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Thermal model for nanosecond laser ablation of alumina

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Abstract

A thermal model based on heat-conduction equation and Hertz–Knudsen equation for vaporization has been employed to simulate nanosecond pulsed laser based ablation of alumina. Heat transfer in the laser irradiated target with allowance for phase transitions, provides estimates for temperature distribution within the target and material ejection rate via ablation. Good agreement between calculated and experimentally measured data on mass ablation rate per pulse and its dependence on incident laser fluence from 5 to 22 J/cm^2, validated our theoretical model. Observed deviation between calculated and experimentally measured ablation rates at high average laser fluence levels was explained by ablation induced progressive degradation of target surface. Absence of abrupt increase in ablation rate with increased laser fluence suggested material ejection largely via normal boiling rather than explosive boiling mechanism. Calculated maximum surface temperature of the target was found to lie well below empirically estimated thermodynamic critical temperature for alumina, corroborating our observations on absence of onset of explosive boiling in alumina target on laser irradiation. Our simulation study enables proper selection of laser fluence, successfully minimizing laser induced target damage, as well as, degradation of micro-structural and mechanical properties of alumina films deposited via pulsed laser ablation.

Keywords: B. Surfaces; D. Al_2O_3; E. Refractories

1. Introduction

Pulsed laser ablation (PLA) forms the basis for an extensive range of applications such as, Pulsed Laser Deposition (PLD) for thin films and coatings [1], surface engineering of solids for device applications [2], vapor generation for analytical characterization of materials [3], and material processing and nanostructuring [4,5]. The underlying mechanism of material removal on laser irradiation depends on the type of material being processed, processing laser parameters, as well as, the prevailing ambient conditions. Laser pulse duration critically determines the physical process of coupling and dissipation of laser energy into the target material [6]. Material ablation with nanosecond pulsed laser is largely thermal in nature. Laser energy absorbed in the medium gets transferred to the lattice through electron–phonon coupling leading to heating and phase transitions at characteristic temperatures, culminating in evaporation of the target material. The complex mechanism of laser ablation could lead to material removal, either in the form of fine vapor, liquid droplets, solid flakes or uneven and large fragments. Reliable prediction of laser ablation rates and associated features has often been facilitated through theoretical simulation of pulsed laser ablation that closely describes the complex mass removal mechanisms [7–9].

Alumina or Aluminum oxide (Al_2O_3) is a refractory material widely used as, optical thin films and protective coatings [10], as a bio-ceramic material on account of its superior bio-compatibility [11], as a tribological coating [12], and in thermal barrier coatings [13]. It is an inert ceramic having a high degree of chemical compatibility with most reactive reagents, excellent wear resistance and hardness and can also be polished to a high degree of surface finish. High hardness and wear resistance endow alumina with excellent tribological qualities making it one of the most commonly used materials for hard coatings [14]. For high temperature applications the hard and thermally stable α-phase of alumina is preferred [15]. Alumina coatings deposited through Chemical Vapor Deposition [16], Metal organic chemical vapor

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deposition [17], Pulsed Laser Deposition [18], Magnetron sputtering [19], and Plasma spray deposition [20] techniques, have all been reported.

Of the techniques employed for deposition of alumina thin films the pulsed laser based process has been particularly promising. PLD has been successfully demonstrated for deposition of multi-component, as well as, single component films of ceramics, semiconductors, metals, dielectrics and insulators. PLD has been used to generate a wide range of coating morphologies ranging from amorphous to crystalline, smooth, dense and optically homogeneous to rough and porous [1]. PLD technique not only allows decoupling of vacuum hardware and the energy source responsible for material evaporation, but also provides desired control of properties of deposited films through appropriate choice of laser parameters. That heating of the substrate is not always necessary with PLD technique is an attractive associated advantage since heating has often been observed to result in substrate degradation.

