

Solidification Analysis on Fabrication of Poly-Si Thin Films by Using Laser Crystallized Technology

雷射結晶技術製造多晶矽薄膜之凝固分析

Chang Chien-Hung^{1,2}, Chao Long-Sun²

張建宏^{1,2} 趙隆山²

¹Department of Mechanical Engineering Air Force Institute of Technology

^{1, 2}Department of Engineering Science, National Cheng Kung University

¹ 空軍航空技術學院機械工程科

² 國立成功大學工程科學系

Abstract

A model has been developed for the solidification of thin Si films induced by excimer-laser annealing. In this paper, a mathematical model was built to analyze the temperature fields of the working pieces under different control parameters, such as laser energy intensity, substrate temperature etc. The numerical scheme is the finite difference method and the specific heat/enthalpy method was used to handle the release of latent heat. The pulse energy of laser was treated as the source term in the energy equation.

From the computing results, it can be found that the SiO₂ layer could effectively decrease heat transferred to the glass substrate and keep most heat from laser in the Si layer. Increasing the laser energy intensity and the temperature of glass substrate could increase the melting depth of Si and decrease the solidification rate of poly-Si. Adjusting the pulse overlap ratio properly can induce the temperature gradient in the x direction, perpendicular to the incident direction of laser, which will increase the grain size in the x direction. It is also found that during the temperature elevation, the Si film with smaller thickness has the higher temperature, but the shorter duration of solidification.

Keywords: poly-Si thin film, laser crystallized technology, specific heat/enthalpy method

摘要

本文提出有關準分子雷射退火之矽薄膜凝固模式。並建立一數學模式，來探討準分子雷射多晶矽薄膜製程中，雷射熱源之各項參數(如雷射能量密度、基板溫度等)對於加熱工件溫度分布之影響。數值方法是採用有限差分法，以等效比熱-熱焓法來處理潛熱之釋放效應，以熱源項處理雷射能量。由模式分析的結果可以發現，二氧化矽層可有效減緩熱量傳遞到玻璃基板，並使熱量集中於 a-Si 薄膜。增加雷射能量密度與玻璃基板溫度可增加矽薄膜的溶化深度，並降低多晶矽的凝固速率。適當調整脈衝雷射覆蓋率可增加 x 方向的溫度梯度。較小厚度的矽薄膜比較大厚度的矽薄膜有較高的溫度，但凝固時間則較短。

關鍵字：多晶矽薄膜、雷射結晶技術、等效比熱熱焓法

Introduction

Poly-Si thin film transistors (Poly-Si TFTs) have become the key element of the Active-matrix Liquid Crystal Displays (AMLCD) in the next generation [1]. Therefore, the poly-Si thin film on the glass substrate has become an important fabricated process. Currently, the processing of poly-Si thin film is mainly due to heating up the amorphous silicon (a-Si) on the glass substrate with the use of excimer laser to make it melt, and then cooling down to form the thin film. However, it is unable to achieve the field-effect mobility of an electronic component using laser-crystallized technology sometimes [1]. This is primarily due to the high power of excimer laser, which causes the silicon film to instantly melt and solidify. This leads to the worse crystal growth and the smaller grain, which hinder the movements of electric carriers. To obtain high field effect mobility, it needs to adjust the control parameters of the laser process. However, the common processing researchers do not know much information regarding change in operating condition that would cause change in the temperature field of the entire work piece. They frequently use trial and error to find the working condition resulting a large expenditure on wealth as well as labor. However, if we can understand in advance about the cause to change in temperature field and how it would be affected under what circumstance, it will be a big help to the research.

Generally, there are two kinds of laser- the continuous and the pulse types. The excimer laser is the latter one. In the

past, laser processing is primarily done by the continuous type and the pulse type is used less. The major applications are concentrated in laser cutting, drilling, welding, repairing and heating etc. The working pieces are thick and most of them are metal. Later on, utilization of the laser has become more widespread, for example, the cutting-off technology of polymer by excimer laser; making poly-Si thin films by using the laser crystallized technology.

