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Numerical Modelling on Stress and Dislocation Generation in Multi-Crystalline Silicon during Directional Solidification for PV Applications

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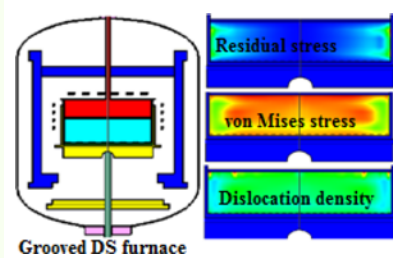
Numerical modelling has emerged as a powerful tool for the development and optimization of directional solidification process for mass production of multi-crystalline silicon. A transient global heat transfer model is performed to investigate the effect of bottom grooved furnace upon the directional solidification (DS) process of multi-crystalline silicon (mc-Si). The temperature distribution, von Mises stress, residual stress and dislocation density rate in multi-crystalline silicon ingots grown by modified directional solidification method have been investigated for five growth stages using finite volume method at the critical Prandtl number, Pr = 0.01. This paper discusses bottom groove furnace instead of seed crystal DS method. It achieves an advanced understanding of the thermal and mechanical behaviour in grown multi-crystalline ingot by bottom grooved directional solidification method. The von Mises stress and dislocation density were reduced while using the bottom grooved furnace. This work was carried out in the different grooves of radius 30 mm, 60 mm and 90 mm of the heat exchanger block of the DS furnace. In this paper, the results are presented for 60 mm radius groove only because it has got better results compared to the other grooves. Also, the computational results of bottom grooved DS method show better performance compared the conventional DS method for stress and dislocation density in grown ingot.

Keywords: bottom grooved directional solidification, simulation, silicon, solar cell, stress, dislocation density



1. INTRODUCTION

In the recent days, many renewble energy technologies are promoted and given much more attention with the diminution of fossil fuels and impairment of the environment throughout the whole global. The PV solar energy is one of the new alternative renewble energy technology in tradational energy sourses.[1] The silicon solar cell industries have been quickly developed in last two decades. The electricity generation from solar cells is deemed to be one of the central technologies of this century. Either mono or multi- crystalline silicon material is extensively involved in solar world. Czochralski or Floating zone crystal growth methods are mostly used to grow mono-crystalline silicon which is mainly used in electronic devises as well as solar cell applications. Multi-crystalline silicon (mc-Si) is grown by directional solidification(DS) method to provide large scale multi-crystalline silicon.[2] Now-a-days, the multicrystalline silicon (mc-Si) is the prevalent material for PV solar cells and its performance improvement is required in order to enhance the conversion efficiency. Silicon material is applied in 90% of commercial solar cell modules. Especially, a market share of multi-crystalline silicon has more than 60% in the photovoltaic materials. It is a crucial material with advantages of high conversion efficiency and low production cost compared with other solar materials.[3,4]

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Nakajima *et al*.[5] have controlled the dendrite arrangement in the dendrite casting method by the cooling pad with different thermal conductivity. Also they have reported the enhanced cooling conditions.[6] Li *et al*. controlled the grains to get a high quality mc-Si by introducing the notched – crucible.[7] Zhou *et al*. have shown that the final dislocation density can be reduced by the modified cooling process in mc-Si ingot production.[8] Yang *et al*.[9] have investigated the effect of temperature distribution, melt convection, c-m interface and thermal stress of mc-Si growth process in the DS process. The thermal distribution, c-m interface shape and energy consumption and heat loss from different portions of the system has been analyzed.[10] Wang *et al*. have reported the cooling spot furnace for higher cooling rate at the center portion of the crucible than the other parts of the charge.[11] In the present work, we have introduced a groove in the bottom of the heat exchanger block in the DS furnace to achieve spot cooling. The c-m interface, von Mises stress and dislocation density were analyzed by using conventional and modified furnace. The numerical simulations were carried out to demonstrate the performance of bottom grooved heat exchanger block in the DS system.

The yield of usable mc-silicon wafers as well as the efficiency of the solar cells are harmfully affected by inherent impurities such as SiC, carbon, nitrogen, oxygen, etc, and generation of stress and dislocation density. Many researchers have discussed the effects of melt flow fluctuation by thermal force, crystallization front shape, melt-crystal interface, distribution of some of non - metallic impurities in grown ingot at the time of directional solidification process. Dislocation density is an important physical parameter to study the crystal quality and its response on the efficiency of solar cells. The thermal stress is the main cause to generate the dislocation in grown multi-crystalline silicon either by conventional or modified DS method. During directional solidification of molten silicon, thermal stress such as von Mises stress, residual stress are the main type of stress in the grown mc-Si crystal. Generation of thermal stress depends on the variation of cooling rate, induces the thermal expansion at different degrees in the ingot and temperature differences in grown ingot. Now-a-days, computer aided design packages based on spectral element, finite difference, finite volume and finite element method have attained great importance in many industries. These software tools are widely used in furnace designing and manufacturing, final testing of mechanical and thermal systems, especially in PV industries.[12]

