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Modelling of pulsed electron beam induced graphite ablation: Sublimation versus melting

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Abstract. Pulsed electron beam ablation (PEBA) has recently emerged as a very promising technique for the deposition of thin films with superior properties. Interaction of the pulsed electron beam with the target material is a complex process, which consists of heating, phase transition, and erosion of a small portion from the target surface. Ablation can be significantly affected by the nature of thermal phenomena taking place at the target surface, with subsequent bearing on the properties, stoichiometry and structure of deposited thin films. A two stage, one-dimensional heat conduction model is presented to describe two different thermal phenomena accounting for interaction of a graphite target with a polyenergetic electron beam. In the first instance, the thermal phenomena are comprised of heating, melting and vaporization of the target surface, while in the second instance the thermal phenomena are described in terms of heating and sublimation of the graphite surface. In this work, the electron beam delivers intense electron pulses of ~100 ns with energies up to 16 keV and an electric current of ~400 A to a graphite target. The temperature distribution, surface recession velocity, ablated mass per unit area, and ablation depth for the graphite target are numerically simulated by the finite element method for each case. Based on calculation findings and available experimental data, ablation appears to occur mainly in the regime of melting and vaporization from the surface.

1. Introduction
Over the past few years, short powerful electron pulses have gained significant attention owing to technologically important applications in various fields ranging from thin film deposition to electron beam lithography [1,2]. Recently, pulsed electron beam ablation (PEBA) has arisen as a relatively novel and potential alternative, due to many superior technical capabilities, to the well-established pulsed laser ablation (PLA) method. In PEBA, electron pulses, instead of pulses of photons, of high current and voltage are employed to ablate the target. Energetic electron pulses during PEBA are generated by a beam source known as channel-spark discharge [2-4]. The resulting electron beam is characterized by short pulse duration (~100 ns) and high power density (10^8 W/cm^2). The beam is directed at a target surface by low-energy electrons (~10-20 keV), penetrating into a target surface. This causes a quick vaporization, as a result of non-equilibrium heating, of a small portion of the target surface and ensuing plasma plume expansion toward a substrate. Accordingly, thin films with superior properties can be deposited at a considerably lower substrate heating, compared to other deposition techniques such as single stage co-evaporation or sputtering [5]. PEBA offers several attractive advantages over PLA, but one of the most distinguishing capabilities is its ability to ablate a wide range of materials, including...
optically transparent or reflective materials, owing to the electrons shorter absorption depth as compared to photons [4-6].

It is has been demonstrated experimentally that pulsed electron beam ablation can be an efficient technique for the synthesis of thin films, nanostructures and clusters [7-9]. Despite PEBA growing experimental developments, the mechanism of surface ablation during beam-target interaction is still not well understood. It has been well established that ablation induced during nanosecond PLA can typically take place through three types of thermal processes: (i) explosive boiling (phase explosion), (ii) normal boiling and (iii) normal vaporization [10]. Depending on pulse duration and laser intensity, any kind of thermal mechanism can take place during laser ablation. When laser irradiance is in the range of $10^9$ W/cm², normal vaporization is one of the major dominating thermal processes in PLA [11]. Electron beam ablation of a target surface is a complex process that comprises absorption of pulsed electron beam energy, heating, phase transition and ablation of the target surface. Owing to the short spatial and temporal scales involved in beam-target interaction, ablation is not always easily amenable to experimental studies as in situ characterizations of the ablation process can be quite challenging. Accordingly, due to the inherent limitations and complications involved in the ablation process, mathematical modeling of the phenomena involved during ablation can prove to be an efficient tool to gain a better understanding of electron-beam induced ablation. In the present work, a brief discussion is given of the thermal model used to describe ablation during electron beam interaction with the graphite target. The cases of melting and sublimation of a graphite target are considered and the results are discussed in terms of temperature distribution, surface recession velocity, ablated mass per unit, and ablation depth.

2. Mathematical model

The theoretical description of electron beam interaction with a graphite target is based on the models described in detail elsewhere [12,13]. Target heating upon interaction with a nanosecond pulsed electron beam can be defined in the solid phase by the heat conduction equation in a one-dimension as,

$$\rho_s C_p \frac{\partial T}{\partial t} = \nabla \cdot (k_s \nabla T) + Q(x,t) \quad (0 < t \leq t_{SM})$$  \hspace{1cm} (1)

where $\rho_s$, $C_p$, and $k_s$ are the solid phase density, specific heat capacity, and thermal conductivity of the target material, respectively. The term $Q$ is the heat source, $x$ is the distance from the target surface along the incident direction, and $t_{SM}$ is the time required by the target surface to attain the sublimation or melting point.

In the case of fluid (vapor/liquid) phase, thermodynamics of the surface target can be defined by introducing the recession velocity in the one-dimensional heat conduction equation [14],

$$\rho_f C_p \left( \frac{\partial T}{\partial t} - u_s \frac{\partial T}{\partial x} \right) = \nabla \cdot (k_f \nabla T) + Q(x,t) \quad (t_{SM} < t \leq \tau)$$ \hspace{1cm} (2)

where $\rho_f$, $C_p$, and $k_f$ are the target material fluid phase density, specific heat capacity, and thermal conductivity, respectively. The terms $u_s$ and $\tau$ are the surface recession velocity and duration of the electron beam pulse, respectively.

