Investigation on influence of dimensionless numbers in molten silicon during multi-crystalline silicon growth process

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Multi-crystalline silicon is key material with advantages of low-production cost and high conversion efficiency of PV solar cells. The paper deals with the numerical investigation of melt flows during directional solidification(DS) of silicon growth process. The simulation is made in two dimensional axi-symmetric model by the finite-element method. The melt flow pattern in the crucible has significant effects on the formation of defects and distribution of impurities concentration in the grown crystals**.** The study of fluid dynamics based dimensionless numbers is used to control the melt flow pattern for optimizing the DS process and improving ingot quality. The influence of the axial velocity field, vorticity, diffusive heat flux and convective heat flux on the molten silicon were investigated for various Prandtl numbers between 0.001 to 2 with the help of numerical technique. In this paper fundamental thermodynamic phenomena were simulated and analysed for the two different Prandtl numbers at constant Rayleigh number Ra=1000. The aim is to increase the grain size and reduce dislocation density through control of the melt flow pattern. The numerical results also provide a basic understanding of the heat transfer characteristics during directional solidification of silicon growth process.

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1. Introduction

The global increase of energy consumption leads to increasing environmental problems caused by greenhouse gases. It can only be resolved if the power generation from renewable energy sources can be strongly raised in the future. The electric power generation by conversion of sun light via solar cells is one of the assuring options of renewable energy sources. At present, the dominating semiconductor for PV solar cells is multi-crystalline and mono-crystalline silicon [1]. Currently, around 90% of industrial solar cells are made from silicon ingots. The photovoltaic market has increased remarkably in recent years, and mc-silicon has a market share of more than 60% in all photovoltaic materials. Multi-crystalline silicon is an important material with advantages of low-production cost and high conversion efficiency of solar cells [2, 3]. The photovoltaic industry is a player in the renewable energy segment, and the electricity generation from photovoltaics solar cells is deemed to be one of the key technologies of the 21st century[4]. The multicrystalline silicon (mc-Si) is the predominant material for solar cells, which is still relatively poorly understood material, even after 30 years of research. Numerical simulation as a main tool in modern crystal growth technology is used for promotion of bulk crystal growth processes. Simulation of fluid flows in crystal growth processes has an important research area in theoretical and applied mechanics. Theoretical modelling has played an important role in developing technologies used for growing multi-crystalline silicon for high performance solar cells[5]. The application of solar cell devices requires large diameter crystals with a large grain size, low defect density and uniform dopant distribution. Directional crystallization of mc-Si ingots is the most cost effective technique for the production of silicon wafers in the PV industry. As crucible becomes larger, the effect of fluid flow and thermal field on ingot quality, such as dislocation density and dopant uniformity, becomes more significant. Besides making large ingots, furnace manufacturers are also looking into new direction that can help increase the growth rate and improve the quality of the ingot[6,7]. The thermal field and stress, formation and distribution of impurities, and dislocation density in a grown mc-Si ingot are directly affecting the crystal quality. These properties have direct effects on the solar cells efficiency [8]. Bellmann and Vizman et al., reported the effects of thermally driven flow and interface shape on impurity distribution during directional solidification of silicon[9,10].

In the past three decades, many works have been devoted to the numerical analyses of various melt crystal growth processes using various models. Most of these models can be generally divided into local models and global models. However, the melt flow in a crucible is usually conducted as a local two-dimensional (2D) axi-symmetric model when using a square crucible in the directional solidification system. Therefore, a 2D thermal and fluid flow analysis of a molten domain in growth system is necessary to gain a better understanding of growth phenomena and insight into the physics of growth processes. In this work, the melt convection due to the temperature difference of melt for various Prandtl numbers

is discussed in the time -independent model with the Newtonian incompressible Navier-Stokes equation in Boussinesq approximation and the convection-conduction equation. The computations were performed in a 2D axi-symmetric model by the finite-element numerical technique. The influence of the Prandtl number at various ranges in molten silicon is discussed during multi-crystalline silicon growth process. The diffusive heat flux, X- velocity field, vorticities and convective heat flux on the silicon melt are simulated and analysed for various Prandtl numbers between 0.001 to 10 at constant Rayleigh number 1000 with the help of numerical technique. Finally here, two Prandtl numbers are determined and simulation results of various fluid flow properties are given for those two Prandtl numbers 0.001 and 2 and simulated for controlling the melt flow pattern. The numerical results also provide a basic understanding of the heat transfer characteristics during directional solidification of silicon growth process. The simulation results indicate a possible way to control the melt flow pattern and improve the crystal growth process for mc-Si ingot.