In this paper, numerical investigation was made based on finite volume method for a large-scale industrial furnace with side and top multi-heater, bottom grooved directional solidification furnace which can produce 400 kg mc-Silicon ingots. Time-dependent 2D simulations were performed for the temperature distribution, stress generation and dislocation density rate during solidification process at the various stages. Quantitative analyses of temperature distributions, von Mises stress, residual stress and dislocation density rate are presented in crystal grown with 60 mm radius of bottom grooved DS furnace. The results may be helpful in understanding the grown crystal characteristics and further optimization of furnace design and growth parameters.

2. DS MODEL DESCRIPTION

The configuration of the conventional and modified DS furnace has been shown in Fig. 1(a) and (b) respectively. 60 mm radius groove has been made in the heat exchanger block in the modified DS furnace. The modified blocks are used for controlling the temperature gradient at the bottom of the crucible. This DS system mainly consisted of chamber wall, silicon nitride (Si_3N_4) coated silica crucible, thermo couples, graphite resistance heater, Argon gas tube, graphite susceptor, bottom heat exchanger block, insulations. The

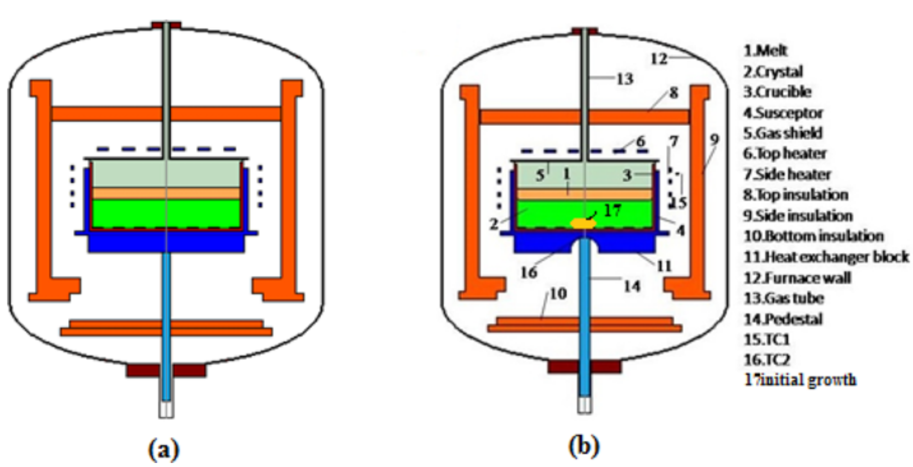

Fig. 1. Schematic diagram: (a) conventional furnace (b) bottom grooved DS furnace.

solar grade silicon feed material is loaded into a rectangular silica crucible. The graphite susceptors are used to support the crucible walls for avoiding deformation at the higher temperature during the growth process. For the purifying growth environment, the argon gas is used with the optimizing flow rate in DS system. A desirable temperature gradient is held by adjusting the heating power and slowly moving the side insulation upwards during the bulk crystal growth phase.[13] This DS system is capable of producing an mc-silicon ingot with the size of 840 mm diameter and 250 mm height. The considered system may be built either with a 2D axi-symmetric geometry to produce a cylindrical ingot or symmetrical geometry to produce rectangular ingot.

Table 1. Physical properties of DS system.

Material	Variable	Value
Silicon (crystal)	Heat conductivity, k (W/m·K)	110.612 - 0.1507T + 0.000109T2 - 4.0094E - 008T3 + 5.668E − 012T^3
	Emissivity	0.9016-.0026208T
	Density, ρ (kg/m^3)	2339.5-0.03267
	Latent heat, ΔH (J/kg)	1800000
	Heat capacity, C_p (J/kg·s)	1000
	Poisson's ratio	0.217
	Young's modulus, E (Pa)	1.653E+11
Silicon (melt)	Heat conductivity, k (W/m.K)	66.5
	Emissivity	0.3
	Density, ρ (kg/m^3)	3194-0.3701T
	Melting point, T_m (K)	1685
	Heat capacity, C_p (J/kg·s)	915
	Dynamic viscosity, μ (Pa·s)	0.008
	Latent heat, ΔH (J/kg)	1800000
Quarz	Heat conductivity (W/m·K)	4
	Emissivity	0.85
	Density, ρ (kg/m^3)	2650
	Heat capacity (J/kg·s)	1232
Graphite	Heat conductivity, k (W/m·K)	146.8885-0.17687T
	Emissivity	0.8
	Density, ρ (kg/m^3)	1950
	Heat capacity, C_p (J/kg·s)	710
Susceptor	Heat conductivity, k (W/m·K)	105
	Emissivity	0.8
	Density, ρ (kg/m^3)	1720
	Heat capacity, C_p (J/kg·s)	1000
Insulation	Heat conductivity, k (W/m·K)	0.5
	Emissivity	0.8
	Density, ρ (kg/m^3)	500
	Heat capacity, C_p (J/kg·s)	100
Argon	Heat conductivity, k (W/m·K)	0.01
	Heat capacity, C_p (J/kg·s)	521
	Dynamic viscosity, μ (Pa·s)	8.466E-6+5.365E-8T-8.682E-12T^2
	Pressure (Pa)	50000
	Molar mass (kg/k·mol)	40

