NUMERICAL SIMULATION OF PULSE LASER ABLATION

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Abstract — Pulsed laser deposition is a powerful technique for thin film deposition of various materials. Pulsed laser ablation is an important process in the Pulsed laser deposition technique. This paper presents a comprehensive numerical model using finite element method considering temperature dependent material properties, plasma shielding effect and temperature dependent absorptivity and absorption coefficient to predict temperature distribution and ablation depth. The numerical simulation is performed using TiC as target material. The model takes into account the effect of delay between successive pulses during multiple laser pulses to predict temperature distribution and ablation depth.

INTRODUCTION

Pulsed Laser Deposition (PLD) is a simple and highly versatile technique to grow thin films of materials of good quality. It is a simple technique in which laser energy pulses are used to remove material from the surface of a target. High power laser pulses are directed on the target. When the target material is exposed to laser energy, sufficient heating of surface takes place leading to the melting and evaporation of the target material. This process is also called laser ablation. This ablation process produces a plasma plume, which expands rapidly away from the target surface. The ablated material collects on a substrate that is kept some distance away from the target, upon which it condenses leading to the formation of a thin film. There are a large number of variables that affect this process such as laser fluence, background gas pressure and substrate temperature. Depending upon the individual application these variables change. Much of the early research on PLD focused on individual materials and applications, rather than understanding processes occurring during material transportation from target to substrate. Even though the PLD technique is widely used, the fundamental processes occurring during the transfer of material from target to substrate are not fully understood and are subject of current research [1, 2].

The PLD technique has significant benefits compared to other film deposition techniques such as: i) relatively high deposition rates. ii) Stoichiometric transfer of material from target to substrate. iii) Extremely clean process (since an external energy source is used). iv) Housing of a number of target materials is possible by using a carousel, so multilayer films can be deposited without breaking of vacuum when changing between materials [2]. Among the research applications of PLD are: high temperature superconducting thin films, coatings for tribological applications, biomedical applications, manufacturing of micro or nano components [2, 3].

THERMAL MODELLING

Bulgakova and Bulgakov [4] developed a numerical model to evaluate the ablation rate and temperature distribution in the target under near threshold ablation conditions. They considered three mechanisms associated with the ablation process, namely, normal vaporization, normal boiling and explosive boiling (phase explosion). They considered the material removal mechanisms in PLA with infrared nanosecond pulses which are typically used for thin film deposition. Based on the measurements of ablation rate as a function of laser fluence, evidence for the transition from normal vaporization to phase explosion was obtained for a number of materials. They did not consider the non-thermal (electronic) processes induced by infrared laser radiation in the target. The time dependent temperature distribution along the target depth T (z, t) was given by the one dimensional heat flow equation as,

$$C_p\rho\left(\frac{\partial T}{\partial t}-u(t)\frac{\partial T}{\partial z}\right)=\frac{\partial}{\partial z}k\frac{\partial T}{\partial z}+[1-R]BI_s(t)\exp(-Bz) \quad (1)$$

where, C_p, ρ, k, R and B are specific heat, density, thermal conductivity, reflectivity and absorption coefficient of target material and u (t) is the velocity of surface recession. The value of u (t) is calculated considering that vaporized material flow follows the Hertz-Knudsen equation and the vapour pressure above the vaporized surface is given by the Clausius-Clapeyron equation. The laser light intensity reaching the target surface is given by

$$I_s(t)=I_0(t)\exp[-\Lambda(t)]=I_0(t)\exp\left[-\int_0^\infty \alpha_p(n,T)dz\right] \quad (2)$$

where, I_0 (t) is incident laser intensity, α_p (n, T) is the plasma absorption coefficient, which depends on the plasma density and temperature. Λ (t) is the total optical thickness of the plasma given by

$$\Lambda(t)=a\Delta z+bE_a \quad (3)$$

here, Δz is penetration depth per pulse, E_a represents density of absorbed radiation energy, a and b are time independent coefficients given by

$$a=\frac{\rho f(T_v)}{m}, \qquad b=\frac{(\gamma-1)}{k_b}\frac{\partial f}{\partial T}\bigg|_{T_v} \quad (4)$$

where, T_v is the temperature at which particles vaporize. The experimental evidence for the transition from normal vaporization to phase explosion during PLA of graphite, niobium and YBCO superconductor was obtained and the corresponding values of threshold fluence were determined. The thermal heating process in the irradiated targets was characterized using model calculations.