2. DS model description

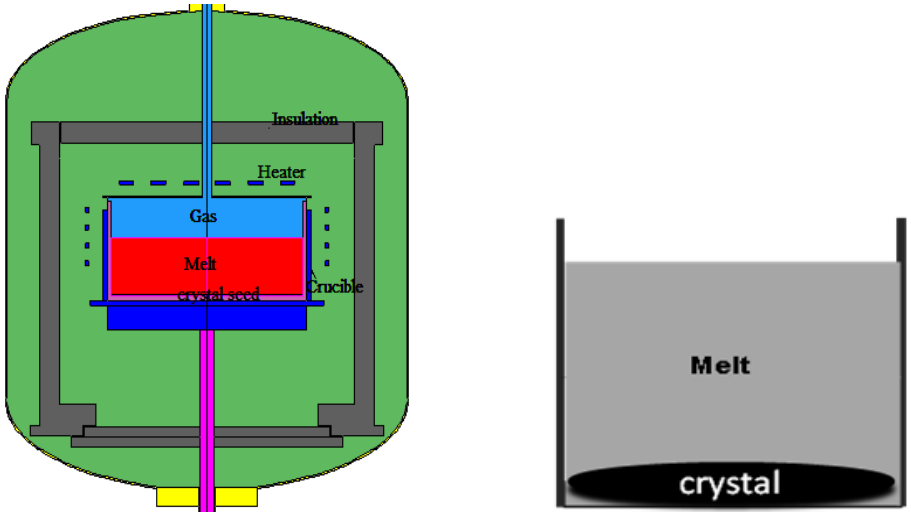

Fig. 1. (a) Schematic diagram of the directional solidification system (b) Physical system(melt)

The schematic diagram of industrial-scale DS system is shown in Fig. 1(a). The system is used for growing mc-Si ingots traditionally for solar cell application. The DS system mainly consists of silicon nitride (Si_3N_4) coated silica crucible, graphite susceptor, gas tube, heat exchange block, graphite resistance heater, insulations, chamber wall. The silicon feed material is loaded into a silica crucible. The crucible walls are supported by graphite susceptors to avoid deformation at high temperature. The furnace is well sealed with a water-cooled wall and operates at a low pressure. The inert argon gas is used for purifying the growth environment in the system. A seeded DS process for growing mono like multi-crystalline ingot includes two major phases: the seed preservation phase and the bulk crystal growth phase. At the seed preservation phase, the mono-crystalline silicon seeds should be preserved (not totally melted) by accurately adjusting the heating power. Then, with the bottom insulation moving downwards, the temperature at the seed-melt interface is decreased below the melting point with the increase in the radiation heat loss from the gap between the side and bottom insulations. The mono-crystalline silicon seeds begin to grow upwards and the bulk crystal growth phase starts. During the bulk crystal growth phase, a suitable growth temperature gradient is maintained by adjusting the heating power and slowly moving the bottom insulation downwards [11]. But considering the axi-symmetry of the problem, the resolution of the equations is done in a two-dimensional(2D) rectangular–shaped silica crucible with molten silicon. The computational domain considered for the present analysis is shown in Fig.1(b). Thermal boundary conditions are applied on the inside wall of crucible. Table .1 shows thermo-physical properties of silicon(liquid) [12, 13].

Table 1. Thermo- physical parameters of silicon (liquid).