Silicon and other DS elements of thermo-physical properties are given in Table 1.[14]

3. MATHEMATICAL MODEL

The temperature distribution of DS system during the solidification process is calculated based on Fourier's fundamental laws of heat transfer. The governing equation of heat transfer is[15]

$$-\nabla\cdot(\lambda_{ik}\nabla T)+\rho\Delta H\frac{\partial f_s}{\partial t}=\rho C_p\frac{\partial T}{\partial t} \tag{1}$$

where ρ, λ, C_p and H are the density, thermal conductivity, heat capacity under constant pressure and latent heat respectively. Here, f_s is the solid fraction of the mc-silicon during the directional solidification.

Fainberg and Leister have given the axisymmetric displacement-based model for the solidification region, which may be used for solidification of ingot. The governing momentum balance equations in axisymmetric model can be written as[16]

$$\frac{\partial}{r\partial r}(r\sigma_{rr})+\frac{\partial}{\partial z}(\sigma_{rz})-\frac{\sigma_{\varphi\varphi}}{r}=0 \tag{2}$$

$$\frac{\partial}{r\partial r}(r\sigma_{rz})+\frac{\partial}{\partial z}(\sigma_{zz})=0 \tag{3}$$

where, σ_{rz} is the shear stress. σ_{rr}, σ_{zz}, and $\sigma_{\varphi\varphi}$ are the normal stresses in the radial, axial, and azimuthal directions, respectively.

Recently Smirnova *et al*. have discussed the numerical calculation of thermal stress and dislocation in silicon ingot at different cooling processes and also those are verified experimentally. Normally High stress level leads to the inelastic creep deformation. Hence, it is coupled with movement of dislocations. Stress and dislocation distributions are modeled within the Haasen-Alexander-Sumino (HAS) model giving the relationship between plastic deformation and dislocation density.

In this model, the total stain ε_{ij} is assumed to be subdivided by components

$$\varepsilon_{ij}=\varepsilon_{ij}^{e}+\varepsilon_{ij}^{T}+\varepsilon_{ij}^{c} \tag{4}$$

where ε_{ij}^{e}, ε_{ij}^{T} and ε_{ij}^{c} are elastic strain, thermal strain and creep strain respectively.

In HAS model the creep strain rate and the multiplication rate of mobile dislocation density N_m can be expressed as follows:[17,18]

$$\frac{d\varepsilon_{ij}^{C}}{dt}=\frac{1}{2}bN_m\frac{1}{\sqrt{J}}S_{ij}\nu \tag{5}$$

$$\frac{dN_m}{dt} = Kk_0(\tau_{eff})^{p+l}\exp\left(-\frac{Q}{k_BT}\right)N_m - \frac{N_m v}{L} \quad (6)$$

in which,

$$\tau_{eff} = \sqrt{J_2} - D\sqrt{N_m} \quad (7)$$

$$D = R\frac{Eb}{4\pi(1-v^2)} \quad (8)$$

$$J_2 = \frac{1}{2}\sum_{i,j} S_{ij}^2 \quad (9)$$

$$S_{ij} = \sigma_{ij} - \delta_{ij}\frac{1}{3}\sum_k \sigma_{kk} \quad (10)$$

$$v = k_0(\tau_{ef})^p\exp\left(-\frac{Q}{k_BT}\right) \quad (11)$$

In equations (5)-(11), Q is the Peierls potential, k_B is Boltzmann's constant, S_{ij} is the deviatoric stress, b is the value of the Burgers vector, τ_{eff} is the effective stress, J_2 is the second invariant of the deviatoric stress, v is Poisson's ratio, and E is the Young's modulus. D and R are the strain hardening factor and relative strain hardening factor, respectively. k_0, K, p, and l are material constants; the σ_{ij} are the stress tensor components; L is the average grain size; and v is the average velocity of mobile dislocation. The last term in equation 6 is formulated in our approach to account for the fact that grain boundaries are expected to be effective barriers to dislocation glide.[19]