Fang *et al.* [5] described a model considering both the vaporization effect and the plasma shielding effect for the high power nanosecond PLA of multi-elemental oxide superconductors. They solved the heat conduction equations with initial and boundary conditions to describe the target temperature, taking vaporization and plasma shielding into account. The following balance equation can be written before vaporization sets in

$$C_p\rho\frac{\partial T}{\partial t}=\frac{\partial}{\partial x}k\frac{\partial T}{\partial x}+B\beta I_s(t)\exp(-Bx) \qquad (0<t\le t_{th}) \quad (5)$$

where C_p, ρ, k, B are as mentioned earlier, β is absorptivity of the target and τ is the width of laser pulse. After considering the vaporization and plasma shielding effects, the temperature of the target in the period from $t=t_{th}$ to t=τ is given by

$$C_p\rho\left(\frac{\partial T}{\partial t}-u(t)\frac{\partial T}{\partial x}\right)=\frac{\partial}{\partial x}k\frac{\partial T}{\partial x}+B\beta I_s(t)\exp(-\alpha_{IB}H)\exp(-Bx) \qquad (t_{th}\le t\le\tau) \quad (6)$$

where, u (t) is as mentioned earlier, α_{IB} is the inverse absorption length and H is plasma length. The incident laser intensity expressed by Gauss function, given by

$$I_0(t)=I_0\exp\left[\frac{-(t-\tau_m)^2}{\tau^2}\right] \quad (7)$$

where, I_0 is the maximal laser power density, τ_m is the time at which the power is maximum and τ is the time corresponding to the full width at half maximum pulse width.

FINITE ELEMENT MODELLING

To simulate laser-matter interaction, a finite element numerical model [7] has been developed using the commercial software ANSYS 11 [8]. This time dependent problem was solved sequentially, considering incremental time step of 1ns over a time period of 20 ns. During the simulation, the output of the preceding time step becomes an input to the succeeding time step. The target is represented by a mesh of finite elements. The target initial temperature is 298 K. When the temperature of an element is higher than the melting point temperature (Tm) at the end of a particular step, melting is assumed to have occurred and latent heat of melting (Lm) is taken into account for calculation in the model. The ablation is assumed to occur when the temperature of the surface elements is higher than the boiling point temperature (Tb), in this condition also, the model takes into account the phase change effect by considering the latent heat of vaporization (Lv) into calculation. Material removal in ANSYS is achieved through killing of the elements, which are deactivated by multiplying their stiffness (or conductivity or other analogous quantity) by a severe reduction factor. All the properties associated with the deactivated elements are set to zero, such as elements loads, specific heat, mass and damping.

Target representation

The target geometry used for simulation is given in Fig. 1. To minimise the computer processing time, the target is supposed to be rectangular in shape with dimension of 10µm x 3µm and only half of the target is simulated because of the axial symmetry of the problem. Moreover, only half of the simulated region is irradiated in order to account for the lateral heat losses to the non-irradiated part of the sample. The size of each element is 20nm x 20nm.

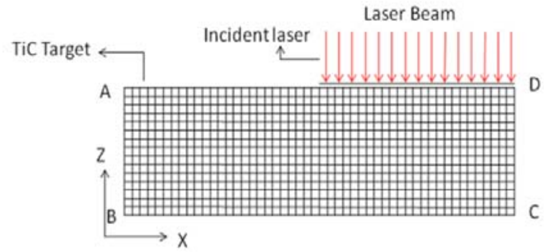

Fig..1. Geometry for laser ablation of target [7]

The element used for the analysis is PLANE 55. PLANE 55 can be used as plane element or as an axi-symmetric ring element with 2D thermal conduction capability. The element has four nodes with single degree of freedom, temperature, at each node. The element can be applied for 2D, axi-symmetric, steady state or transient thermal analysis.

Assumptions

The following assumptions are considered in the model [7]

- A 2D heat conduction equation is assumed.
- Heat source distribution is assumed to be of temporal Gaussian profile.
- Since the molten volume of the target material is very low in PLA process, Convective heat transfer effects are not considered.
- Due to very small interaction time between laser and target surface, Radiation effect has also not been taken into account.

Considerations

The following aspects are considered in the model [7]

- Thermal analysis is transient in nature.
- Temporal Gaussian profile of laser.
- Temperature dependent mechanical and thermal properties of material.
- Dynamic absorptivity and absorption coefficient.
- Absorption of laser in plasma.
- Delay between pulses.