The research about heating up working pieces by laser was being done by many researchers. Hsu and Mehrabian [2] discussed the laser processing phenomena such as instant melting and solidifying. Ion et al.[3] used dimensionless method and the empirical data to discuss about the power of carbon dioxide laser and the temperature variation of material. Kou et al. [4] utilized the finite difference method to solve three-dimensional heat transformation problem of laser processing. Chan et al. [5] used a two-dimensional transient model to simulate the flow field of a molten pool under the irradiation of a moving laser. Basu and Srinivasan [6] utilized the finite difference method and a two-dimensional steady model to analyze the flow field in the pool under the irradiation of a static laser. Sluzalec [7] applied the finite element method to analyze the two-dimensional transient flow in the molten pool under the irradiation of pulse laser. Maier et al. [8] used the finite difference method to study the temperature variation during laser processing. Different kinds of laser processes and laser beam shapes were taken into account. Wei et al. [9] utilized different models to analyze laser heat source and to calculate the molten pool

size. Chen and Huang [10] used enthalpy to replace temperature in dealing with the latent heat and the solid-liquid interface. They utilized the finite difference method to analyze the two-dimensional steady melting process under the irradiation of a moving laser. Liou [11] applied the Runge-Kutta method and the experimental measurement to investigate the ablation of PMMA by an excimer laser. Chou [12] used plastic substrate to replace glass substrate and utilizes HP4156 and HP4285 to measure the properties. The experimental results are compared to the simulation results from ATHENA(process simulation) and ATLAS(device simulation). Kuriyama et al. [13] utilized the finite difference method and the experimental scheme to study the influence of substrate temperature and the buffer layer on the solidifying speed.

Some of the researched mentioned above use the analytic or semi-analytic way, or utilize the numerical methods, such as the finite difference method and the finite element method, to solve the heat-transfer problem of laser processing. Material of the work piece is thick and mainly metal or polymer. In the heat-affected zone of laser, it is easy to form a molten pool or have an ablation phenomenon. In this condition, the convective effect of fluid flow in the molten pool or the dispersive effect of micro-particle in the plume should be considered in the numerical simulation.

The main goal of this paper is to establish a mathematical model to investigate the effects of control parameters in the processing of poly-Si thin film by using excimer laser. The basic

structure of the working piece is glass substrate and thin film of silicon. Since the former one has lower thermal tolerance than metal, the temperature of glass substrate should be paid attention to in this study. Because the thin working piece results in very shallow molten pool, in the mathematical model, the convective effect can be ignored. The excimer laser is a bar shape and pulse type photo- source. Due to the silicon film put on a movable platform, the laser can be regarded as a movable heat source, which induces a transient heat-transfer problem with melting and solidifying phenomena.

Theoretical Analysis and Numerical Method

During the annealing process of excimer laser, it is quite difficult to measure the temperature of a-Si thin film. However, it will be feasible to use a mathematical model to study the problem. Figure 3 shows laser beam being shot onto the work piece. The excimer laser is a Gaussian or an evenly distributed pulse-type heat source. There is a layer of porous SiO_2 in between the a-Si thin film and the glass substrate. The SiO_2 layer is utilized to reduce the heat conducted from the silicon film to the glass substrate.

To establish the heat transfer model of the laser processing, the following basic assumptions need to be made.

- (1) The irradiation area of laser is rectangle along the z direction (vertical to the x-y plane, shown in Fig. 1). It is assumed that the laser strength along z-axis is uniform. Accordingly, the problem can be treated as a two-dimensional one.

- (2) Since the thinness of the work piece as well as the laser-heating area is small, the effect of convective heat transfer can be ignored.
- (3) Besides density, all of the other thermal properties vary with temperature. From the assumptions above, the energy equation can be written as

$$\rho C_p^{eff} \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \left(k \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left(k \frac{\partial T}{\partial y} \right) + Q \quad (1)$$

where ρ is density and k is thermal conductivity. Q is the heat source term, which is used to deal with the radiation energy of laser. C_p^{eff} is the effective specific heat. The enthalpy/specific heat method [15, 16] is utilized to take care of the release or absorption of latent heat during the melting or solidifying process. The thermal conductivity is a function of temperature [13].

The initial and boundary conditions of the model are

(1) Initial condition

Before heating the work piece, its temperature can be adjusted as desired. Accordingly, it can be room temperature, or larger or smaller than room temperature.

(2) Boundary condition

- (i) The convective boundary condition is applied to all the surfaces of the working piece except the bottom one.

$$-k \frac{\partial T}{\partial n} = h(T - T_\infty) \quad (2)$$

where n is the normal direction of boundary, h is the coefficient of convection heat transfer, T_∞ is the environment temperature.

- (ii) The boundary condition of the bottom surface can be heat-insulated or a fixed

temperature, which is the same as the initial temperature.

- (iii) At the interface between silicon film and SiO_2 layer or between glass substrate and SiO_2 layer, the temperature and the heat flux are continuous.