4. NUMERICAL MODEL

Numerical model was performed for the new bottom groove DS system of multi-crystalline silicon ingot using finite volume method. The DS furnace is divided into number of blocks for simulation purpose based on boundaries. Here, new bottom groove DS furnace was introduced. The mesh scheme (Q and triangle grids) is applied as structured, unstructured and both combined for improving the efficiency of computation for DS system. Triangular grids are performed for considered crystal part of DS system. The numerical mesh was generated to be suitable for moving the crystallization front up during unsteady computations of the solidification growth. Unsteady 2D global simulation was carried out based on the thermal phenomena like conduction, convection, radiation, gas flow as well as phase transformation. In this paper, time dependent modelling accounts for conductive and radiative heat transfer in the bottom grooved directional solidification silicon growth system. The Si melt flow was considered as laminar using the Navier-Stokes equations. The melt-crystal interface geometry was analysed at each time step, from starting to end of crystal growth process. The stress and dislocation distributions were simulated using the Alexander-Haasen-Sumino model, which gives the relationship between the plastic deformation and dislocation density in the grown crystal. The time scale of our employed simulation is the same as that of the real experiment. The rectangular crucible is equivalently simplified to a cylindrical shape for the purpose of saving the computational resources. The thermal resistance is kept constant in the whole DS process. This simplification is generally accepted and has been validated with conventional one.[20]

5. RESULTS AND DISCUSSION

Numerical investigation was performed for the thermal field, stress and dislocation multiplication rate of the silicon ingot at different radial grooves (30 mm, 60 mm and 90 mm radius) of heat exchanger block of DS system. For these three different grooves (30 mm, 60 mm and 90 mm radius) of thermo-mechanical values are compared. The maximum values of von Mises stress for various grooves 30 mm, 60 mm and 90 mm are 7.7021×10^6, 6.57×10^6 and 7.201×10^6 Pa respectively. The generation of dislocation density rate of three different grooves 30 mm 60 mm and 90 mm are 21140, 18585 and 267281/s/m^2 respectively. The residual stress of three different grooves 30 mm 60 mm and 90 mm are 7179, 4532 and 6900 Pa, respectively. Generally, stress generation depends on the temperature distribution in grown crystals. Based on above calculation results, the simulation results are presented only for 60 mm groove of heat exchanger block of DS system in this paper. The unsteady numerical modelling was carried out for the bottom grooved heat exchanger DS system of mc-silicon growth process at the critical Prandtl number Pr = 0.01. Figure 1(a), (b) illustrates the furnace elements of conventional DS furnace and modified bottom grooved furnace. The simulation results are presented only for modified bottom grooved DS of mc-Silicon ingot. The DS process mainly depends on thermal field and heater power of DS furnace. Generally heat loss is mainly due to both bottom of the heat exchange block and side insulation of the DS system. In this case, side insulation is moving at the rate of 10 mm/h up to 15 hours, then it becomes stable till the complete crystal growth process. The flow rate of argon gas is 30 L/min and the pressure of DS furnace 0.6 bar. The growth conditions were selected based on literature of conventional DS furnace. The temperature distributions, residual stress and dislocation density rate were simulated during solidification processes at five various growth stages for bottom grooved furnace. The first computation was performed for the time when the system was in the completion of the melting process and initial stage of nucleation formation. Time-dependent 2D modelling of thermal history calculation was made for the 5

