Accelerating voltage effect on the ablation process during pulsed electron deposition

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Abstract. In this study, we have analyzed the effect of electron beam accelerating voltage on the maximum temperature of a graphite target during pulsed electron beam ablation (PEBA). To this end, a two stage, one dimensional thermal model is used. The target is subjected to an electron pulse with accelerating voltages of 10, 13, 15, 17 and 18 kV. The surface temperature, vaporization front velocity, ablated depth, and ablation rate are estimated from the solution of the model. Simulation results have shown that target surface temperature is not proportional to the accelerating voltage. It has been found that the surface temperature increases with the accelerating voltage from 10 kV to 15 kV, and reaches a maximum value (7500 K) at 15 kV. After 15 kV, the temperature decreases with increasing accelerating voltage. Similar trends have been observed in the vaporization front velocity, ablated depth and ablation rate, with maximum values (75 m/s, 2.1 µm, and 4 µg/mm²) at 15 kV. The calculation results are in good agreement with relevant experimental data from the literature.

1. Introduction
Over the past few decades, the interaction of intense pulsed energy beams with solid-state materials has gained growing interest for the fabrication of high quality thin films. Pulsed electron beam ablation has recently emerged as a potential alternative to well-known pulsed laser deposition (PLD) for the fabrication of thin films with superior qualities. PEBA is conceptually similar in overall physics to PLD, with distinctive advantages of efficient energy transfer (electric-electric: ~30 %), lower operating costs, and ease of use [1-3]. Thin film deposition during PEBA is significantly influenced by many factors such as power density, beam energy, background gas pressure, and target to substrate distance. Target ablation is mainly affected by two critical factors, which are beam energy and power density delivered to the target surface. Experimental results have shown that it is not always favorable to operate the electron beam at higher voltage in order to increase the target heating rate. The fundamental principle of PEBA involves the generation of a highly energetic pulsed electron beam with a short pulse width (~100 ns) and high power density (10⁸ W/cm²). The energetic electron beam impinges on the target surface, which leads to rapid sublimation of a fine fraction of the surface and to the ejection and expansion of a plasma plume. The latter will eventually condense as a thin film on a properly placed substrate in a vertical direction to the target. Due to high power density delivered to the target surface (10⁸ W/cm²), fast heating and vaporization take place far from thermodynamic equilibrium. Solid-to-vapor transition is principally independent of the phase diagram and composition of the target material, resulting in congruent sublimation and potentially, target stoichiometry preservation in the deposited thin film [1,4].

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Despite the fact that PEBA has been used in the fabrication of thin films for two decades now, very limited attempts have been made to model and optimize the critical parameters involved in the ablation process [5,6]. In an early attempt, Kowalewicz and Redel [5] have proposed a model, which describes irradiation by a polyenergetic electron beam, energy dissipation via conduction and the effect of the vapor pressure of the ablated mass on the temperature of the target. Recently, Tricot et al. [6] have suggested a simple model of heat conduction in a target irradiated by a polyenergetic pulsed electron beam, but does not include phase change during ablation. Both of these models do not account for the receding surface of the target during ablation. In order to thoroughly assess PEBA for the deposition of thin films with superior properties, it is essential to interpret how the critical parameters, particularly the accelerating voltage behaves during the ablation process. As mentioned earlier, beam accelerating voltage is one of the key parameters towards thin film fabrication control. In this study, we present the effect of accelerating voltage on the ablation efficiency of graphite.

2. Mathematical model

The interaction of an electron beam with a target material is quite a complex physical process and takes place in a sequence of phenomena. When the target surface is exposed to electron beam radiation, the incident beam energy is mostly absorbed by the electron subsystem. Later, due to interactions between electrons and phonons, energy is transferred to phonons in the lattice. The energy transferred to the phonons propagates in the target material through the mechanism of heat conduction [7]. The thermal model for electron beam-target interaction in the regime of sublimation can be categorized to occur in two stages: (i) heating of the target surface, and (ii) sublimation of the target surface. Therefore, the temperature distribution inside the target surface along the depth can be described by a two stage one-dimensional heat conduction model. The latter depends on the incident electron beam parameters and the thermophysical properties of the target material. The diameter of the electron beam is normally in the range of several millimetres, which is much greater than the diffusion depth. Therefore, heat conduction can be assumed to take place predominantly in one dimension along the target depth [8].

