Simulations of Heating Sol-Gel Thin Film by Laser Pulse Train

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Abstract: Simulation of laser pulse-train (25ns, 60 kHz and 3000 pulses) heating Sol-Gel thin film using COMSOL Multiphysics software is investigated. The results show two kinds of temperatures formed on film surface by laser pulse-train heating. One is a single pulse induced transient peak-temperature, which is up to ~1635°C on both Si and glass substrates. The other is the accumulated-temperature generated by accumulating residual-temperature at the end of each pulse period, which is up to ~75°C on Si and ~500°C on glass substrates, respectively. The peak-temperature only depends on the laser intensity. However, the accumulated-temperature not only depends on the laser intensity, but also the pulse repetition rate and the thermal properties of the substrate in particular.

Keywords: Laser pulse train, laser heating, Sol-Gel thin film and heat simulation**.**

1. Introduction

Laser heating has been proven to be an effective and efficient method for low temperature material processing, thanks to the localized heating and minimum thermal impacts to the substrate and the surroundings [1]. Using organic based Sol-Gel material as precursors to fabricate functional thin film by direct laser heating has attracted a enormous attention in terms of achieving cost-effective and environmentally viable film deposition. Different from the hard and dense film, as-spun Sol-Gel film appears relatively soft and porous, and hence the laser heating becomes more challenge. For example, laser intensity has to be kept at an appropriate level to complete annealing and avoid material removal. So far, there are some reports on heating Sol-Gel thin film using continuous wave (CW) and pulsed lasers [2-4]. The CW laser heating is able to sustain heat for a relatively longer time. However, it is strongly dependent on the thermal properties of the material. Instead, the pulsed laser heating is more efficient and independent on the thermal properties of the material. But, it only sustains heat within pulse duration. Laser pulse-train heating offers an alternative method, because of it's unique function in combining the CW and the pulsed laser heating. Laser pulse train heating of Sol-Gel film has been experimentally investigated in our group.

Numerical simulation of laser heating is a very important approach to understand the temperature spatial/temporal profile, since it is difficult to accurately measure the localized temperature [5-7]. However, the simulation of laser pulse-train heating is vey challenging and needs more computing time in comparison with the CW and the pulsed laser heating due to the small duty-cycles of pulse-train.

In this paper, we report for the fist time to the best of our knowledge the numerical simulation of pulse-train heating Sol-Gel thin film using COMSOL Multiphysics software.

2. Description of Simulation Models

2.1 Laser Source

The laser used in the heating experiment is 266nm DPSS laser (Coherent AVIA series) with pulse duration of 25ns, and repetition rate of 1-300 kHz. A beam shaper was used to convert the Gaussian beam profile to 1D top-head distribution, as shown in Figure 1. The beam dimension is 900um (2*Wy) x 230um (2*Wx) @1/e^2.

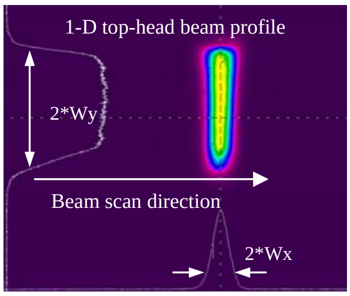


Figure 1: Measured 1D top-head beam profile using CCD camera via Fluorescence glass plate

The intensity of the 1D top-head laser beam is expressed by equations (1)–(3).

Geometry of the top-head laser beam:

$$g = e^{-2(\frac{x-x_0}{W_x})^2 - 2(\frac{y-y_0}{W_y})^{10}} \qquad (1)$$

Beam dimension:

$$S = 2\sqrt{2} * W_x * W_y \qquad (2)$$

Laser intensity:

$$I_0 = g * P_0 / S \qquad (3)$$

Figure 2 shows a schematic diagram of the pulse-train with 25ns pulse duration, 60 kHz frequency, and a duty cycle of 1/664 (16.6μs of pulse period).