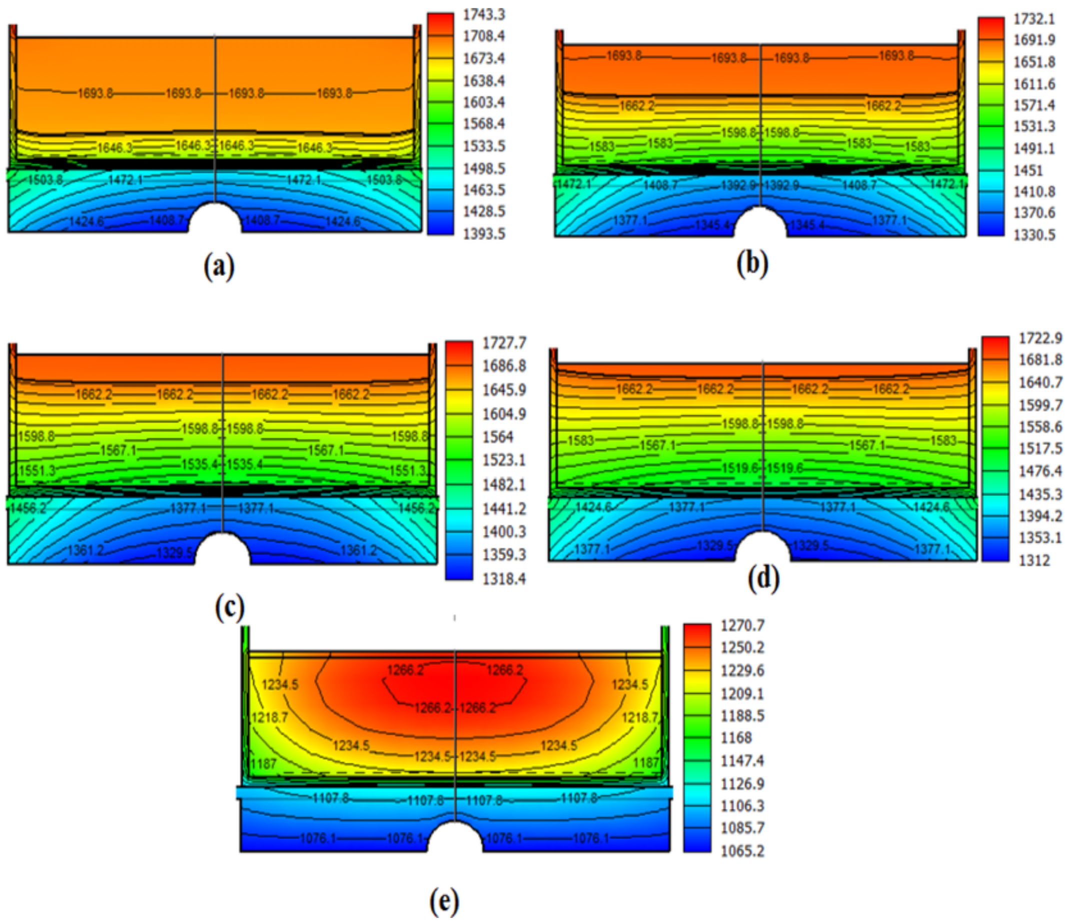

Fig. 2. Temperature distribution of mc-silicon at different growth stages: (a) 5 hr, (b) 15 hr, (c) 25 hr, (d) 35 hr, and (e) 45 hr, Unit: K.

various growth stages from nucleation formation to the end of solidification. This modelling approach confirms realistic conditions for our subsequent simulations. In our considered simulation system, melt, crystal within the crucible and heat exchanger block (bottom groove) were extracted from Fig. 1. Also we have done detailed study of the physical parameters in grown mc-silicon part only.

The temperature distribution of the crystal and melt with crucible and bottom grooved heat exchanger block of DS system at the growth process time of 5 hr, 15 hr, 25 hr, 35 hr and 45 hr is shown in Fig. 2. The simulated isotherm lines in crystal are almost flat at the starting of growth from bottom of the crucible. The isothermal lines shapes are changed from flat to convex at the 15 hr as shown in Fig. 2(b). At the same time, the isotherms near melt - crystal interface are almost flat at most of the region and lightly concave in periphery region in the final stages. These simulation results indicate that there is a large radial temperature gradient in the peripheral region compared with central region. It may cause a large thermal stress in outer region of grown mc-Si crystal, which is confirmed from Fig. 3. Inhomogeneous temperature distribution may couple with locally different expansion/contraction in grown crystal. Theory of thermo-elasticity deals with the stress and thermal field correlation in the grown crystal system. Figure 3 shows the simulated generation of von Mises stress of the grown mc-crystal by bottom grooved DS furnace along the crystal symmetry axis at the five solidification stages. From Fig. 2 and Fig. 3, it is seen that the generation of von Mises stress was small where the melt - crystal interface and isotherm lines are flat. At the same time, the generation of von Mises stresses are high at the outer region of grown crystal whether the melt-crystal geometry and isotherm lines are convex or concave. Already several studies are made on the influence of the melt-crystal interface shape in conventional DS process. In this paper, computations have been made on the thermal and mechanical behaviour of mc-Silicon ingot grown by new bottom grooved modified DS furnace. Especially, we have found that the generation of von Mises stress was suddenly increased at the final stage of solidification process, same as conventional DS process of mc-Si ingot which is shown in Fig. 3. But it is low compared with the conventional grown crystal.