Most ceramics are poor absorbers of heat and much less reflective than metals. Hence laser beams tend to penetrate effectively and deposit energy to larger depths within ceramics than metals. Amount of laser energy deposited into the target critically determines ablation parameters including ablation yield. Laser ablation based optimized processing of ceramics involves critical choice of processing laser parameters and aims at successfully avoiding generation of cracks due to thermally induced stresses within the target, explosive boiling and ejection of fragments and large droplets from the target. All these phenomena are not only detrimental for controlled and high precision micromachining of ceramic targets, but also compromise the quality of ceramic thin films and coatings deposited using a PLD approach. An understanding of the mechanism and dynamics of material-removal process following a theoretical model based approach serves not only to accurately estimate material ablation rates but also facilitates selection of laser parameters for optimization of the application technique.

At high laser fluence levels, a complex coupled process involving vaporization, ionization, gas dynamics including vapor and plasma expansion with possible shock wave generation, have been proposed [21,22]. However, for laser fluence levels maintained close to ablation threshold, simpler simulation models describing laser ablation beginning with laser induced heating and evaporation of target in the presence of heat transfer within the target, and gas dynamics to describe the generated vapor, have been reported [3].

In this paper, we have employed a thermal model for nanosecond pulsed laser ablation based on a heat conduction approach within the target and phase transition resulting in normal boiling and vaporization. Although, plasma formation and laser plasma interaction have not been considered explicitly in our model, screening effect of the plasma has been broadly incorporated through scattering and absorption losses experienced by the process laser beam as it propagates through the ablation plume. Heat transfer within the target with allowance for phase transitions provides temperature distribution generated in the target when irradiated with a nanosecond laser pulse. Velocity of the ablating vapor and ablation rate have been estimated following a Hertz–Knudsen approach avoiding description of vapor plume involving gas dynamic equations [9]. This simulation study of laser ablation of alumina enables processing conditions to be related to target ablation rate. In addition, our theoretical simulation facilitates proper selection of laser fluence thereby successfully avoiding onset of explosive boiling in laser irradiated alumina target hence minimizing laser induced target damage, as well as, degradation of micro-structural and mechanical properties of deposited PLD alumina films.

2. Numerical simulation

2.1. Model

Laser energy is coupled into the target material via electronic excitation. Equilibrium between electrons and lattice vibrations occurs via electron–phonon interaction, typical relaxation times ranging up to 100 ps [23]. Hence, when simulating laser matter interactions in nanosecond time domain one can safely assume laser energy to have been coupled as thermal energy into the target. On irradiation with a laser pulse energy is partly reflected at the target surface and the rest is absorbed within a short penetration depth characterized by the absorption coefficient of the target material. The absorbed energy is distributed within the target largely via conduction. If the laser energy coupled into the target is sufficient for melting and vaporization, material ablation occurs.

Physical processes that occur during laser ablation include energy deposition into the material followed by heat transfer within the target, radiative and convective boundary conditions, thermo-dynamics of phase-changes, a moving melt and solid interface, fluid flow and evaporation via normal boiling, as well as, explosive boiling should the target temperature approach the critical temperature of the target material. Hence, an exact theoretical model becomes extremely cumbersome and complex. However, based on the specific nature of dynamics being explored several associated processes and mechanisms often become small enough to be safely neglected. This allows considerable simplification of the numerical approach.

A simplified thermal model of laser induced heating and material removal validated against experimental data on nanosecond laser heating and vaporization of ceramics [24] has been utilized to describe laser ablation of alumina. Broadly the following assumptions have been made:

1. Heat radiated from laser irradiated target surface estimated by σT^4 (where T is the temperature of the hot target surface and σ is the Stefan–Boltzmann constant) has been observed to be $\sim 2\times10^3$ W/cm^2 for maximum surface temperatures as high as 4300 K typically reached in case of PLA of alumina. This is negligible in comparison to typical average laser intensity of 10^{11} W/cm^2 deposited by incident laser beam. Hence, radiative heat loss from the target has been neglected.
2. For transient laser irradiation involving a single laser pulse and in absence of cumulative effects arising from successive laser pulses, heat transport via convection has been neglected [25].