K/6 & 0 & 0 & 0 \\
C_{11} - 2K/3 & 0 & 0 & \text{Sym.} & C_{44} + K/3 & 0 \\
0 & 0 & 0 & C_{44} + K/3 & 0 & 0 \\
0 & 0 & 0 & 0 & C_{44} + K/3 & -\sqrt{2}K/6 \\
0 & 0 & 0 & 0 & C_{44} + K/3 & 0
\end{bmatrix}$$

(7)
where $K = C_{11} - C_{12} - 2C_{44}$. Experimental measurements of the Young’s modulus are available [18] and the corresponding temperature-dependence of the elastic constants has been implemented. The final equation for the problem is the force balance:

$$\frac{\partial \sigma_{ij}}{\partial x_j} = 0.$$  

(8)

3. Calculation results

Comparison of the FZone results between 5” crystal diameter cases with and without reflector in Fig. 3 show the shapes of phase boundaries and the vertical distribution of the apparent ambient temperature on the crystal surface calculated from equation 4. Depth of the crystallization interface increases using the reflector because of the significantly higher apparent ambient temperature and therefore lower heat losses from the crystal surface. The physical parameters of the heat transfer are temperature of the ambient space 600 K, temperature of the inductor 400 K and emissivities of the inductor and the reflector 0.3. Average temperature of the reflector 1150 K is obtained without any cooling and is close to estimated 1000 K for a realistic FZ process [16]. However, water cooling would be preferable in order to avoid deformation of the silver reflector near its melting point. With larger depth of the crystallization interface and therefore smaller interface angle, in general an increase of thermal stress is expected.

![Figure 3. Comparison between FZone results of axisymmetric heat transfer for cases of 5” crystal diameter without reflector (blue lines) and with reflector (red lines). Shapes of phase boundaries with inductor given in black (left) and the apparent ambient temperature on the crystal surface and temperature of the reflector (right).](image)

Von Mises stress invariant is commonly used as a universal measure for the stress impact on dislocation generation and fracture [3]. Influence of the ridge and process parameters on von Mises stress on the crystal surface is shown in Fig. 4 where two values of pull rate, two crystal orientations and two radii are compared. For all the cases stresses in 5” crystal are higher that in 4” crystal by roughly 20 MPa. The azimuthal variation of stress due to the ridge is considerably stronger than due to the material anisotropy. Stress maximum without the ridge can be located on the triple point line as in cases (c, d) with high pull rate and below the triple point line as in cases (a, b) that have lower pull rate. There is a sudden decrease of the stress on the ridge which is always accompanied by a pronounced stress maximum just by the side of the ridge. The maximum is located mostly up to 5 mm below the triple point line (a, c, d) but can reach 20 mm as in (b). Stress on the triple point line is approximately 10 MPa lower than the maximum by the side of the ridge below the triple point line. Larger azimuthal variations of von Mises stress along the triple point line are observed for case with the reflector as shown in Fig. 5. Stresses for a round $\langle 1\bar{1}0 \rangle$ crystal have a four-fold symmetry with relatively low variations compared to $\langle 111 \rangle$ orientation with a more pronounced three-fold symmetry of von Mises stress. Slight
increase of the stress by the side of the ridge is noticed for all cases except for the undulated ridge which gives largest stress at the tip of the ridge. This peak exceeds the maximal value for case without ridge by 1 MPa. For \(\langle 1 1 1 \rangle\) crystals the stress minimum at the ridge is slightly smaller than for \(\langle 1 0 0 \rangle\) crystals although the stress is larger elsewhere.

![Figure 4. Distribution of von Mises stress on the crystal surface for different orientations and pull rates with crystal diameters of 4" (left) and 5" (right). Comparison between crystals without and with ridge. \(z = 0\) corresponds to the triple point. Unified colour scale for all of the cases with 5 MPa interval between isosurfaces.](image)

Literature data suggest that cleavage in silicon crystals occurs along the \{1 1 1\} slip planes [19]. These planes for \(\langle 1 0 0 \rangle\) and \(\langle 1 1 1 \rangle\) crystal orientations are illustrated in Fig. 6 where the angle between the planes and the vertical is 54.7° for \(\langle 1 0 0 \rangle\) and 70.5° as well as 0° for \(\langle 1 1 1 \rangle\)
orientation. For a quantitative measure, the normal stress on the slip planes, $\sigma_n = \sigma_{ij} n_i n_j$, is examined. Distribution of the normal stress on 5” crystal with pull rate $v = 2.5$ mm/min in Fig. 7 presents all of the possible cases for the two different crystal orientations depending on the slip plane direction. Higher normal stress for ⟨111⟩ compared to ⟨100⟩ crystal orientation is found. The stress is significantly reduced on the ridge. Strongest stress maxima are found on the crystal surface besides the ridge which exceed the values for crystal without ridge. Fracture of the crystal is the least likely to happen along the horizontal slip planes for ⟨111⟩ crystal as the normal stresses on these planes are at least an order of magnitude lower than for the other directions of the slip planes.