Symbol and Description		Values (units)
β	heat expansion coefficient	5.5×10^{-6} (K^{-1})
γ	surface tension	0.74 (N/m)
$d\gamma/dT$	rate of change of surface tension	-2.8×10^{-4}(N/m.K)
ρ	density	2530 (kg/m^3)
C_p	specific heat capacity	1040 (J/kg.K)
T_{melt}	melting point temperature	1685 K
η	dynamical viscosity	7×10^{-4} (kg/ ms)
λ	heat conductivity	67 (W/m.K)
σ	electrical conductivity	1.2 ×106(S/m)
L	latent enthalpy	1.8×10^{6} (J/kg)
ΔT	temperature difference	10 K

3. Mathematical equations

The transport of heat, mass and momentum is especially essential in bulk crystal growth processes. Flow in the molten phase is indispensable for transport of heat and mass convection in bulk crystal growth systems. The convection induced by temperature gradients in the silicon melt system is analysed in the time independent model. The top and bottom melt surfaces are thermally insulated and the left and right silicon nitride coated quartz walls are maintained at isothermal constant temperature difference (10K). The flow field is assumed to be steady and incompressible. By incompressibility it is meant that the variation in density due to temperature differences is neglected except where they modify the body force term in the momentum equation. This is generally referred to as Boussinesq approximation. The governing equations for the melt region are described by coupled Navier-Stokes and energy equations with solidification. The Boussinesq approximation is used to account for buoyancy force in momentum equation via temperature dependence of density in gravity term [13, 14,15]

Assuming axi-symmetric conditions the differential equations governing steady-state, incompressible fluid flow are given as follows. Continuity equation

$$\nabla.\vec{u} = 0 \tag{1}$$

Navier-Stokes equation

$$-\nabla.\eta[\nabla\vec{u} + (\nabla\vec{u})^T] + \rho\vec{u}.\nabla\vec{u} = -\nabla P + F \tag{2}$$

Navier- Stokes equation with Boussinesq approximation

$$-\nabla.\eta[\nabla\vec{u} + (\nabla\vec{u})^T] + \rho\vec{u}.\nabla\vec{u} = -\nabla P + \rho g\beta(T - T_{ref}) \tag{3}$$

The first term gives the rate of momentum gain by viscous transfer, the second by convection, and the third by pressure forces, where η, ρ ,u, p, g, β ,T. are dynamic viscosity, density, velocity vector, pressure, acceleration due to gravity, thermal expansion and reference temperature respectively. F (considering only Boussinesq approximation) is force per unit volume.

The transport of heat and mass in melt flow can be determined from solution of the appropriate governing equations. The energy balance equation is given by the following expression

$$\nabla.(-k\nabla T + \rho C_P T\vec{u}) = 0 \tag{4}$$

In the above equation, the expression within the brackets is the heat flux vector, containing a conductive and a convective part, where, k, T $C_{p,}$ are thermal conductivity, temperature, heat capacity, respectively.

The Rayleigh number(Ra) denotes the ratio of buoyant to viscous forces. which is also defined as the product of the Grashof number and the Prandtl number. The buoyancy- driven convective state of a silicon melt fluid is governed by the thermal Rayleigh number

$$Ra = Gr \times \Pr = \frac{g\Delta TL^3\beta h}{\eta\rho^{-1}KCL^{-1}} \tag{5}$$

The Prandtl number(Pr) denotes the ratio of momentum and thermal diffusivities, which characterizes the similarity between temperature and velocity distributions in a configuration of similar boundary geometry, for liquid semiconductors, Pr<<1.

$$\Pr = \frac{\eta\rho^{-1}}{KCL^{-1}} \tag{6}$$

The boundaries for the Navier-Stokes equations are impermeable, no-slip conditions. The no-slip condition results in zero velocity at the wall, with pressure within the domain remaining undefined[16].

$$\vec{u}.\vec{n} = 0, \vec{u}.\vec{t} = U \tag{7}$$

$$\vec{t}.[-pI + \eta(\nabla\vec{u} + (\nabla\vec{u})^T]\vec{n} = 0 \tag{8}$$

where, $p = 2\eta(\partial\vec{u}/\partial\vec{n})$, U is scalar velocity, $\vec{t}$ and $\vec{n}$ represent the boundaries of tangential and normal vectors, respectively. Since the boundary conditions involve $\vec{u}$ alone, the pressure can be eliminated. Thus, the boundary condition expresses that the gradient velocity field along the melt free surface is balanced by the shear stress [17].

