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Finite element simulation of pulsed laser ablation of titanium carbide

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Abstract

In the present paper, a 2D finite element model based on the heat-conduction equation and on the Hertz-Knudsen equation for vaporization was developed and used to simulate the ablation of TiC by Nd:YAG and KrF pulsed laser radiation. The calculations were performed for fluences of 8 and 10 J/cm^2, which according to experimental results obtained previously, correspond to large increases of the ablation rate. The calculated maximum surface temperature of the target for both lasers is higher than the estimated value of TiC critical temperature, corroborating the hypothesis that the increase of the ablation rate is explained by the explosive boiling mechanism.

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Keywords: Laser ablation; Finite element simulation; Titanium carbide

1. Introduction

Titanium carbide (TiC) is a transition metal carbide widely used as a coating material because of its unique combination of physical and chemical properties, such as high melting point, high hardness and high wear resistance [1]. Several techniques have been used to deposit TiC thin films, including chemical vapor deposition (CVD) [2], plasma-enhanced CVD [3], laser CVD [4], ion-beam assisted deposition (IBAD) [5], plasma spraying [6], magnetron sputtering [7] and pulsed laser deposition (PLD) [8–14]. Among these methods, the latter process is particularly promising because of its simplicity, flexibility and capability to preserve the stoichiometry of the material [15].

To optimize the quality of TiC films deposited by PLD, it is of fundamental importance to understand the ablation mechanisms involved. For instance, D'Alessio and co-workers [10,11] reported a non-linear variation of the ablation rate of TiC as a function of the radiation fluence, using a frequency doubled Nd-YAG laser ($\lambda = 532$ nm). In the fluence range between the ablation threshold (0.5 J/cm^2) and 8 J/cm^2 the ablation rate initially increases with fluence, then becomes approximately constant, as expected from a plasma mediated vaporization process [16]. Above 8 J/cm^2, the ablation rate increases drastically and the deposited films become covered by numerous droplets of resolidified material. A similar behaviour was observed more recently by Oliveira et al. [17] in a study of the pulsed laser ablation of TiC using a KrF excimer laser ($\lambda = 248$ nm) but the threshold for increasing ablation rate was 10 J/cm^2. Similar variations of the ablation rate were also observed in the PLD of other materials, and explained by the occurrence of explosive boiling (or phase explosion) [18–20]. According to Martynyuk, Kelly and Miotello [21–23], explosive boiling is the primary material ablation mechanism for sufficiently high fluences and short pulse durations. In these conditions, the target will reach temperatures near the thermodynamic critical temperature of the material. At this point, the rate of homogeneous bubble nucleation rises catastrophically and the target makes a transition from superheated liquid to an equilibrium mixture of vapor and liquid droplets.

In the present paper, a 2D finite element model (FEM) was developed and applied to the simulation of the pulsed laser ablation of TiC using Nd:YAG ($\lambda = 532$ nm) and KrF ($\lambda = 248$ nm) laser radiation. The calculations were performed for fluences of 8 and 10 J/cm^2, and the possibility of explosive boiling was investigated. The results achieved suggest that explosive boiling is probably the mechanism responsible for the large increase of the ablation rate observed experimentally at those fluences.

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2. Numerical simulation

2.1. Background

In the absence of convective and radiative energy transport, the temperature distribution T induced by absorption of laser radiation in a material is given by the heat conduction equation, which, in 2D, can be written as [24]:

$$\rho(T)C_p(T)\frac{\partial T(x,y,t)}{\partial t} = \nabla[k(T)\nabla T(x,y,t)] + Q(y,t) \quad (1)$$

where x and y are the space coordinates and ρ, C_p, k the mass density, specific heat at constant pressure and thermal conductivity of the target material. The source term $Q(y, t)$ represents the laser energy absorbed by the sample and is expressed as:

$$Q(y,t) = I_s(1-R)\alpha\exp(-\alpha y) \quad (2)$$

where R and α are the reflectivity and the absorption coefficient of the target material, y the spatial coordinate in the direction normal to the sample surface and I_s is the temporal laser irradiance at the sample surface. According to Bulgakov and Bulgakova [25], I_s is given by:

$$I_s(t) = I(t)\exp[-\Lambda(t)] \quad (3)$$

where Λ is the optical thickness of the ablation plume, is given by $\Lambda(t) = ah(t) + bE_a(t)$. In this expression, a and b are time-independent coefficients, h the ablation depth and E_a is the density of the radiation absorbed by the plasma. The laser beam temporal profile $I(t)$ is supposed to be Gaussian with full width at half maximum τ.