Additionally, the shear stress $\tau = \sqrt{(\sigma_{ij} n_j)^2 - \sigma_n^2}$ on the {111} slip planes can be attributed to dislocation generation. Azimuthal plots of the normal and shear stresses on the slip planes along the triple point line are given in Fig. 8 with the same order of slip plane directions as in Fig. 7. These stress projections exhibit more non-uniform behaviour compared to von Mises stress. Shear stresses are smaller than the normal stresses except for the horizontal slip planes with negligible stress. Considerable difference in the magnitude of the stress is between 4” and 5” diameter crystals. The vertically undulated ridge in the ⟨111⟩ case increases the normal stress at its peak.

Intrinsic point defects – vacancies and self-interstitials – are formed during the crystal growth which in turn lead to grown-in defects affecting the crystal quality. A criterion of $(v/G)$, where $v$ is the growth rate and $G$ is the axial temperature gradient, determines the majority species
Figure 7. Distribution on the normal stress on \{111\} slip planes for a 5” diameter crystal with pull rate \(v = 2.5\) mm/min without ridge (top) and with ridge (bottom) for two different growth orientations. Arrow denotes position of the ridge. Plane normals from left to right: \((\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}})\) or \((-\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}})\), \((\frac{1}{\sqrt{3}}, -\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}})\) or \((-\frac{1}{\sqrt{3}}, -\frac{1}{\sqrt{3}}, \frac{1}{\sqrt{3}})\), \((-\frac{2\sqrt{3}}{3}, \frac{2}{3}, 1)\), \((-\frac{2\sqrt{2}}{3}, 0, -\frac{1}{3})\), \((0, 0, 1)\).

Figure 8. Azimuthal distribution of the normal stress (top) and the shear stress (bottom) on the \{111\} slip planes along the triple point line for 4” crystal (blue lines) and 5” crystal (red lines for straight ridge and black lines for ridge with undulations) with pull rate \(v = 2.5\) mm/min. \(\langle 100 \rangle\) orientation with the ridge at 45° and \(\langle 111 \rangle\) orientation with the ridge at 60°. Plane normals in the same order as in Fig. 7.

According to [20]. A critical value of \((v/G)\) separates the interstitial-rich and vacancy-rich growth modes. Influence of the thermal stress on the \((v/G)_c\) has been shown [2]. Kulkarni parameter set [21] is used to calculate the critical value:

\[
(v/G)_c = 1.5582 \times 10^{-1} - 1.48 \times 10^{-3} p + 10^{-5} p^2
\]

where units for \((v/G)_c\) are mm\(^2\)/min/K and \(p\) is the pressure in MPa. See Fig. 9 for radial distribution on the crystallization interface of ratio of the critical value with and without the stress effect. When thermal stress is included in the calculation of the \((v/G)_c\), the crystal tends to be more vacancy rich from the centre until 80% of the radius value and more self-interstitial rich in the outer part. The results for each case slightly differ between the two crystal orientations at
the centre and become almost equal at the triple point. The relative changes of the \((v/G)_c\) are up to 30% of the value without stress effect taken into account in both directions of the constant value. The local effect of ridge is observed only in the last 2 mm before the triple point.

![Figure 9](image_url)

**Figure 9.** Normalized radial distribution of ratio between the critical values of \(v/G\) with the actual stress influence and with zero stress on the crystallization interface for \(\langle 1\ 0\ 0 \rangle\) orientation on the left and \(\langle 1\ 1\ 1 \rangle\) on the right. The dashed and solid lines correspond to cases without and with the ridge respectively.

4. Conclusions
Considering that ridges on the crystal surface are an essential part of dislocation-free growth, the present study shows the possible influence of the ridge on thermal stress for different process parameters. A ridge on crystal surface has been resolved by refining the mesh locally and calculations using the 3D anisotropic elasticity model reveal strong influence within the perturbed region. The stresses always decrease at the top of a straight ridge and have symmetric maxima on the sides but can also increase at the centre of the ridge with undulations. The stress projections on the slip planes have been obtained and show larger normal stresses compared to the shear stresses by approximately 8 MPa for \(\langle 1\ 0\ 0 \rangle\) and 20 MPa for \(\langle 1\ 1\ 1 \rangle\) orientation. Sides by the ridge are critical spots for initiation of dislocation generation and fracture as the stresses there can exceed the values for a perfectly round crystal by roughly 5 MPa (up to 8%). With the chosen reflector configuration, the thermal stress is larger compared to the standard case by 6 MPa on average and also azimuthal variations increase. Although the ridge has a local influence, the azimuthal span of the affected region on the crystal surface near the triple point line is approximately 30°. The ridge is shown to decrease the critical \(v/G\) on the crystallization interface within few millimetres from the triple point but the change is smaller compared to the overall changes due to the stress effect which range from 0.7 at the centre to 1.3 at ETP times the level without stress.
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