Temperature dependent material properties

Both temperature profile and the ablation depth in PLA process vary with material properties such as thermal conductivity and specific heat, therefore it is necessary to consider the temperature dependent material properties. The model is simulated for titanium carbide (TiC) material. The material properties of TiC [21] are given in Table 1 and Table 2

Table 1. Temperature-dependent properties of TiC [6]

Temperature-dependent properties of TiC									
T (K)	298	400	800	1200	1600	2000	2400	3340	5080
K [W/(m K)]	23	24	31	36	40	43	45	8	4
Cp [J/(kg K)]	550	650	830	890	900	915	930	950	950

Table 2. Material properties of TiC [6]

Material properties of Titanium carbide						
Tm (K)	Tb (K)	ρ (kg/m^3)	Lm (J/kg)	Lv (J/kg)	a (m$^-$	b (m$^-$
3340	5080	4910	10^6	10^7	4 x 10^5	5 x 10^{-4}

BOUNDRY CONDITIONS

Equations (8) and (9) give the heat conduction equations that are solved for entire domain during the analysis within different temperature limits.

$$-k(T)\frac{\delta^2 T}{\delta z^2} = Q(z,t) \quad T<Tm$$

(8)

The above condition implies that incident laser energy onto the surface is conducted into the work piece, when target temperature is less than its melting temperature.

$$-k(T)\frac{\delta^2 T}{\delta z^2} = -L + Q(z,t)$$

T>Tm

(9)

It implies that when the target temperature exceeds its melting point, latent heat (L) comes into picture.

The boundary conditions are given by following equations

$$T(x,t)\big|_{z=0} = T_0$$

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The above two conditions are under the assumption that AB and BC are far away and at the same temperature.

$$-k(T)\frac{\delta T}{\delta z}\bigg|_{x=10\mu m} = 0$$

Initial condition

Initially the temperature of the body is equal to ambient temperature

$$T(z,0) = T_0$$

RESULT AND DISCUSSION

Ablation depth per pulse

In the model, the ablation depth per pulse is calculated by killing the elements. When the temperature of the elements is higher than the boiling point temperature, these elements are assumed to be ablated. In this condition too, the model takes into account the phase change effect by considering the latent heat of vaporization in the calculation. The ablation depth is calculated knowing the numbers of elements of the mesh that reach the boiling temperature for TiC (5080 K). The ablation depths and temperature profiles calculated for a single laser pulse of 20 ns on-time for fluence levels of 4, 7, 10 and 15 J/cm^2. Fig 2 shows temperature profile and ablation depth for laser fluence of 15 J/cm^2

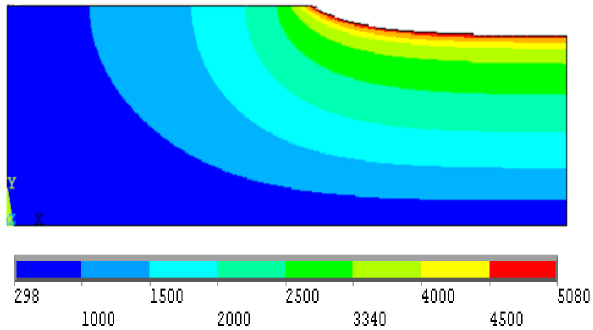

Fig. 2. Temperature distribution profile for laser fluence 15 J/cm^2

Consideration to off-time (Delay between successive pulses)

In this model the off-time is considered after single pulse on-time. The on-time has been taken as 20 ns and off-time has been considered to be 20 ns in the simulation. During the off-time, target is cooled by radiation and conduction heat losses, so the target temperature decreases. During the off-time the heat conduction takes place from high temperature to low temperature of target. The model does not consider the convective heat transfer. The thermal conduction equation is given by

$$Q_{conduction} = -K\frac{\delta T}{\delta Z}$$

(10)

The surface cooling of the target material due to radiation during the off-time has also been considered and is given by

$$Q_{radiation} = \in \sigma(Ti^4 - Tj^4)$$

(11)

It is understand that this effect is negligible for short pulse ablation process because there is very less time available between the two successive pulses for this effect to take place. We consider this effect in order to observe the overall effect of each mechanism governing the ablation process.

Temperature variation with on-time and off-time

Figure 3 shows surface temperature variation during on-time and off-time for the laser fluences in the range of 4 J/cm^2 to 15 J/cm^2 obtained after single pulse.

