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Numerical modeling of thermo physical properties on molten Si in multi-crystalline silicon growth process

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Numerical investigation of thermal characteristics of Si melt during multi-crystalline Si growth processes like temperature distribution, velocity field, stream line, Peclet numbers and Reynolds numbers were carried out in a 2D and 3D axis symmetric model by the finite-element technique. One of the greatest technological and scientific challenges of multi-crystalline silicon growth is to achieve homogeneity of the material properties in the grown crystal. It is affected by fluctuations of the growth rate of the crystal. One of the possible causes for fluctuations of the growth rate is the non-stationary convection in the melt. The aim here is to investigate the complex heat transport phenomenon existing in the Si melt during the DS growth process with three different temperature gradients. Being a non-linear process, the dimensionless numbers indicated above play vital role in parametric analysis. The simulation results in the present study indicated the existence of the flow regimes and their dependency on the temperature gradient under study. The obtained results are in reasonable agreement with the predictions of the theoretical approach. The studied dimensionless numbers will be beneficial in understanding the flow regimes during the DS process of Si.

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Keywords: Heat transfer; Fluid flow; Silicon; Thermodynamics; Simulation

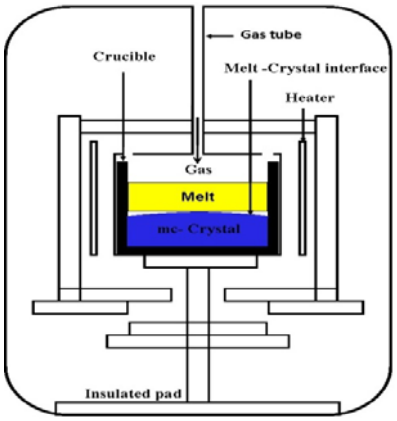
1. Introduction

The unsustainable nature of fossil fuels as an energy source, from the point of view of future availability and environmental impact, has spurred an interest in diversification of energy sources, with particular interest in renewable energy. The photovoltaic (PV) 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[1]. The application of multi-crystalline silicon crystals in Solar cells enormously increased during the recent decades. A large portion of solar cells is presently made from multi-crystalline silicon using directional solidification(DS) method. One of the greatest technological and scientific challenges of multi-crystalline silicon growth is to achieve homogeneity of the material properties in the grown crystal.

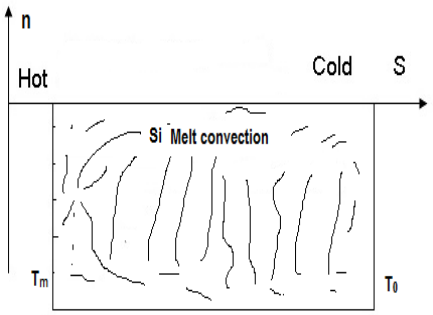
The quality of the multi-crystalline silicon mainly depends on the interface and transport phenomenon occurring in the melt during the growth process. One of the possible causes for fluctuations of the growth rate is the non-stationary convection in the melt, generated by the thermal and kinematic boundary conditions. The physical process in solidification of liquid is of great importance in natural and industry world[2]. It can be described by mathematical model considering the transport of mass, momentum and energy. Theoretical analyses and numerical simulations with these models have been used in various crystal growth techniques to understand the physical process [3,4]. The quality of grown crystal by directional solidification is largely effected by the melt flow convection, which directly affects both the yield of good silicon from an ingot and the solar cell performance. The physics governing the growth of mc-silicon in the directional solidification system involves complex non-linear transport phenomena of heat and mass transfer processes. An understanding of the macro scale forces which drive convection during directional solidification is crucial to control the growth of uniform crystals [5,6].

Reports are available on influence of some dimensionless numbers for some crystal growth methods. There are no reports, using finite element numerical technique, on the influence of Peclet number and Reynolds number on thermal energy transfer in silicon melt during directional solidification process. Peclet and Reynolds numbers have significant role on melt convection due to thermal fluctuations during crystal growth process. In this paper, thermal characterstics are obtained by interpreting the dimensionless numbers based on fluid mechanics and heat transfer. The study was motivated by the intention to answer the question of "how small temperature difference also can create convection in melt flow patterns". We have studied for the first time the influence of Peclet numbers using various temperature differences for silicon melt during directional solidification. Low Peclet number ($Pe<1$) and low Reynolds number are desired for solidification process because melt instabilities occur in high Peclet and Reynolds number[18]. So we have conducted the limited range of parametric analysis. These parameters are also used to study the boundary layer and thermal layer characteristics of the flow. The aim of the study is to carry out the heat transfer characteristics of molten silicon during multi-crystalline silicon growth process.