The heat conduction equation, which allows the calculation of time-dependent temperature distribution along the target depth, $T(x, t)$, before the target surface has started to vaporize is expressed as,

$$\rho_s C_p s \frac{\partial T}{\partial t} = \nabla \cdot (k_s \nabla T) + S(x, t) \quad (0 < t \leq t_s)$$

with the initial condition,

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

and boundary conditions,

$$-k_s \nabla T \bigg|_{x = 0} = Q_s(t) \quad (0 < t \leq t_s)$$

$$-k_s \nabla T \bigg|_{x = l} = 0 \quad (0 < t \leq t_s)$$

where $\rho_s$, $C_p s$, and $k_s$ are the solid phase density, solid state specific heat capacity, and solid phase thermal conductivity of the target material, respectively. $S$ is the heat source term, $T_0$ is the initial temperature of the target surface, $x$ is the distance from the target surface along the incident direction, $l$ is the thickness of the target material, $Q_s$ is the surface power density delivered by the heat source, and $t_s$ is the time required by the target surface to attain the sublimation point.

When the target surface temperature increases and exceeds the sublimation temperature, vaporization will start to take place. During this stage. During the vaporization stage, the thermal field in the target can be calculated by introducing the moving vaporization front in the one-dimensional heat conduction equation [9]. The latter takes the form,
\[ \rho_v C_{pv} \left( \frac{\partial T}{\partial t} - u(t) \frac{\partial T}{\partial x} \right) = \nabla \cdot (k_v \nabla T) + S(x, t) \quad (t_s \leq t \leq \tau) \]  

(5)

Here the initial and boundary conditions are

\[ T(0, t) = T_s(t) \]  

(6)

\[ -k_v \nabla T \bigg|_{x=0} = Q_s(t) - L_v \rho_v u(t) \quad (t_s \leq t \leq \tau) \]  

(7)

\[ -k_v \nabla T \bigg|_{x=l} = 0 \quad (t_s \leq t \leq \tau) \]  

(8)

where \( \rho_v, C_{pv}, \) and \( k_v \) are the vapor phase density, vapor phase specific heat capacity, and vapor phase thermal conductivity of the target material, respectively. \( u(t), L_v, \) and \( \tau \) are the vaporization front velocity, latent heat of vaporization, and duration of the electron beam pulse, respectively. The time required by the target surface to attain the sublimation point can be estimated by [10],

\[ t_s = \frac{3(T_{sb} - T_0)^2 \rho_s C_{ps} k_s}{4Q_s(t)^2} \]  

(9)

During the process of sublimation, the vaporization front velocity can be evaluated assuming that the vaporization flux from the target surface follows Hertz-Knudsen equation [9], and it can be given by,

\[ u(t) = 0.82 \frac{P_s(T_s)}{\rho} \left( \frac{m}{2\pi k_B T_s} \right)^{\frac{1}{2}} \]  

(10)

where \( P_s(T_s) \) is the saturated vapor pressure at the surface temperature \( T_s \), \( m \) is the average mass of target particle, and \( k_B \) is the Boltzmann constant. The solid to the vapour phase transition has been assumed to follow the Clausius-Clapeyron equation [9],

\[ P_s(T_s) = P_b \exp \left[ \frac{m L_v}{k_B} \left( \frac{1}{T_{sb}} - \frac{1}{T_s} \right) \right] \]  

(11)

where \( P_b, L_v, \) and \( T_{sb} \) are the atmospheric pressure, latent heat of vaporization, and sublimation temperature during the atmospheric pressure \( P_b \).

The source of electrons in PEBA is polyenergetic [5]. In order to describe the heat source appropriately, the distribution of energy must be taken into consideration. Monte Carlo simulations are able to estimate the profile of the energy transferred to the target material by the electron beam. Monte Carlo is based on tracking a primary electron of initial energy \( E \) and direction \( \mathbf{v} \), and finding where the next collision will occur based on the calculation of the inelastic mean free path. To achieve statistical sampling, this process is repeated for thousands of electrons using CASINO code [11]. The analytical expression of maximum energy deposited by each group of electrons takes a polynomial form as

\[ E(x) = f_{A,B,C}(x) = \sum_{i=0}^{q} a_i \times x^i \]  

(12)

where \( x \) is the penetration depth along the target material. In order to account for energy losses, the fraction of beam electrons that are effectively used during ablation has been set to 0.6. This parameter has been assessed in a separate study [12].
3. Results and discussion