In general, since laser beam cannot pass through bulk metal, the boundary of heat flux is used to deal with the irradiation energy of laser. Excimer laser is a pulse-type photo-source, with a wavelength range between 193nm~351nm. According to the radiation theory [17], the wavelength range of thermal radiation is between 100 and 10^6 nm. Consequently, the theory of radiation heat transfer is used to handle the laser heat source. Since the laser beam can transmit the thin working piece, the heat source term in the energy equation is utilized to deal with the laser energy. Based on the Beer's law, the source term Q in Equation (1) can be written as [18~19]

$$Q = I_0 a e^{-az} \quad (3)$$

$$I_0 = \begin{cases} I_i(1-R) & \text{heating zone} \\ 0 & \text{the other place} \end{cases} \quad (4)$$

where I_i is pulse laser power per unit area, a is absorption coefficient, R is reflectivity. The magnitude of reflectivity would directly affect the absorptive energy of work piece from laser. Hatano et al. [20, 21] use an optical method to measure the reflectivity of the a-Si thin film. The reflectivity is obviously affected by temperature, especially when the a-Si film is in the liquid state.

The governing equation was solved by using the finite difference method.

Since the finite difference equations are nonlinear equations, the iterative method is used to solve these equations. Compared with glass substrate, the a-Si film is very thin as well as the primary variation zone of temperature. Accordingly, the unequal-spacing grid is used along the y direction. Because the time between two laser shoots is much larger than the duration time of a laser pulse, the un-uniform time step is utilized.

The release of latent heat during melting and solidifying process would influence the temperature distribution. The enthalpy/specific heat method is introduced to treat the effect of latent heat. It is the combined scheme of the enthalpy and the specific heat methods and has the advantages of these two methods, accuracy and fast convergence. The relationship among temperature, effective specific heat and enthalpy can be written as

$$Cp^{eff} = \frac{e^{n+1} - e^n}{T^{n+1} - T^n} \quad (5)$$

where e is enthalpy and Cp^{eff} is effective specific heat.

Results and Discussion

This paper mainly discusses the influences on the transient temperature field of work piece brought by various working parameters, which are energy density of laser, substrate temperature, pulse overlap ratio and Si film thickness. Furthermore, the influence of convection and radiation boundary conditions on the Si surface is also discussed by comparing the convective and radiative energies from the surface with the input energy of laser. In this study, the repetition rate is fixed at 20 Hz, which means that the period of two

adjacent laser pluses is 0.05 second. The pulse time is 20ns, and the reflectivity of silicon film is 0.75 [21].

The Situation of The Influence by The Function of Pulse Laser

Fig. 2 indicates the central-point temperature of irradiated surface varies with time. Due to the incident laser energy, the temperature goes up rapidly to the melting point (1200K) and stays there for a moment. After melting process, the temperature rises quickly again and reaches the maximum value when the pulse finishes at the twentieth nano-seconds. After that, it goes down to the melting point and stays there for a while. After the solidifying process, the temperature decreases less rapidly. Since the maximum temperature (about 2000K) does not reach the boiling point (3540K), it is not necessary to consider the vaporized effect in the mathematical model.

Comparison Between Two Simulated Methods for The Incident Energy of Laser

Two simulated methods are used to handle the laser energy in solving the temperature distribution: one is the heat-flux boundary condition on the irradiated surface, and the other is the heat source term in the energy equation. To compare these two methods, Fig. 2 is used as the test case and its result is shown in Fig. 3. From the figure, it can be found that the difference between these two methods is not significant. It is mainly because the extremely high absorption coefficient of a-Si ($1.5 \times 10^8 \text{ m}^{-1}$) makes most of the irradiated energy absorbed as soon as it

enters the silicon surface, which is very similar to the heat-flux boundary condition.

Temperature Variation Along the Depth Direction and Influence of SiO₂ Layer

Fig. 4 shows the central-point temperature distribution of the irradiated area along the depth direction (the y direction) changes with time from the twentieth nano-second. The temperature of shallow area (close to the work-piece surface) decreases with time, the temperature of deep region goes the opposite way, and finally the whole temperature approaches a uniform distribution. This phenomenon is due to the effects of laser irradiation and heat conduction. Within the twenty nano-seconds, of laser irradiation, the temperature of the portion near the surface rapidly increases. After that, the heat diffuses into the work piece so that the temperature in the shallow region goes down and in the deep one goes up. This temperature variation proceeds until no diffusion is approached and then the temperature becomes uniform along the y direction.

In Fig. 4, it can also be found that the primary variation region of temperature includes the silicon film and part of SiO₂ layer. The region whose depth is larger than 2 μ m is the glass substrate. The change of glass temperature is not big during the whole working process. The main reason is that the SiO₂ layer can effectively slow down the heat transfer from the silicon film to the glass substrate. This would prevent the substrate from distortion after the processing.