$$\eta\cdot[t_x, t_y].\begin{bmatrix} 2(\partial\vec{u}/\partial x) & (\partial\vec{u}/\partial x) + (\partial\vec{v}/\partial y) \\ (\partial\vec{u}/\partial x) + (\partial\vec{v}/\partial y) & 2(\partial\vec{v}/\partial y) \end{bmatrix}\cdot\begin{bmatrix} n_x \\ \vec{n}_y \end{bmatrix} = (\partial\gamma/\partial T).[\partial T/\partial x + \partial T/\partial y] \tag{9}$$

Here the sign $d\gamma/dT$ generally depends on the material, with downward flow ($d\gamma/dT$ <0) or upward flow ($d\gamma/dT$ >0) of the free liquid on Si melt surface. The material (Si melt) parameters used in the mathematical model for the considered molten region of DS system are given in Table 1.

4. Numerical method

The governing equations and the boundary conditions for the fluid flow and heat transfer characteristics in the molten Si system (Fig. 1) are solved numerically using finite element method in which the calculation domain is discretized as triangular element into a finite number of elements. The continuous physical model is divided into finite pieces called elements and laws of nature are applied on the generic element and the results are then recombined to represent the continuum. The computations are made using the two-dimensional(2D) axi-symmetry hypothesis for inner cell silicon melt of rectangular crucible. The time independent Newtonian incompressible Navier-Stokes model for fluid flow, heat and mass transfer are solved using the finite-element numerical technique. The iterative process is tuned for a fast, efficient solution using

nondimensional parameters and a Boussinesq term for the buoyant drive with the incompressible Navier-Stokes equation and the convection and conduction application modes. According to the considered geometry, 119116 triangular elements are partitioned as sub domains. Standard no-slip and zero mass flux conditions are used on the boundaries. The numerical solutions are carried out to obtain the heat and flow charcterestrics for more than 25 Prandtl numbers. Anticipating the existence of thin hydrodynamic boundary layers, the performed mesh is non-uniform, much thinner along the walls (0.0025mm) than in the middle of the melt system (0.011 mm). The dimensionless numbers play a vital role to study the heat transfer mechanism in liquid, which are calculated for molten silicon system using finite element code for various Prandtl numbers at constant Rayleigh number.

5. Simulation results and discussion

Multi-crystalline Silicon is an important semiconductor substrate for manufacturing solar cells. The mechanical and electrical properties of multi-crystalline silicon (mc-Si) are primarily influenced by the quality of the feedstock material and the crystallization process. In this work, numerical calculations, applying finite element analysis (FEA) are presented, in order to simulate thermo physical properties during the crystallization process of industrial size mc-Si ingots. Electrical material properties of the resulting wafers strongly depend on various parameters of the ingot casting. And also mechanical properties of the material are influenced by the crystallization process. For example, changes in temperature distributions during the crystallization determine the amount of dislocations in the crystal, influencing charge carrier lifetimes as well as the strength of the wafers. Controlling and understanding the crystallization process is very essential for improving the electrical and mechanical properties of wafers and cells[18]. Predicting the behaviour of an industrial-scale melt crystal growth process requires a faithful depiction of furnace-scale heat transfer along with a detailed accounting for heat transfer, melt convection, and solid-liquid interface motion. Here, the problem is mainly focused on the utilization of finite element technique to understand the influence of dimensionless numbers and thermal characteristics on molten silicon during directional solidification process.