The value of residual stress in the case of bottom grooved

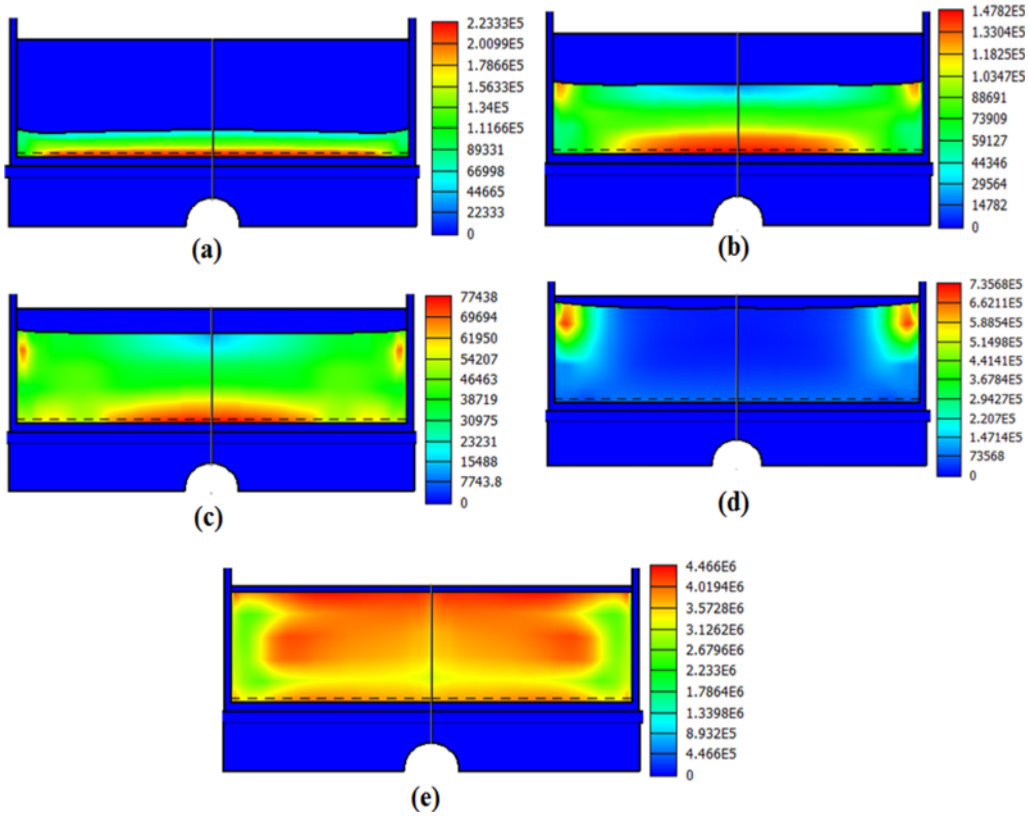

Fig. 3. von Mises stress of mc-silicon at various growth stages (a) 5 hr, (b) 15 hr, (c) 25 hr, (d) 35 hr, and (e) 45 hr, Unit: Pa.