In this work, the melt convection due to the temperature difference of melt is discussed in the time - independent model with the Newtonian incompressible Navier-Stokes equation in the Boussinesq approximation and the convection-diffusion. The computations were performed in a 2D and 3D axis symmetric model by the finite-element numerical technique. Thermal characteristics like temperature distribution, velocity field, stream line, Peclet numbers and Reynolds numbers in Si melt are performed for directional solidification crystal growth processes using numerical technique. The melt flow properties and heat characteristics are simulated and analyzed for more than 40 temperature differences of silicon melt for directional solidification system. Here thermo physical parameters were given only for three temperature differences.



(a)



(b)

Fig. .1 Schematic diagram of the DS furnace: (a) conventional furnace and (b) Si melt system between hot and cold temperature region.

The simulation model is defined as follows: Fig.1 (a) shows the DS furnace for growing multi-crystalline silicon. A DS crystallization apparatus consists typically of a rectangular–shaped silica crucible in furnace which has Si melt with crystal seed. A model is assumed as axis symmetric for small Si melt system with two temperature regions. Fig.1(b) shows in vector form that the melt flow is driven from hot region to cold region. This model is simulated with 2D and 3D stationary behavior on the Si melt system. Temperature is denoted on hot region as T_m and on cold region as T_0. Known thermo-physical properties of silicon are given in the table 1.

Table 1: thermo - physical properties 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)
dγ/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_m 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	0.04K, 0.4K,4K

2. Mathematical modeling:

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. For liquids, such as molten phase in melt growth, flows are described by conservation equations written for momentum (Navier-Stokes equation) and continuity of an incompressible, Newtonian fluid with the application of the Boussinesq approximation [7]-[13].

Navier-Stokes equation

$$-\nabla.\eta[\nabla\vec{u}+(\nabla\vec{u})^T]+\rho\vec{u}.\nabla\vec{u}=-\nabla P+F \qquad (1)$$

Continuity equation

$$\nabla.\vec{u}=0 \qquad (2)$$

Navier- Stokes equation with Boussinesq approximation

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

The first term gives the rate of momentum gain by viscous transfer, the second by convection, and the third by pressure forces, where the variable of properties η, ρ, u, p, g, β,T, are dynamic viscosity , density, velocity vector, pressure, acceleration due to gravity, thermal expansion and reference temperature respectively. The F is considered only as Boussinesq approximation, it is a source term representing external forces per unit volume (N/m^3).

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 [8]. The variables of parameters k, T, $C_{p,}$ are thermal conductivity, temperature and heat capacity respectively.

$$\text{Peclet number } Pe = \beta h / D \tag{5}$$

$$\text{Reynolds number } \mathrm{Re} = \vec{u}.L / \nu \tag{6}$$

Here, □ is Kinematic viscosity. L is characteristic element length. On the melt free surface, we consider slip/2D and axial symmetry condition [13],

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

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

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 [10].

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

Here the sign of $d\gamma/dT$ 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 DS system are given in Table 1.

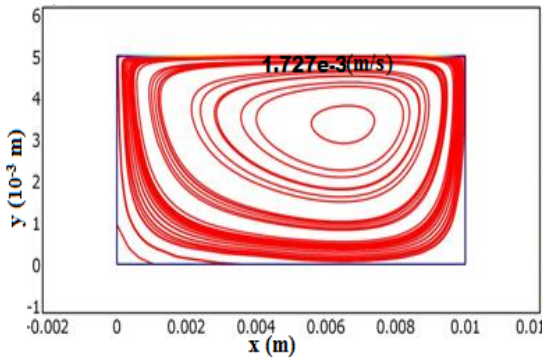
3. Numerical method

The governing equations and the boundary conditions for the fluid flow and temperature fields in the Si melt system 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 [14, 15]. The computations are made using the 2D/3D axis-symmetry hypothesis for inner cell silicon melt of rectangular crucible. According to the considered geometry 123256 triangular elements are partitioned as sub domains. The time-independent Newtonian incompressible Navier-Stokes model for fluid flow, heat and mass transfer, along with weak form of the boundary is solved using the finite-element technique.