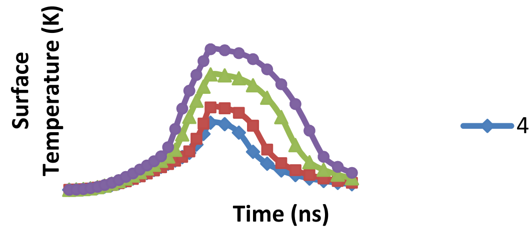

Fig. 3. Surface temperature variation during on-time and off-time after first pulse.

In the Fig. 3, first 20 ns represents on-time. During the on-time, temperature of target increases. Similarly, as the fluence level increases, the target temperature increases. At the end of 20 ns, the maximum surface temperature obtained for the laser fluence of 4, 7, 10 and 15 J/cm^2 is 5172.22 K, 6284.4 K, 8670.72 K and 10507.1 K, respectively. It is observed that, the pattern of increase in temperature with time is similar for all laser fluences (Fig. 3.7).

The next 20 ns is off-time. During the off time temperature of target decreases. The pattern of decrease in temperature is again similar for all the laser fluences. At the end of off- time, at 40 ns, the temperature of target for laser fluence of 4, 7, 10 and 15 J/cm^2 are 693.673 K, 801.867 K, 1135.55 K and 1531.91 K, respectively.

Figure 4 shows the surface temperature variation during on-time and off-time obtained after fifth pulse. Temperature at the end of fifth pulse on-time for laser flunces 4, 7, 10 and 15 J/cm^2 is increased by an ammount of 271.4 K, 305.7 K, 401.53 K, and 486.2 K respectivly compared to the temperature at the end of first pulse on-time. Temperature at the end of off- time after fifth pulse for laser fluence 4, 7, 10 and 15 J/cm^2 is 732.75 K, 839.82 K, 1267.33 K and 1671.45 K, respectivly which is greater than temperature at the end of off-time after the fourth pulse.

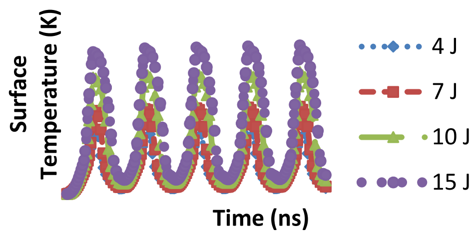

Fig. 4. Surface temperature variation during on-time and off-time after fifth pulse.

we can say that, in the case of multiple laser pulses, the temperature variation in each pulse follows the same pattern during on and off-time. In each successive pulse, temperature of the target increases at the end of on-time and off-time compared to previous pulse.

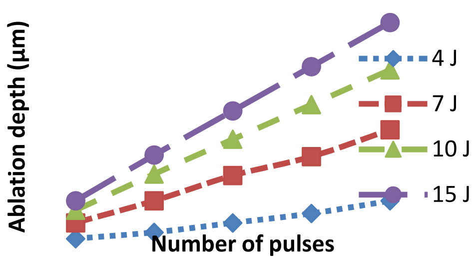

Fig. 5. Ablation depth vs Number of pulses

The ablation depths obtained after a number of pulses for laser fluences of 4, 7, 10 and 15 J/cm^2 are shown in Fig. 6. It is observed that for constant laser fluence, as the number of pulses increase, the ablation depth also increases. At the lower fluence levels, the ablation depth obtained after each successive pulse is more than the ablation depth obtained after previous pulse. Whereas, at the higher fluence levels, the ablation depth obtained during each pulse increases by the same factor. For the same pulse, as the laser fluence increases, the ablation depth also increases.

CONCLUSION

A finite element model of the laser matter interaction to predict ablation depth has been developed. The model takes into consideration temperature dependent material properties, dynamic absorptivity and absorption coefficient of the target material, plasma shielding, time dependent ablation and delay between successive pulses. The model is solved using ANSYS 11 software to determine temperature variation and ablation depth for multiple laser pulses. Major conclusions from this work are summarised below;

- The ablation depth obtained increases with an increase in laser fluence.
- In the model delay between successive pulses is considered. During the delay between pulses (off-time) the target surface is cooled by conduction and radiation.
- Temperature obtained at the end of on-time of successive pulse is greater than temperature at the end of on-time of previous pulse.
- The ablation depth obtained after second pulse is approximately double the ablation depth obtained after first pulse.
- In the multipulse laser ablation, the ablation depth increases approximately by the same factor during each successive pulse.

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