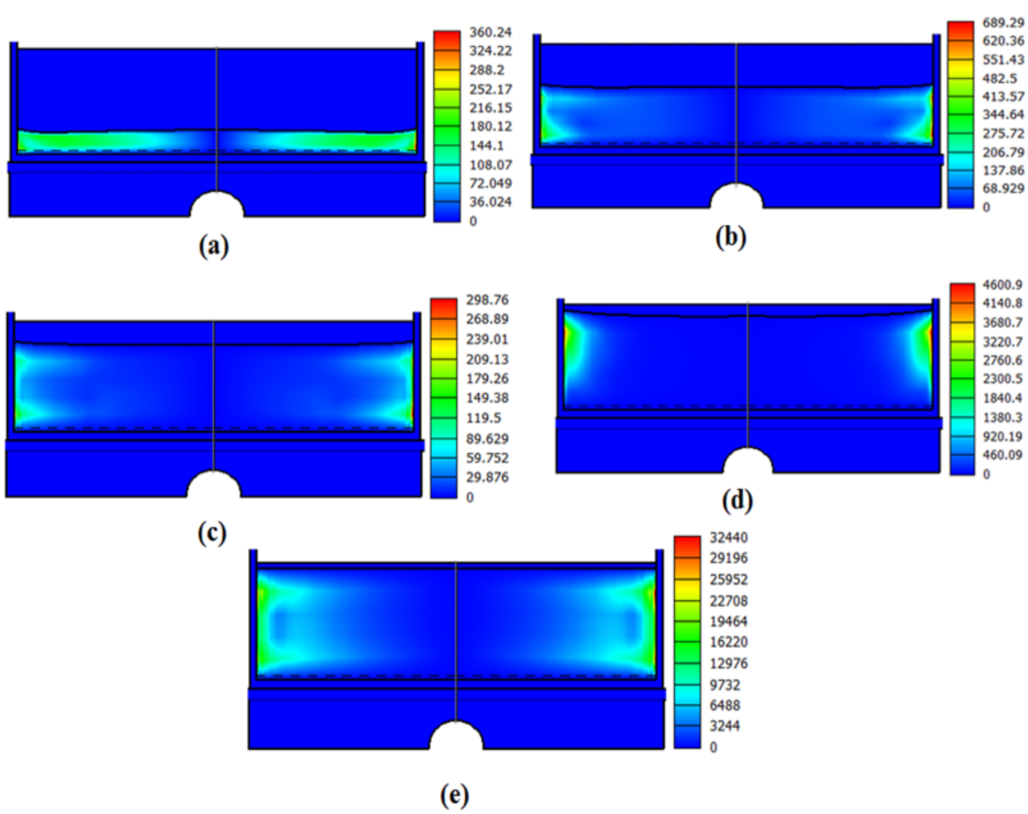

Fig. 4. Residual stress of mc-silicon at various growth stages (a) 5 hr, (b) 15 hr, (c) 25 hr, (d) 35 hr, and (e) 45 hr, Unit: Pa.

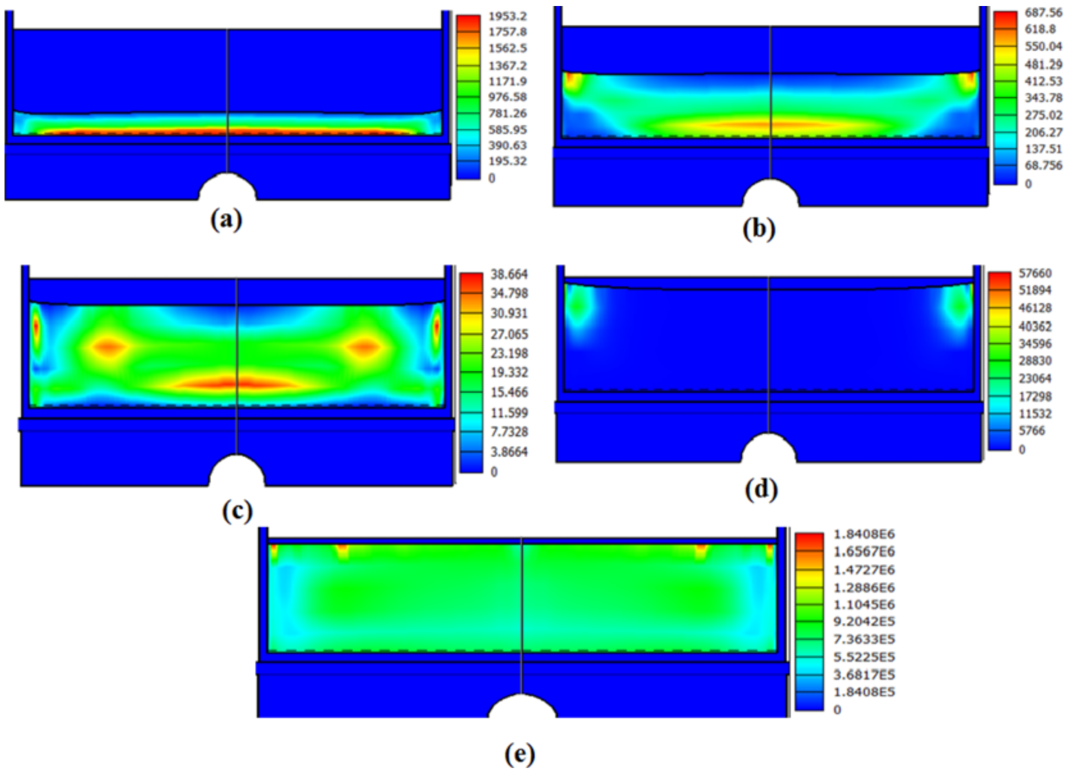

Fig. 5. Dislocation density rate of mc- silicon at various growth stage (a) 5 hr, (b) 15 hr, (c) 25 hr, (d) 35 hr, and (e) 45 hr Unit:(1/s/m).

DS furnace was simulated for the five different stages between every 10 hour of solidification process. The results show that the residual stress of first three stages are below 1000 Pa and it increased to above 30000 Pa at final stage of grown mc-silicon ingot at the time of 45 hr. So we need to concentrate in final stage of growth as same conventional DS growth system. In this new bottom groove model advantage is that the residual stress spread out most of crystal homogeneously which is shown in Fig. 4.