Axial (here X) velocity field, the generation of the vortex, convective and diffusive heat fluxes are analysed from the simulation results with respect to various Prandtl numbers. Computed heat and fluid flow characteristics are evaluated at various Prandtl numbers ranging from 0.001 to 2 in the molten Si. Convection instability occurs in the molten silicon above certain Prandtl number which is studied from the simulation results to analyse various fundamental melt flow properties in the chosen model. In this paper, we choose only two Prandtl numbers 0.001 and 2 with constant Raleigh number 1000 and ΔT 10K. According to the melt flow fluctuation, dopant and impurities distribution should affect the crystal homogeneity in the crystal growth process. The thermal fluctuation of flow pattern depends on flow velocity of the melt. The melt convection is dynamically dependent on the crucible diameter, temperature gradient, gas flow range etc. Melt flow instability in the growth process for large diameter Si multi-crystals is usually intense.

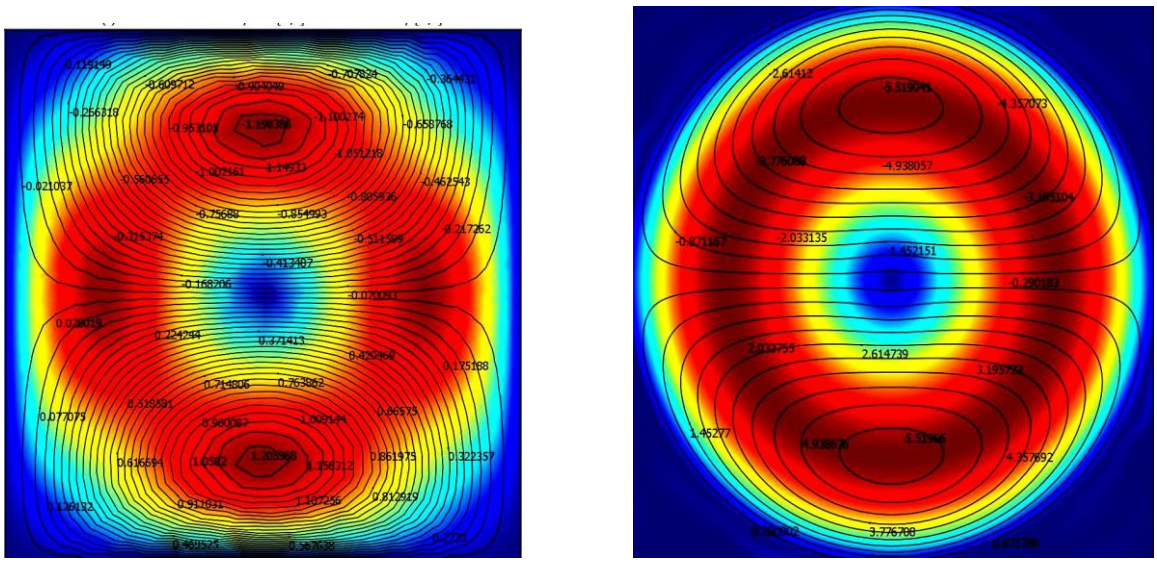

Fig. 2. Axial(X) velocities in the melt when the flow is only driven by buoyancy forces at various Prandtl numbers (a) Pr = 0.001and (b) Pr = 2