4. Simulation results and discussion

Numerical simulation of Si melt flow properties such as stream line flow, temperature distribution, Peclet number and Reynolds numbers were performed for three temperature differences (0.04K, 0.4K and 4K). Growth of multi-crystalline silicon crystals is a complex process involving heat and mass transfer phenomena. The related temperature and concentration fields have a dominant effect on the quality of crystals. Therefore, appropriate control of fluid dynamics and good understanding of the phenomena are very important for the growth of high - quality ingots for solar cell applications. Predicting the behavior 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 such as Peclet number and Reynolds number on the Newtonian flow regimes in melt domain of DS process.

The studied temperature difference $\Delta\mathbf{T}$ is considered along the x direction of the melt domain only. Also, the governing equations for the melt region are described by coupled Navier-Stokes and energy equations. The Boussinesq approximation is used to account for buoyancy force in momentum equation via temperature dependence of density in gravity term. This Boussinesq approximation could be applicable in Navier-Stokes equation in any one of direction whether x or y direction. The convection of melt flow patterns are dynamically dependent on the temperature field (thermal force). The melt flow fluctuation dictates the dopant and impurities distribution along the growing crystal. The fluctuation of melt flow pattern depends on flow velocity of the melt. The melt flow is everywhere tangent to the streamlines [16]. Streamline patterns indicate flows rotating in a counterclockwise sense which is shown in figure 2. Buoyant forces caused by thermal gradients drive counterclockwise circulating flows.



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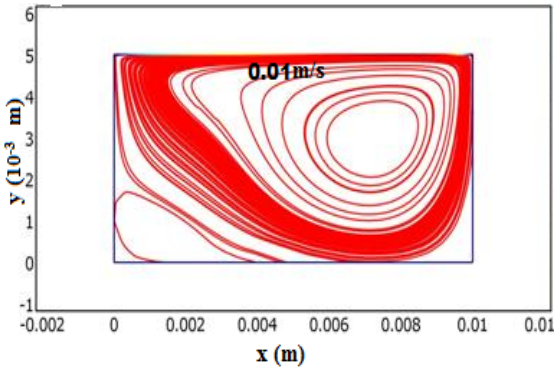

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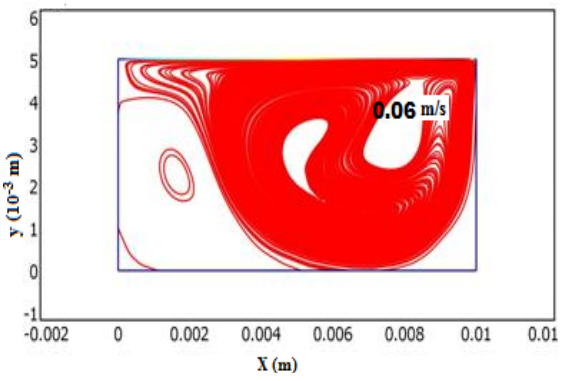
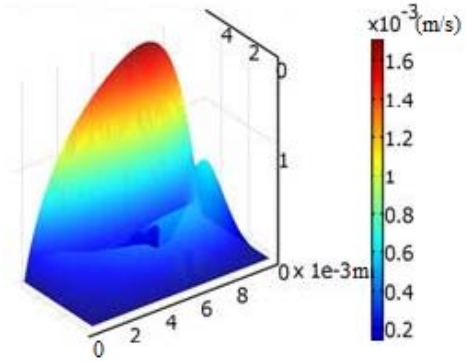


(c)

(a) ΔT= 0.04K (b) ΔT= 0.4K (c) ΔT =4K.

Fig. 2. Stream line flow with velocity(m/s) in 2D for three temperature differences.



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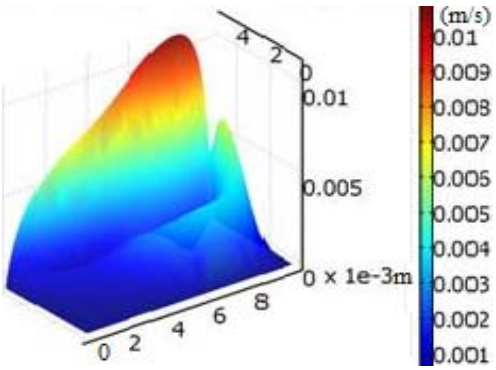