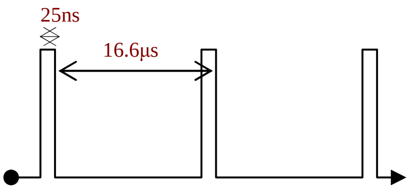


Figure 2 shows a schematic diagram of pulse-train

Design of an appropriate pulse-train function is essential to the numerical simulation. Figure 3 shows a design of the laser pulse-train function using a discontinuous sign (x) function, which is expressed in equation (4)

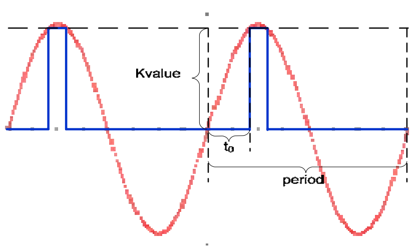


Figure 3: Schematic drawing of pulse-train function

$$PulseTrain(t) = 0.5 * (sign(\sin(\frac{2\pi(t+t0)}{period}) > Kvalue) - 0.5) + 1) \qquad (4)$$

In COMSOL, the sign(x) function was replaced by a smoothed sign function with a continuous first derivative, as shown in equation (5).

$$y = flsmsign(x, scale) \qquad (5)$$

The transition within the interval –scale < x < scale was smoothed.

Design of an appropriate time stepping for pulse train evolution is another essential task to the success of the simulation. This is mainly due to the fact that the pulse-train has a very small duty cycle of 1/664, thus tremendous computing time is required to solve this model. An adaptive time stepping is designed in this study to reduce the computing time without sacrificing the simulation accuracy.

The laser irradiation was modeled as heat flux in the simulation. Low laser powers between 0.1 and 0.8W at 60 kHz were used in both experiment and simulation to avoid the material removal by laser. Table 1 shows the conversion of laser power to laser peak power and laser intensity.

2.2 Geometry and Meshing in Simulation Models

Figure 4 shows the geometry of the simulation model, based on the actual material and structure.

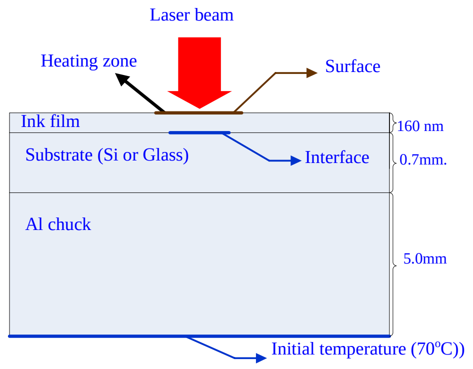


Figure 4: Schematic drawing of geometry of simulation model

(i). Since the 160nm thick Sol-Gel film is much thinner than the substrate, and has very large absorption at 266nm wavelength (absorption coefficient of ~10^5cm^{-1}) the film behaves like light absorber, and has little contribution to heat conduction. (ii). 0.7mm thick crystalline Si and BK7 glass were used as the substrate, respectively. The substrates dominated the heat conduction. (iii). In order to comply with experiment, a 5.0mm thick Al-chuck with initial temperature of 70^{o}C (343k) was also used in the model. Figure 5 shows the mesh configuration. Only laser irradiated zone was finely meshed to ensure the simulation accuracy and the computation efficiency.

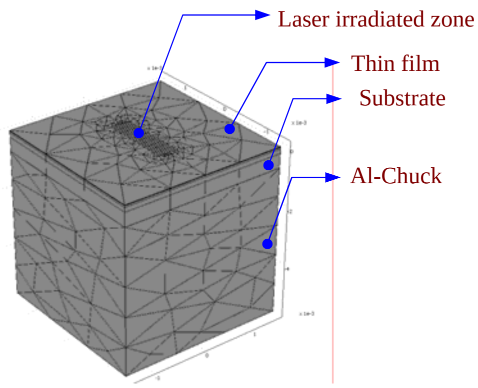


Figure5: Mesh configuration of the model

3D heat conduction model was solved by using COMSOL Multiphysics 3.5 version. In the simulation, "Heat Transfer Module" and "Transient Analysis" were chosen to solve the heat conduction equation (6)

$$Q = \delta_{ts}\rho C_p \frac{\partial T}{\partial t} - \nabla[\kappa \nabla T] \qquad (6)$$

In this simulation, three assumptions were made:

(i) Laser is fully absorbed at the film surface due to the high absorption coefficient (~10^5 cm^{-1}) of Sol-Gel material at 266nm wavelength;
(ii) The thermal conductivity of Sol-Gel film is constant;
(iii) Heat convection and radiation are neglected.