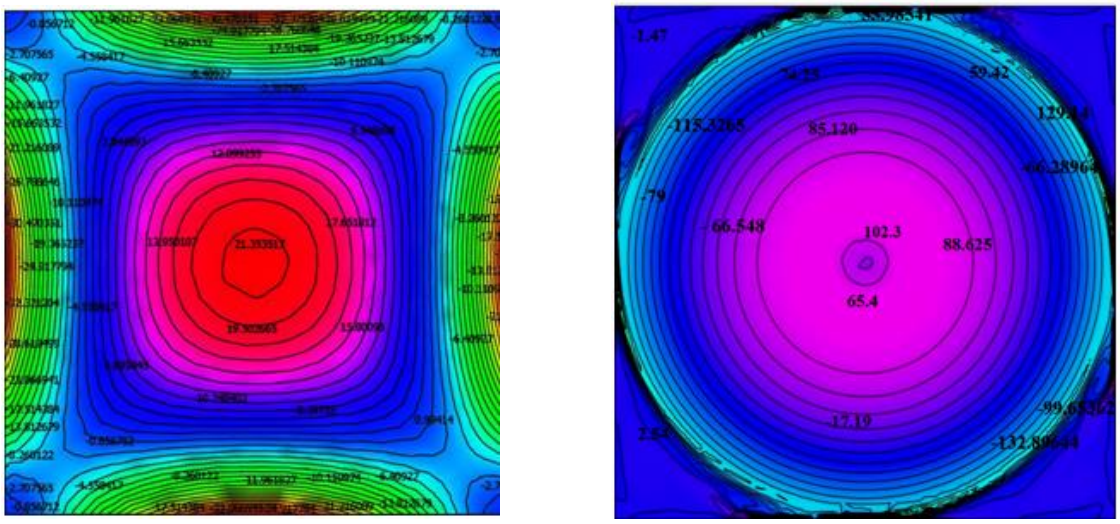

Fig. 3. Vorticity in the melt when the flow is only driven by buoyancy forces at various Prandtl numbers (a) Pr =0.001 and (b) Pr = 2

To analyze the melt flow instability in molten silicon, simulations of thermal studies were performed for an industrial DS-Si crystal growth process, in which the diameter of the grown crystal is as large as 500 mm. The unstable melt flow is simulated using the finite element method. Fig. 2 shows the fluctuation fields of velocity contour with temperature spectrum of silicon melt. It is seen that the instabilities of the melt flow in centre regime and the melt flow at the melt surface are intensive. The melt flow X - axial velocities at the central melt regime with a maximal magnitude compared with periphery regime of melt of an order of approximately 0.02 m/s to 5m/s are obtained for Prandtl number 0.001 and 2 respectively. The velocity field spectrum demonstrated the perturbation of velocity field of melt whose values are decreasing from centre to periphery region (near wall). If velocity values decrease then the boundary layer thickness may increase. This boundary layer thickness directly affected the impurity distribution in melt and quality of the grown crystal. From the calculated velocity values, velocity fields are not much varied at low Prandtl number (Pr= 0.001). At the high Prandtl number(Pr=2) velocity fluctuation occurred heavily, it varied from 5.506 m/s at central regime to 0.023 m/s at periphery regime. So we need to optimize the flow velocity of molten silicon at minimal Prandtl number when we are growing bulk crystal. For that optimal Prandtl number, we may set the temperature gradient and such other physical parameters for growing good quality mc-silicon crystals.

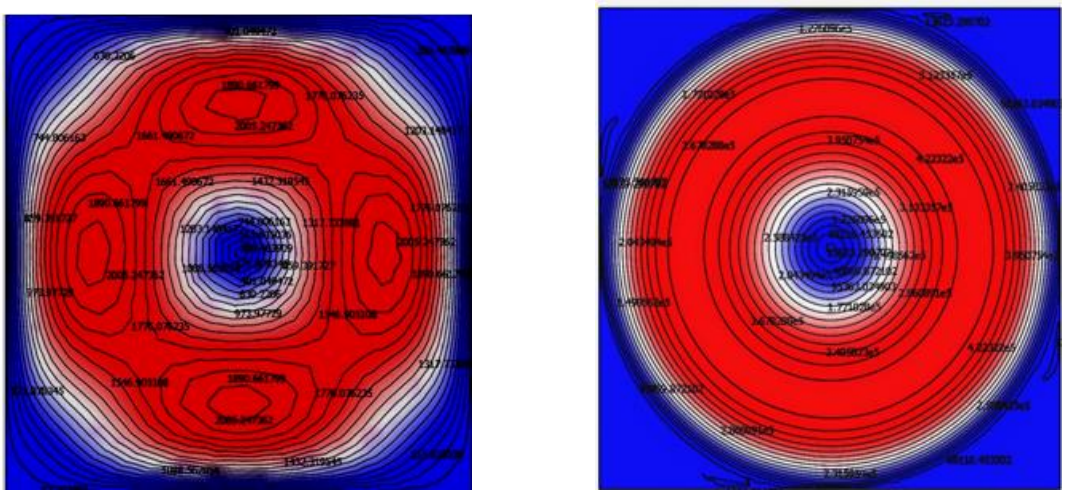

Fig. 4. Convective heat flux in the melt when the flow is only driven by buoyancy forces at various Prandtl numbers (a) Pr =0.001 and (b) Pr = 2