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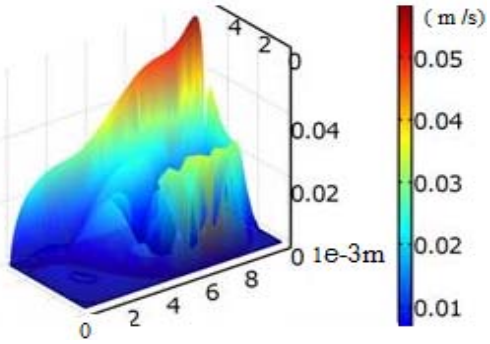
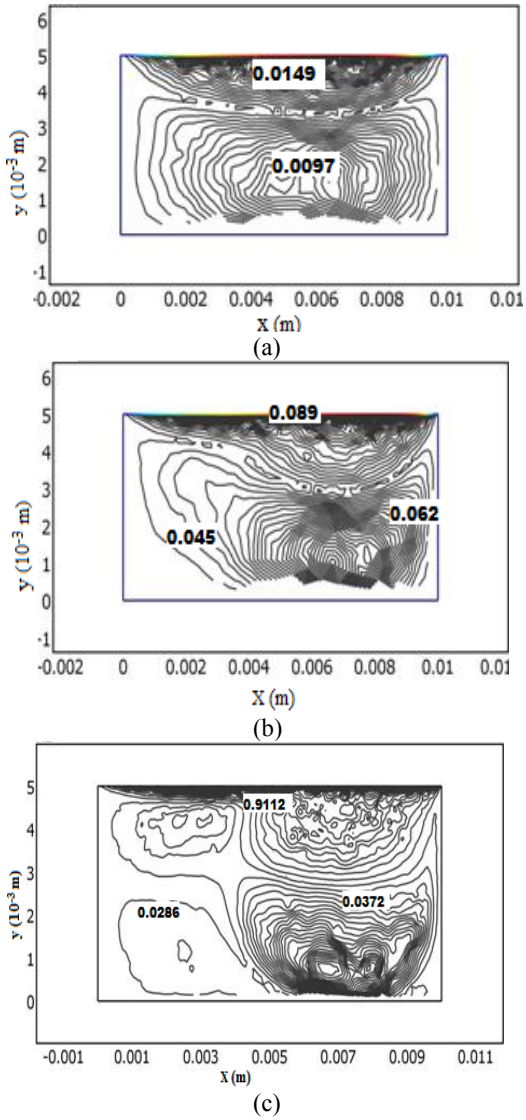


(c)

(a) Δ**T**= 0.04K (b) Δ**T**= 0.4K (c) Δ**T** =4K.

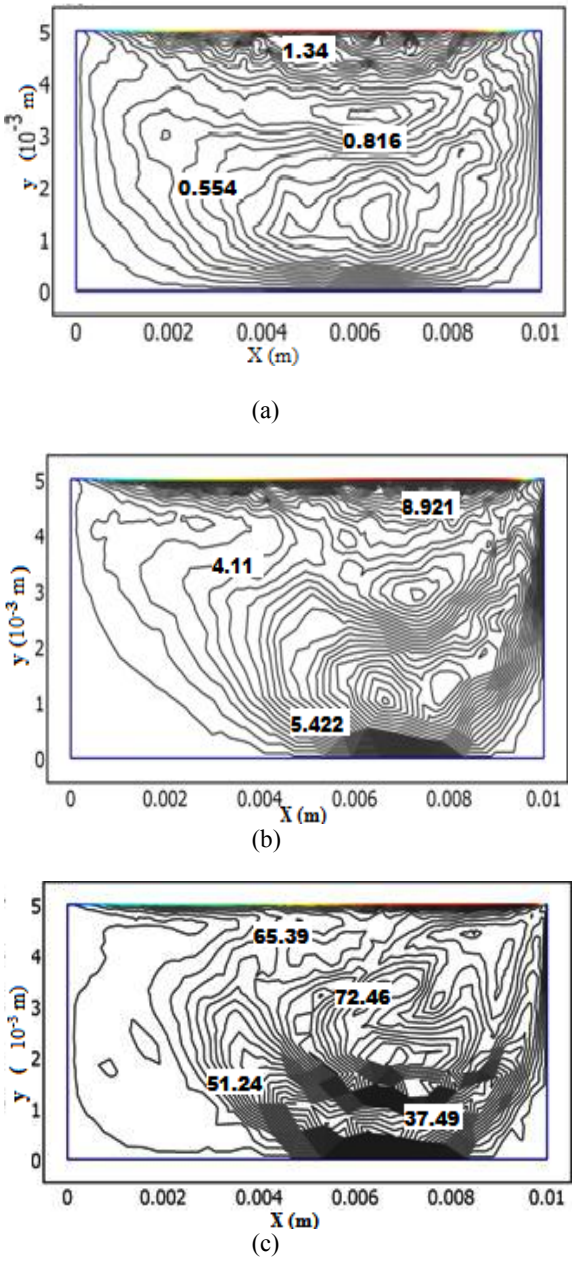
Fig. 3. Temperature distribution(K) with velocity field(m/s) in 3D for three temperature differences.