The measured thermal conductivity of the Sol-Gel film is ~0.1W/m*K, which indicates a polymeric character. Here, we also assume a heat capacity of 1500 J/kg*K and density of 1000kg/m^3 for Sol-Gel material. The thermal conductivities of Si and glass are function of temperature, as shown in Figure 6. The rest parameters were obtained from COMSOL material library. All of the sub-domain settings are listed in Table 2.

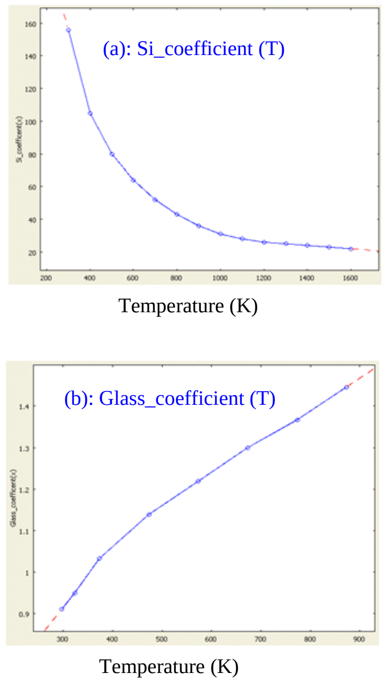


Figure 6: Thermal conductivity (unit: W/m*K) of (a) Si and (b) glass substrate as a function of temperature (unit: K)

In the real experiment, laser beam moved at a speed of vx =0.1mm/s. This gives 2.3s dwell time of the laser beam at one position as a result of 2*Wx/vx, which indicates 13800 pulses fired at this position. However, heat simulation with a pulse-train of 13800 pulses is beyond our computer capability. For the purpose of feasibility investigation, a pulse-train with 3000 pulses was applied to the simulation.

3. Results and Discussion

The results will be presented in describing the heat field on the film surface, including (i) peak-temperature (T_p) generated by individual pulses and (ii) accumulation-temperature (T_A) by accumulating residual-temperature (T_R) of individual pulses. As shown in Figure 7, each pulse first generates a peak-temperature within the pulse duration, and then the temperature quickly decays and leaves a residual-temperature at the end of each pulse period. The residual-temperature will be the initial-temperature (T_i) for the next coming pulse. After certain number of laser pulses (No#i, i=1-3000), the accumulation of residual-temperature becomes pronounced, as expressed in equation (7).

$$T_{A(i)} = T_i + \sum_{i=1}^{3000} T_{R_i} \qquad (7)$$

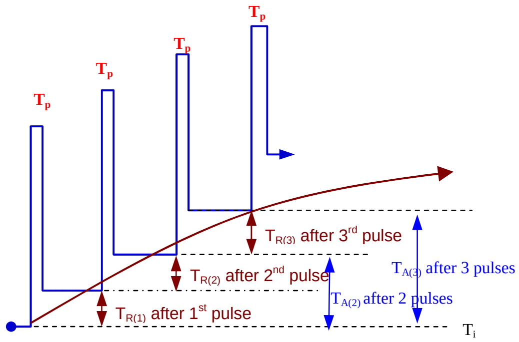
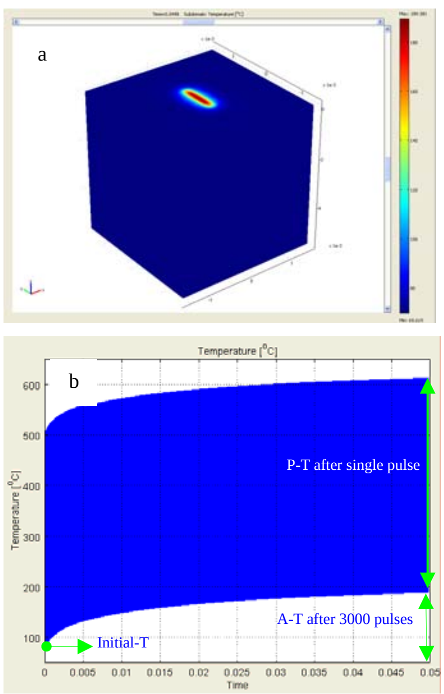