For materials with a metallic electronic structure, such as TiC [26], ablation occurs essentially by thermal ablation processes [24]. According to Kelly and Miotello [22,23], there are three regimes of thermal ablation: vaporization, heterogeneous boiling and explosive boiling (or phase explosion). Only vaporization and explosive boiling are compatible with the time scale of nanosecond pulse duration laser. Since explosive boiling only occurs when the target reaches temperatures near the thermodynamic critical values of the material [21–23], the ablation mechanism assumed in the model is vaporization. The flow of material vaporized from the surface follows the Hertz-Knudsen equation, leading to the ablation rate v given by:

$$v(T) = (1-\beta)\sqrt{\frac{m}{2\pi k_B T}}\frac{p_0}{\rho}\exp\left[\frac{L_V}{k_B}\left(\frac{1}{T_B}-\frac{1}{T}\right)\right] \quad (4)$$

where T_B is the boiling temperature at the pressure p_0, k_B the Boltzmann constant, β the back flux coefficient and L_V is the latent heat of vaporization of the material. In Eq. (4), it is assumed that no vapor is present in the atmosphere and that the vapor pressure above the vaporized material is given by the Clausius-Clapeyron equation.

2.2. Finite element model (FEM)

To simulate the ablation of TiC, a finite element numerical model was developed using the commercial package ANSYS [27]. The target is represented by a mesh of finite elements that changes over time so as to simulate materials removal. This time-dependent problem was solved sequentially, as a series of 1 ns time steps, linked together by using the output of step n as the initial conditions for problem $n + 1$. The target initial temperature is 298 K. If the temperature of an element is higher than the melting temperature (T_m) at the end of a particular step, melting is assumed to have occurred and the latent heat of melting (L_m) is taken into account in the calculation. Similarly, ablation is assumed to occur when the temperature of the surface elements is higher than the boiling temperature. If this happens an ablation depth h is calculated from Eq. (4) and compared with the element thickness, Δy. If $h > \Delta y$ the element is supposed to be vaporized and the surface temperature of the remaining material corrected to take into account the latent heat of vaporization. The geometry used for the simulations is illustrated in Fig. 1. To minimize the computer processing time the target is supposed rectangular with 10 μm × 3 μm 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 (Fig. 1). The size of the elements is 2 nm × 10 nm in the upper half of the substrate and 10 nm × 10 nm in its lower half. The use of a thinner mesh in the upper part of the

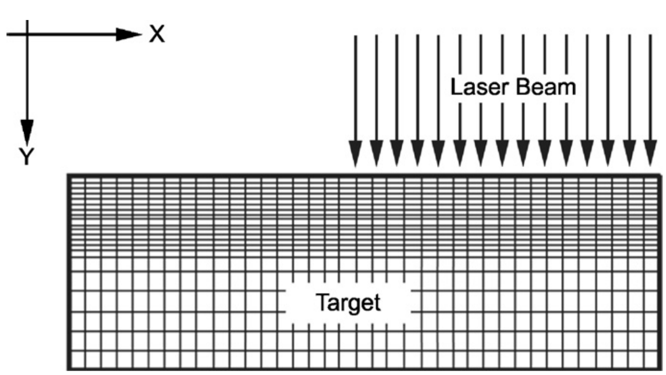

Fig. 1. Geometry for laser ablation of TiC.

Table 1
TiC parameters [1,28,29]

T_m (K)	T_b (K)	ρ (kg/m^3)	L_m (J/kg)	L_V (J/kg)	β
3340	5080	4910	10^6	10^7	0.18

Table 2
Temperature-dependent parameters [1]

T (K)	300	400	800	1200	1600	2000	2400	>2400
K [W/(m K)]	23	24	31	36	40	43	45	45
C_p [J/(kg K)]	550	650	830	890	900	915	930	930