The vorticity is another main complex physical phenomenon in directional solidification of Si growth process. The vorticity generation is to create melt instability in molten silicon. Vorticity is related with the linear integrals of the velocity. It could be controlled by setting suitable temperature gradients with dimensionless numbers. It may also control by applying external force such as rotating crucible or subjecting the magnetic field

on the melt flow. In this paper specifically, we examine minimizing the vorticity in the melt region which is an effort directed at decreasing large-scale turbulent motions inside the melt and to indirectly minimize oscillations in the melt-crystal interface during crystallization. Thus, we have considered to optimize the minimum vorticity in melt regime through control of Prandtl numbers. Fig. 3 displays the vorticity which is very low at Prandtl number 0.001 compared with Prandtl number 2 in the molten silicon. In our case, it is obtained from simulation results of melt flow contour. The positive value of vorticity corresponds to the melt surface moving upwards and the negative value of vorticity corresponds to the melt surface moving downwards. The clockwise flow of the upper layer melt also indicates that thermal buoyancy force plays less important role in determining the melt flow near the melt free surface. Fig 3 (a) shows the vortices which are clearly seen in simulation results at Prandtl number 0.001. The main vortex is generated in central melt regime and small secondary vortices are formed near the walls of melt system. In this case, we have understood, the upward and downward flow direction (through positive and negative vorticity values), separated buoyancy(under) and surface tension(upper) effect in molten silicon. However these vortices are controlled using adjustable physical parameters in directional solidification. At the same time for the Prandtl number 2, Fig.3(b) shows that without secondary vortex the main vortex is strongly generated at the whole system. The upward and downward fluid motion is mixed in most of the region. The vortex values reached more than 100(1/s). This type of vortices leads to the growth of bad quality crystals. So we need to control the vortex using possible way in the molten silicon during solidification process.

Heat flux (convective, conductive and diffusive heat fluxes) is one of the important mechanisms in transport phenomena of crystal growth process. It is defined as the rate of heat energy transfer in melt through a given area, per unit area. A typical heat flux contour structure, obtained for two different Prandtl numbers of 0.001, and 2 are shown in Fig 4 and 5. The convective heat flux plays an important role in the control of dopant, and impurity distribution of grown crystals. The main concern is the occurrence of convective instability in the melt and can be understood by temperature fluctuations in the melt flow. It causes crystallographic defects during melt growth. This factor has a significant impact on the strength of the convection and consequently the shape of the melt - crystal interface in the DS crystal growth. The convective heat fluxes are simulated for two Prandtl numbers of the molten silicon. The convective heat flux values are randomly increased when the Prandtl number is increased from 0.001 to 2. Fig.4. (a) & (b) show that convective heat flux values are randomly increasing from approximately 2000 to 200000 with respect to Prandtl numbers Pr=0.001and 2 on the molten silicon. It influences on the melt crystal interface and distribution of impurity concentration, defects in crystal growth. When convective heat flux is randomly increased then it may influence the flow to become turbulent. From our simulations, the convective heat flux is increased highly at Prandtl number 2 compared with Prandtl number 0.001. So we have determined the effect of flow regime due to convective heat flux. If we control convective heat flux then melt-crystal interface geometry can be optimized during solidification process.