When vaporization becomes substantial at higher surface temperature, velocity due to surface recession during this stage can be calculated by Hertz-Knudsen equation [14],

$$u_s = (0.82) \frac{P_s}{\rho} \sqrt{\frac{m}{2\pi k_B T_s}}$$ \hspace{1cm} (3)

where $P_s$ is the saturated vapor pressure of the ablated particles, $k_B$ is Boltzmann constant, $T_s$ is the surface temperature, and $m$ is the average mass of target particle. The saturated vapor pressure at the target surface can be determined according to the Clausius-Clapeyron equation [14],
\[ P_S = P_0 \exp \left[ \frac{mL_v}{k_B} \left( \frac{1}{T_{SB}} - \frac{1}{T_S} \right) \right] \] (4)

where \( P_0 \) and \( T_{SB} \) are the normal atmospheric pressure and sublimation/boiling temperature under \( P_0 \), respectively. The heat source, \( Q \), term used in this study has been described somewhere else thoroughly in detail [12,13].

3. Results and discussion

Due to the rapid absorption of electron beam irradiation onto the target surface, the target temperature, which is initially at ambient condition (298 K), rises very quickly. The temperature profiles of the target surface calculated at an accelerating voltage of 16 kV and beam efficiency of 0.6 for the cases of melting and sublimation are illustrated for the entire pulse width in figure 1. For both cases, it can be observed that the surface temperature rises slightly quickly initially, reaches a maximum value of ~7700 K, then falls off towards the end of the pulse. The decline in surface temperature is significantly slower than the initial temperature rise. As it can be seen in figure 1, the surface temperature is still higher than the boiling/sublimation temperature at the end of the pulse. This can be most likely explained in light of the polyenergetic character of the electron beam [15,16]. Beam electrons appearing at the end of the pulse convey a power density of \( \sim 10^6 \) W/cm\(^2\) owing to high beam current and short penetration depths in the target, which, in turn, sustains the ablation process. Furthermore, it can be observed from figure 1 that the maximum temperature attained by the surface is well below \( 0.9T_c \) (where \( T_c \) is the critical thermodynamic temperature) for graphite. Accordingly, calculated surface temperature profiles suggest no indication of phase explosion (explosive boiling) during PEBA. Normal vaporization is the most likely thermal process involved in the ablation of the target surface. Around the two cusps exhibited by the temperature profile, as shown in figure 1, the difference in surface temperature corresponding to melting and sublimation reaches a maximum value of \( \sim 110 \) K.

![Figure 1. Target surface temperature corresponding to melting and sublimation (indicated in the inset) as a function pulse duration.](image)

Surface recession velocity calculated for the different thermal phenomena for the pulse width of 100 ns is depicted in figure 2. As can be observed, for sublimation the calculated recession velocity from the target surface increases rapidly and reaches a maximum value of 95 m/s at about 66 ns, i.e., when the target surface temperature is at maximum (7700 K). For the case of melting, the recession velocity increases very gradually over the period of the pulse duration and reaches up to 23 m/s at about 66 ns.
Upon reaching the maximum value, and for both cases, it can be observed that the surface recession velocity decreases abruptly until the end of the pulse.

![Figure 2. Surface recession velocity profiles corresponding to melting and sublimation as a function pulse duration.](image)

Surface recession velocity can be integrated over the pulse duration to calculate ablated depth and ablation mass per unit area for both thermal cases. These are reported for the entire pulsed width in figures 3 and 4. As a result of the maximum surface recession velocity reaching maximum value for the case of sublimation, it can be expected that the magnitudes of ablation depth and ablated mass per unit area in this case, i.e., sublimation, will be higher than their counterparts in the case of melting, as can be appreciated in figures 3 and 4.

![Figure 3. Ablation depth profile corresponding to melting and sublimation as a function pulse duration. The experimental ablation depth is reported for comparison.](image)
4. Model assessment
To assess the validity of the model, appropriate parameters calculated from the model have been contrasted with available experimental results, under the prevailing conditions of this study, in the literature [5,17-19]. The reported experimental value of the ablation depth has been estimated to be around 1.0 µm [5,17]. This is in good accordance with the ablation depth of 0.75 µm obtained from the model based on melting as illustrated in figure 3. Experimental observations have indicated that the ablated mass per unit area via PEBA is about 5-10 times greater than its counterpart via PLA for a similar energy fluence [5,18]. The reported experimental value of ablated mass per unit area via PLA has been estimated to be around 0.30 µg/mm² for a graphite target [18]. The calculated ablated mass per unit area from the model accounting for melting is about five times greater than the reported experimental value, as illustrated in figure 4. According to the results obtained from the present model calculations and from appropriate previous experimental studies [17,19], target ablation seems to take place primarily in the regime of melting and vaporization from the surface.

5. Conclusions
A two stage, one-dimensional heat conduction model is presented to investigate thermal phenomena responsible for ablation in PEBA. A finite element analysis is applied to model equations to estimate the temporal dependence of the surface temperature for a graphite target. Surface recession velocity, ablation depth and ablated mass per unit area are also calculated. Calculation results are compared with available experimental results to assess the validity of the model. It is concluded that the target ablation via PEBA takes place via melting instead of sublimation.

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