Another important parameter, the dislocation density rate is calculated for bottom grooved DS of mc-silicon ingot at various stages of solidification. The von Mises stress generation may directly influence the multiplication of dislocation density in grown mc-silicon ingot and affect the solar cell efficiency. The relationship between the stress and dislocation density generation in grown silicon ingot is shown in Fig. 3 and Fig. 5. The simulation results show that the dislocation density rate is increased towards the end of grown crystal from 1000 to around 10^6 compared with initial stage. There are many reasons attributed to generation of dislocation density in grown crystal. Stress is main reason to generate the dislocation density. So we have to concentrate in final stage of solidification and optimize the control parameters.

We have concluded from the above simulation results that thermal field von Mises stress creation, residual stress, dislocation density generation are interlinked and directly affect the crystal quality and solar cell efficiency.

6. CONCLUSIONS

A transient global heat transfer model was used to investigate the effect of new bottom grooved heat exchanger block of DS of multi-crystalline silicon (mc-Si). The numerical modeling has been made in 2D axis-symmetry using finite volume method. The temperature distribution, thermal stress and dislocation density rate have been simulated and analysed. The modified heat exchanger block system was used for controlling the temperature gradient at the bottom of the crucible. The obtained results agree with conventional DS of mc-silicon ingot. The von Mises stress and dislocation density of mc-silicon were analyzed while using the bottom grooved heat exchanger block of DS furnace. The work was carried out in the different grooves of radius of the heat exchanger block of the DS furnace. In this paper, the results are presented only for 60 mm radial groove of DS furnace at various growth stages which we found better one compared with other radial grooves of DS furnace by using optimization of simulation results. The obtained results confirm that the crystal quality depends largely on the final stage of solidification process compared with other stages. In future this bottom groove model may lead to much better mc-silicon ingot growth than conventional method.

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REFERENCES

1. L. Sun, L. Liu, Z. Li, and W. Zhao, *Materials for Renewable Energy and Environment,* **1**, 95 (2011).
2. G. Muller and Friedrich, *AIP Conf. Proc.* **1270**, 255 (2008).
3. J. W. Shur, B. K. Kang, S. J. Moonb, W. W. Sob, and D. H. Yoon, *Sol. Energ. Mat. Sol. C.* **95**, 3159 (2011).
4. A. F. B. Braga, S. P. Moreira, P. R. Zamperi, J. M. G. Bacchin, and P. R. Mei, *Sol. Energ. Mat. Sol. C.* **92**, 418 (2008).
5. K. Nakajima, K. Kutsukake, K. Fujiwara, K. Morishita, and S. Ono, *J. Cryst. Growth* **319** 13 (2011).
6. T. F. Li, H. C. Huang, H. W. Tsai, A. Lan, C. Chuck, and C. W. Lan, *J. Cryst. Growth* **340**, 202 (2012).
7. T. F. Li, K. M. Yeh, W. C. Hsu, and C. W. Lan, *J. Cryst. Growth* **318**, 219 (2011).
8. N. G. Zhou, M. H. Lin, L. Zhou, Q. F. Hu, H. S. Fang, and S. Wang, *J. Cryst. Growth* **381**, 22 (2013).
9. X. Yang, W. Ma, G. Lv, K. Wei, T. Luo, and D. Chen, *J. Cryst. Growth* **400**, 7 (2014).
10. J. Wei, H. Zhang, L. Zheng, C. Wang, and B. Zhao, *Sol. Energ. Mat. Sol. C.* **93**, 1531 (2009).
11. T. Y. Wang, S. L. Hsu, C. C. Fei, K. M. Yei, W. C. Hsu, and C. W. Lan, *J. Cryst. Growth* **311**, 263 (2009).
12. Q. Chen, Y. Jiang, J. Yan, and M. Qin, *Prog. Nat. Sci.* **18**, 1465 (2008).
13. W. Ma, G. Zhong, L. Sun, Q. Yu, X. Huang, L. Liu, and Chen, *Sol. Energ. Mat. Sol. C.* **100**, 231 (2012).
14. L. Braescu and T. Duffar, *J. Cryst. Growth* **310**, 484 (2008).
15. J. W. Huang, W. S. Hwang, C. H. Hwang, and Y. W. Chang, *IOP Conf Series: Materials Science and Engineering* **33**, 012059 (2012).
16. J. Fainberg and H. J. Leister, *Comput. Method. Appl. M.* **137**, 167 (1996).
17. H. Alexander and P. Haasen, *Solid State Phys.* **22**, 27 (1968).
18. S. Nakano, X. J. Chen, B. Gao, and K. Kakimoto, *J. Cryst. Growth* **318**, 280 (2011).
19. J. P. Hirth and J. Lothe, *Theory of Dislocations*, Krieger Publishing Company, Malabar, USA (1983).
20. W. Zhao, L. Liu, L. Sun, and A. Geng, *J. Cryst. Growth* **401**, 296 (2014).