The variation of flow velocity and flow pattern with studied temperature difference are depicted in Fig. 2 in 2D model. The simulation results in velocity fields of 0.00173m/s, 0.01m/s and 0.06m/s for ΔT = 0.04K , 0.4K and 4K respectively. Fig. 3 shows the temperature distribution along with melt flow velocity field in 3D numerical technique. Fig. 2 and Fig. 3 clearly indicate the role of temperature difference in influencing the flow pattern, velocity and temperature distribution. As per the literature, if the velocity field is controlled below 0.02m/s it is better for controlling melt convection in silicon growth process. The boundary layer thickness may also be calculated using this velocity field in order to understand the magnitude of impurities dissolving inside the wall during the growth process. The present simulation results in higher melt velocity field when the temperature difference ΔT is more than 0.4K.



(a) ΔT= 0.04K (b) ΔT= 0.4K (c) ΔT =4K

Fig.4 Contour of Peclet number(no unit) for three temperature differences.



(a) ΔT= 0.04K (b) ΔT= 0.4K (c) ΔT =4K

Fig. 5. Contour of Reynolds number (no unit) for three temperature differences.

The temperature distribution is mainly involved to grow high quality crystals. The temperature distribution along the growing crystal is a fundamental parameter which characterizes temperature field and influences the morphological instability and defects formation. The mechanical and thermal stress are maximum avoided in the grown crystals when we control the temperature distribution as homogeneous in the whole melt of the growth system[17]. The obtained results suggested that the temperature distribution at the bottom of the crucible is more uniform when the temperature difference is 0.04K

and instability occurs when ΔT =4K. The instability of the flow can be understood from the presence of multiple flow regimes at the bottom. Also, the flow velocity is much low at the bottom when compared to the top region of the melt. The Peclet number plays a vital role to study heat transfer in liquid that is similar to the Reynolds number in fluid mechanics. The Peclet number explained the relative importance of convective transport of thermal energy when compared with molecular transport of thermal energy (conduction)[18]. Figure. 4 shows the simulated results on Peclet number of the silicon melt for three temperature differences. It is found that the Peclet cell numbers are small for silicon melt which lies with theoretical value of semiconductor materials (Pe < 1). If the Peclet number values are above one then the melt has the characteristics of fluctuations with high convection as Peclet number is directly proportional to the rate of energy transport by convection. High Peclet number is not desirable to maintain the melt-crystal interface as plane during solidification process. Further, the increase in temperature difference ended up with breakages and localized regimes having distinct Peclet numbers which embrace the instability in the melt flow.

Fig.5 shows the simulated Reynolds number contours for the Si melt under three ΔTs. Even a small change of temperature differences greatly influences the values of the Reynolds number. If the Reynolds number is increased then the eddy and vortices formation may occur in melt flow. Eddies and vortices create dislocations and inhomogeneity in the impurity distribution in the grown crystal. These are undesirable for growing high quality multi-crystalline silicon for solar cell applications. The simulated contours of Reynolds number for ΔT=0.4K and 4K clearly evidences the formation of vortices in the flow regime and the increased values of Reynolds number for ΔT=4K supports the fact of increased convection in the melt. Under these circumstances, the simulated results may more helpful in optimizing the experimental condition to modify or control the flow in the melt during growth be process. Further, the global simulation modeling will be performed in the near future to study of the solidification of the Silicon from melt having optimum heat and mass transfer characteristics and its effect on melt -crystal interface.

5. Conclusion

Numerical investigation on thermo phyical proprties of Si melt in a directional soldification system were carried out in 2D and 3D model using finite element tecnique. A stationary model of the framework has been employed in incompressible Navier-Stokes equation in the Boussinesq approximation and the convection-diffusion equation. The velocity fields were found using 2D and 3D model. The stream line flow velocities simulated in 2D axis symmetry model for various temperature differences were analyzed. And also, the temperature distribution with velocity field was simulated in 3D model for the present system of study. The influence of dimensionless numbers like Peclet number and Reynolds number on the flow characteristics were investigated. The incorporation of the studied dimensionless numbers in understading the flow regimes in Si growth of directional soldificational process will be more beneficial to further improve the quality of the growing crystals.

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