Figure 7: Schematic drawing of peak temperature and residual temperature with number of pulses



Figures 8: (a) simulated 3D temperature field; (b) temperature increase with increase of number of pulses.

Figure 8 shows (a) the simulated 3D temperature field, and (b) the plot of the surface temperature as a function of time stepping, corresponding to a sequence of 3000 pulses. The substrate is glass and the laser intensity is 133kW/cm^2.

Above result clearly shows that the surface temperature is the sum of the peak-temperature and the accumulation-temperature. The peak-temperature is independent on the number of pulses. The accumulation-temperature, however, increases with increasing number of laser pulses.

In the following part, we will discuss influences of laser intensity and thermal properties of substrate on the peak and accumulation temperature, respectively.

Figure 9 shows the simulated peak-temperatures of the film on Si and glass substrates, respectively versus laser intensity. The peak-temperature linearly increases with increasing the laser intensity. There is little difference of peak-temperature on Si and glass substrate. This is due to the fact that the dwell time of peak temperature is too short (~25ns) to differentiate the two substrates.

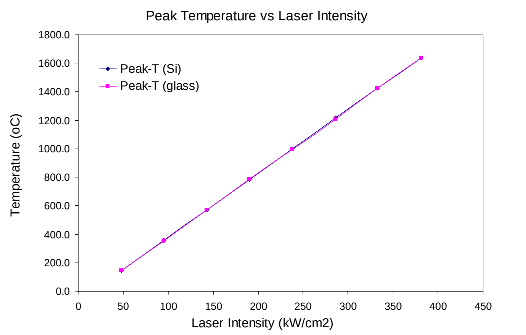


Figure 9: Peak temperature of the film on Si and glass substrate versus laser power density.

Figure 10 shows plots of accumulation-temperature of the film on (a) Si and (b) glass substrates, respectively, as the function of time stepping at laser intensity from 48 to 381kW/cm^2 with an initial temperature of 70^{o}C. In Figure 10, the accumulation-temperature first rapidly increases at time stepping <~0.1s (corresponding to 1000 pulses), and then towards to saturation with further increasing time stepping due to the heat dissipation via substrate. (1) For the case of using Si substrate, after 3000 pulses the accumulation-temperature does not significantly increase. For example, at the laser intensity of 381kW/cm^2 the accumulation-temperature is only 76.13^{o}C, which is only 6.13^{o}C above the initial-temperature. The low accumulation-temperature mainly results from the high thermal conductivity of silicon. Thus, when using Si substrate, the accumulation-temperature has little contribution to the heating process. (2) However, for the case of using glass substrate, after 3000 pulses, the accumulation-temperature largely increases. For example, at the laser intensity of 381kW/cm^2 the accumulation-temperature reaches ~500^{o}C, which is 430^{o}C above the initial temperature. The high accumulation-temperature attributes to the low thermal conductivity of glass. So, when using glass substrate, the accumulation-temperature plays an important role in the heating process.