Influence of Energy Density on Temperature

The higher the energy density means the more energy the work piece can obtain. For different energy densities, Fig. 5 indicates the central-point temperature of irradiated surface varies with time. From the figure, it can be observed that the higher energy density has the higher relative temperature, the faster heating rate, and the slower cooling rate. After the laser pulse, the temperature goes down to the melting temperature and stay there for a while. The higher energy density has the longer time staying at the melting point, which means the longer solidification time.

Influence of Substrate Temperature

The substrate temperature can be adjusted by pre-heating and then the work piece is heated by the laser. For different substrate temperatures, Fig. 6 illustrates the central-point temperature of irradiated surface varies with time. Similar to the higher energy density, the higher substrate temperature has the higher relative temperature, the faster heating rate, and the slower cooling rate. The higher substrate temperature has the longer time staying at the melting point. Apparently, it has longer solidification time.

Influence of Overlap Ratio

Under the same condition, the third pulse is chosen for comparison. Fig. 7 shows the temperature distribution along the work-piece surface (x direction) changes with time after the third pulse under the condition that the energy density of laser is 400 mJ/cm², the overlap ratio is 50% and the substrate temperature is

300°C. It can be clearly observed that the temperature of the overlap region is higher and the cooling rate is lower than those of un-overlap area. Besides, between the overlapped and un-overlapped areas, there exists an obvious temperature gradient in the x direction, which would enhance the crystal growth in this direction.

Influence of a-Si Film Thickness

Fig. 8 shows the central-point temperature of irradiated surface varying with time for the a-Si film thicknesses of 0.1 μm and 0.05 μm . From this figure, it can be observed that before 300 nano-seconds, the thickness of 0.05 μm has the higher temperature than that of 0.1 μm . However, the thickness of 0.1 μm stays at the melting temperature (the horizontal part in Fig. 8) longer than that of 0.05 μm . After 300 nano-seconds, both temperatures go down together more slowly than those going down from the highest temperature within 300 nano-seconds. The thickness of 0.05 μm has a little bit lower temperature than that of 0.1 μm . All these temperature variations are mainly due to the low specific heat of a-Si. The smaller thickness has the smaller mass or heat capacity.

Conclusions

This paper is to study the influences on the temperature field of work piece brought by various working conditions. From the computational results, it can be concluded that:

1. In the period of laser irradiation, most of the heat enters the work piece along the y direction, which leads to large temperature gradient in this direction. Contrarily, the temperature gradient in the x direction is less obvious.
2. SiO_2 can effectively slow down the heat transfer from the Si film to the glass substrate and make most of the heat from laser stayed in the Si film.
3. Increasing the energy density of laser and the substrate temperature can have the longer time of solidification.
4. Choosing the proper overlap ratio can induce the temperature gradient in the x direction, which could assist the crystal growth of Silicon in the x direction.
5. The duration of laser pulse is 20 ns. Within 300 ns, the temperature of the thinner Si-film is relatively higher than that of the thicker one. However, the thicker film stays at the melting point longer than the thinner one. After 300 ns, the temperature thinner film is a little bit lower than that of the thicker one.

From these results, it can be concluded that the proposed model could predict the temperature distributions of the working piece and the analyses of different control parameters could provide preferable working conditions, which are good for the growth of large grain of poly-Si

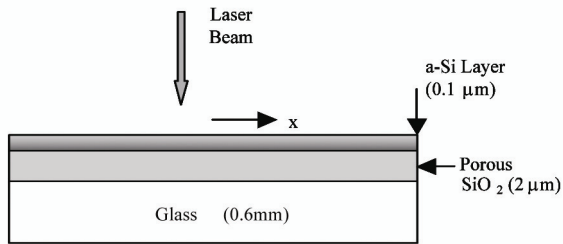


Figure 1. Schematic diagram of laser-heating work piece.

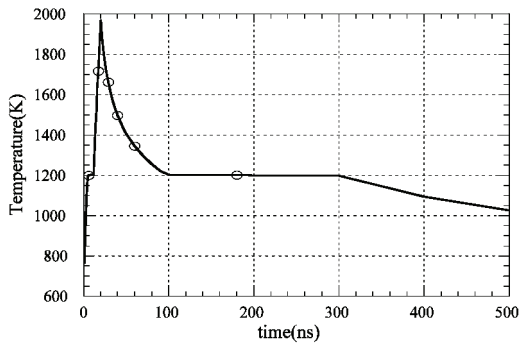


Figure 2 Central-point temperature of irradiated surface versus time for energy density of 400 mJ/cm², and substrate temperature of 300°C.