In our calculations, the target is initially at ambient room temperature (300 K). Due to the electron beam energy deposition in the target material, the surface is heated very rapidly. The calculated target surface temperature for various values of the accelerating voltage is depicted for the entire pulse width (~100 ns) in Fig. 1. As anticipated, the maximum temperature is located at the target surface. It can be seen that the surface temperature and heating rate increase rapidly for the accelerating voltage in the range of 10 to 15 kV. This finding is likely due to the polyenergetic nature of the electron beam [5]. The majority of electrons produced by the beam for the voltage in the range of 10 to 15 kV have lower energies. As a result, the maximum energy of the electrons is deposited near the target surface. The highest temperature is observed for an accelerating voltage of 15 kV at about 7500 K between 60 and 70 ns. At higher voltages, the heating rate falls due to an increase in the penetration depth of the electrons, which offsets the intensity of the beam. As the acceleration voltage increases the energy of the electrons in the beam increases while, at the same time, the highly energetic electrons will penetrate deeper in the target. This results in greater losses of the beam energy, which becomes predominate at higher voltages (above 15 kV), and eventually cancels out the effect of the acceleration voltage on the beam energy. For 17 kV, it can be seen that initially the heating rate is about the same as 15 kV, but it slightly decreases after 40 ns yielding a maximum temperature of 7300 K. The evolution of the target surface temperature with time and dependence on voltage seem to have a similar trend for all voltages except for 18 kV. At 18 kV, the temperature profile goes through a short plateau at ~20 ns corresponding to a target surface temperature of 4000 K. This is the result of phase change due to sublimation, which starts to occur around 4000 K. Afterwards, there is a sudden climb in the temperature whereby the surface temperature reaches 6700 K. For lower voltages than 18 kV, sublimation seems to occur much faster so that not plateauing of the temperature can be observed.

The vaporization front velocity from the target surface is illustrated in Fig. 2. As can be seen, at 15 kV the velocity reaches a maximum value of 75 m/s at about 65 ns, i.e., when the surface temperature of the target is at maximum (7500 K). Beyond the latter, the surface front receding velocity decreases as the target surface temperature drops. A similar trend can be observed for the other values of the accelerating voltage.
The ablated depth and ablation rate can be obtained from the surface front receding velocity. These are plotted as a function of pulse duration in Fig. 3 and 4, respectively. As expected for the case of 15 kV, the calculated ablated depth and ablation rate are larger than at other voltages. The calculated ablated depth is around 2.1 µm and the ablation rate is about 4 µg/mm². Similar profiles of both ablated depth and ablation rate for the remaining accelerating voltage values can be observed.

Figure 3. Ablated depth for various accelerating voltages as a function of pulse duration.

4. Model validation
Experimental data on the solid-state target ablation by pulsed electron beams are very scarce and more so on graphite. To validate the model, the estimated parameters for different values of the accelerating voltage are compared with relevant experimental data in the literature [4,5,13-16]. For all voltage values, the ablated rate ranges between 9 and 50 µg/pulse or 50×10¹⁶ and 250×10¹⁶ atoms/pulse, which is in good agreement with reported experimental data of a few tens of µg/pulse [15] or some 10ⁱ⁰ atoms/pulse.
Experimental results have shown that the ablation rate in PEBA is at least 10 times larger than its counterpart in PLD. This is true in our case where the ablation rate in PEBA is 10-20 times the rate in PLD for a graphite target [14,15]. In terms of the heating rate, our results are in qualitative (no absolute values have been provided by the authors) agreement with experimental data [4,13]. The latter indicate that the heating rate is at maximum at a voltage of about 15 kV. Finally, electron beams have been reported to reach a penetration depth in the target of 1-2 µm [15,16]. This is in good accordance with our calculated ablated depth as reported in Fig. 3.

Figure 4. Ablation rate for various accelerating voltages as a function of pulse duration.

5. Conclusion
We have proposed a two stage, one-dimensional heat conduction model to investigate graphite ablation during PEBA at various electron beam accelerating voltages. The temporal dependence of the temperature of the target has been numerically estimated. Vaporization front velocity, ablated depth and ablation rate have also been calculated. The findings have shown that an optimum value of the accelerating voltage is around 15 kV. The latter corresponds to the maximum value of the surface front receding velocity, ablated depth and ablation rate. The calculation results have been compared with available experimental data and have been found to be in overall good agreement.

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