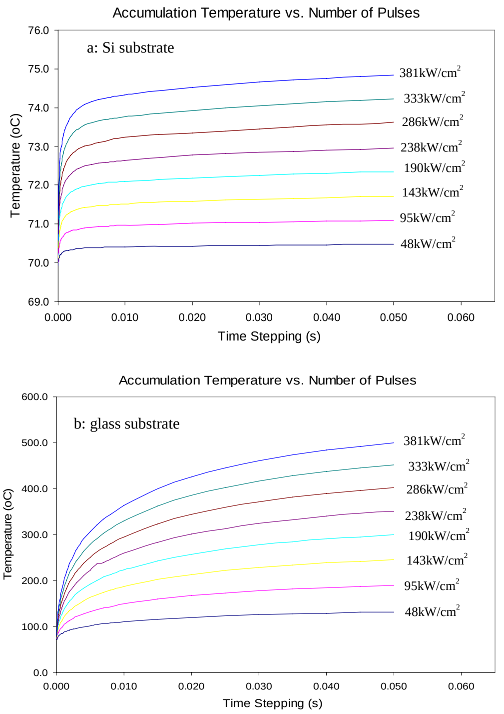


Figure 10: Plots of accumulation temperature as a function of time stepping at laser power density of 48, 95, 143, 190, 238, 286, 333 and 381kW/cm2, respectively.

Figure 11 show a linear increase of the final accumulation-temperature (after 3000 pulses) of the film on Si and glass substrates, respectively as a function of the laser intensity. This result indicates that the formed accumulation-temperature on Si substrate is too small to affect the material processing even at reasonably high laser intensity. Instead, the accumulation-temperature on glass is pronounced and comparable to the peak-temperature, which will definitely contributes to the material annealing process.

In this study, the laser pulse-train only included 1/46 of total applied 138000 pulses (2.3s dwell time at 60 kHz) at one position, due to the limits of computation. However, according to the trend of the accumulation-temperature increase as function of the number of pulse, the increase of the accumulation-temperature after 3000 pulses seems to be not in a large magnitude especially for the case of using Si substrate.

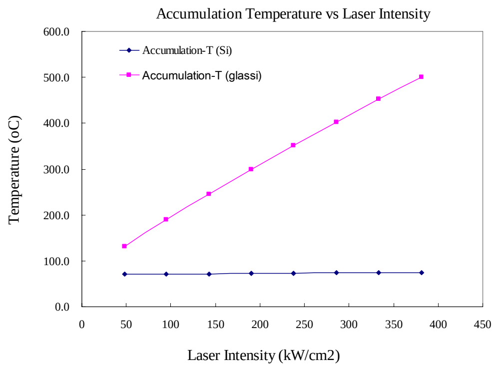


Figure 11: Accumulation temperature of thin film on Si and glass substrate, as a function of laser intensity

4. Conclusion\

Our numerical simulations demonstrate that laser pulse-train heating produces transient peak-temperature by individual pulse and accumulation-temperature by accumulating the residual heat, respectively. Transient peak-temperature is only laser intensity dependent. However, the accumulation-temperature is strongly dependent on (i) thermal conductivity of material, (ii) pulse repetition rate (pulse period), and (iii) laser intensity. In order to achieve high accumulation heat, low thermal conductive material and short pulse period are required. Further investigation on simulating the laser heating process with a moved laser pulse-train is ongoing.

5. Nomenclature

Q	Heat -source (W)
δ_s	Time-scaling coefficient
ρ	Density (kg/m3)
C_p	Heat capacity (J/kg*K)
κ	Thermal conductivity (W/m*K)
T	Temperature (K)
t	Time (s)

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7. Appendix

Table 1: Conversion of laser power to laser intensity at 25ns pulse width and 60 kHz repetition rate

Laser power (P:W)	Laser Peak Power (P0: W)	Laser Intensity(I0: kW)
0.1	67	48
0.2	133	95
0.3	201	143
0.4	267	190
0.5	335	238
0.6	402	286
0.7	469	333
0.8	536	381

Table 2: Sub-domain settings of the simulation models

	κ (W/m*K)	C_p (J/kg*K)	ρ : (kg/m^3)
Film	0.1	1500	1000
Si	Si_coefficent (T)	C_p (T) (COMSOL material library)	ρ (T) (COMSOL material library)
Glass	Glass_coefficent(T)	2540	708
Al	κ (T) (COMSOL material library)	C_p (T) (COMSOL material library)	ρ (T) (COMSOL material library)