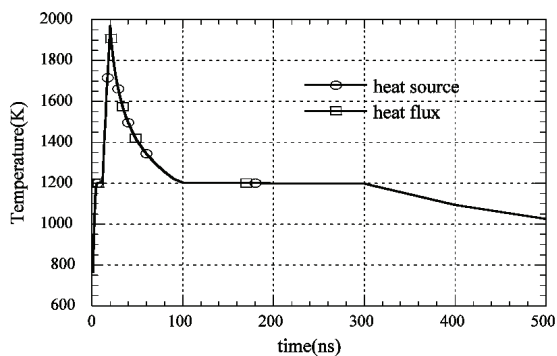


Figure 3 Central-point temperature of irradiated surface versus time for two simulated methods of handling the laser energy.

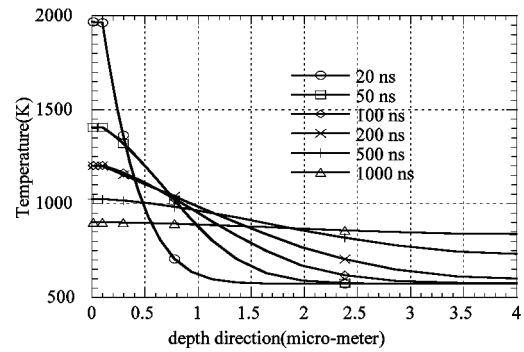


Figure 4 Central-point temperature distribution of the irradiated area along the depth (y) direction varying with time from the 20th ns.

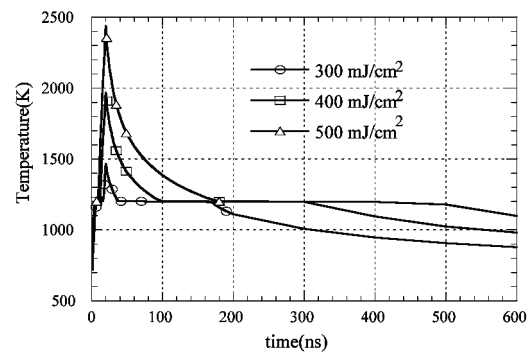


Figure 5 Central-point temperature of irradiated surface versus time for different energy densities of laser.

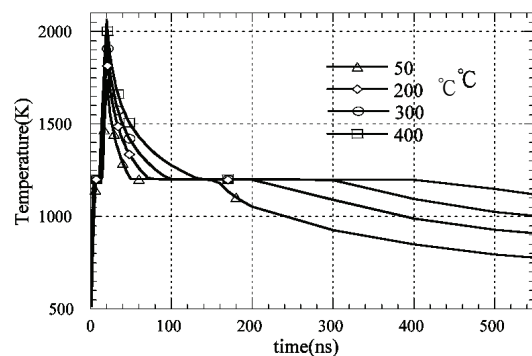


Figure 6 Central-point temperature of irradiated surface varying with time for different substrate temperatures.

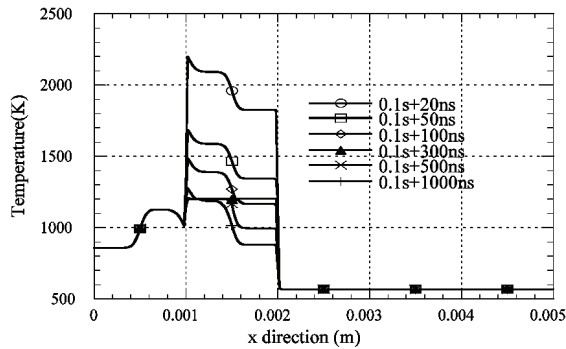


Figure 7 Temperature distribution along the work-piece surface (x direction) varying with time after the 3rd pulse for energy density of 400 mJ/cm², overlap ratio of 50% and substrate temperature of 300°C.

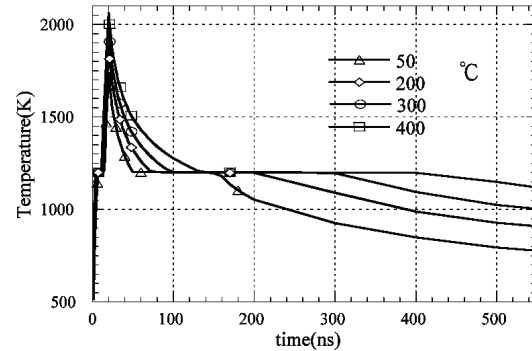


Figure 8 Central-point temperature of laser-irradiated surface versus time for a-Si film thicknesses of 0.1μm and 0.05μm

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