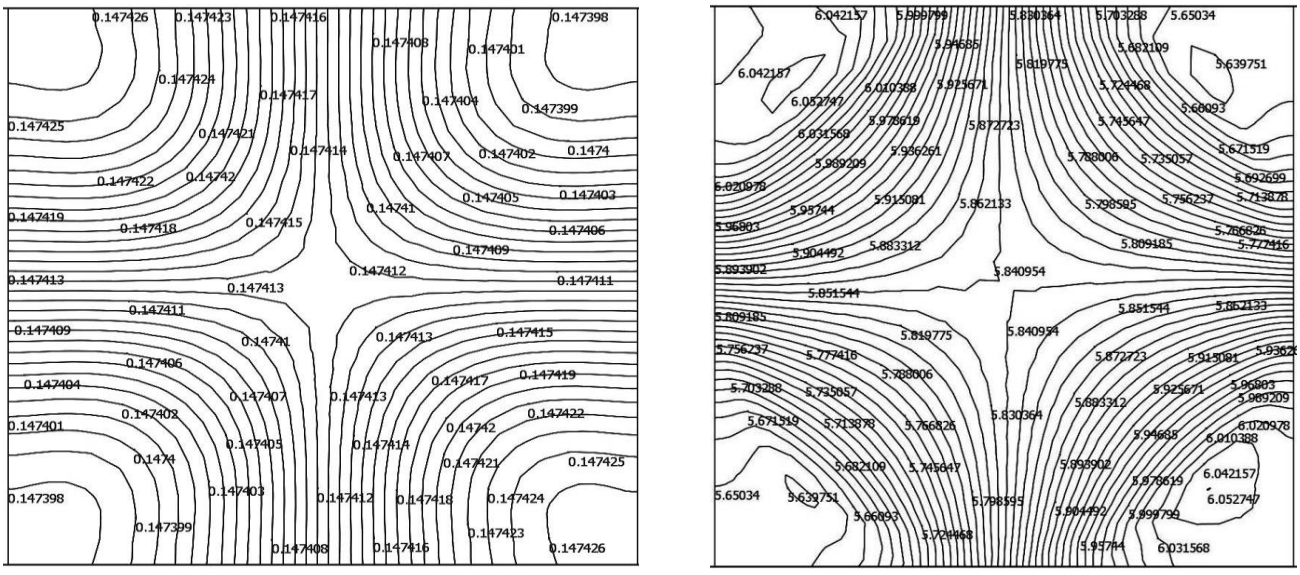

Fig. 5 Diffusive heat fluxes are calculated in molten silicon at two Prandtl numbers (a) Pr =0.001 and (b) Pr = 2.

Fig. 5 shows the diffusive heat flux contour of molten Si. The diffusive heat flux is influenced in the central region of melt except the corner of melt regime. It indicates that the fluid flow pattern is also affected by the diffusive heat flux from bottom to top due to buoyancy force. But the calculated values are not much increased compared with the convective heat flux. So the convective heat flux only highly dominated and influenced the molten

flow fluctuations during bulk multi-crystalline silicon growth process. The diffusive heat flux may not influence the flow structure according to the variation of Prandtl number. This diffusion heat flux may affect the impurity and dopant distributions in grown crystals. According to the crystal growth process impurities and dopants distributions are the main factors. So we should not avoid diffusive heat flux analysis. From the above simulation results, we may conclude that the lower heat flux controls the melt instabilities. In practical consequence, the thermodynamic factors like velocity field, heat flux, vorticity, surface tension and temperature gradient affect the crystal quality.

6. Conclusions

Numerical model of heat transfer on a molten silicon was performed using finite element method in directional solidification of silicon growth process for solar cell applications. The influence of Prandtl numbers on the heat transfer characteristics were investigated. The dimensionless numbers such as Prandtl number, Rayleigh number can significantly influence the thermal and velocity fields in the solidification process. If the Prandtl number is large(Pr>1), the large melt fluctuations occur in molten silicon and these thermal fluctuations will lead to great thermal stress and high dislocation in growing crystals. Axial velocity field, generation of vorticities, convective and diffusive heat fluxes were investigated for various Prandtl numbers. For the constant temperature gradient, the perturbation velocity field in central regime of the melt was reduced from 5.5m/s at Pr = 2 to 1.2 m/s at optimal value of Pr = 0.001. The temperature distribution, the melt convection and the m-c interface shape in a directional solidification process can be controlled and optimized by the study of fluid flow properties based on understanding of dimensionless numbers. Some typical results were presented, showing the importance and effectiveness of computer modelling and numerical simulation in analyzing and improving multi-crystalline Si crystal growth processes.

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