Spectrochimica Acta Part B 122 (2016) 1–8

Contents lists available at ScienceDirect

Spectrochimica Acta Part B

journal homepage: www.elsevier.com/locate/sab

Evaluation of explosive sublimation as the mechanism of nanosecond laser ablation of tungsten under vacuum conditions

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A R T I C L E   I N F O

Article history:
Received 28 February 2015
Received in revised form 12 April 2016
Accepted 22 May 2016
Available online 24 May 2016

Keywords:
Laser-induced breakdown spectroscopy
LIBS
Laser ablation modeling
Laser ablation mechanism
ITER
Tungsten

A B S T R A C T

A non-equilibrium mechanism for nanosecond laser ablation is suggested herein, and its predictions are compared to the results of W experiments performed under vacuum conditions. A mechanism of particle formation is explained via this model, with partial sublimation of the superheated irradiated zone of the target considered to be the mechanism of laser ablation. In this study, a mixture of vapor and particles was explosively generated and subsequently prevented the rest of a laser pulse from reaching its intended target. This mechanism was found to play an essential role in the ablation of W under vacuum conditions, and it provides a theoretical justification for particle formation. Moreover, special considerations were taken into account for the expansion of plasma into a vacuum. The model was evaluated by measuring the mass of ablated particles using a quartz crystal deposition monitor and time-resolved optical emission spectroscopy. The results of this model were found to be in good agreement with experimental values.

1. Introduction

Laser ablation and resulting laser-produced plasma have been found to have numerous applications in a variety of scientific and technological fields, ranging from materials, chemical analytics, and biological sciences to the exploration of life on other planets [1–4]. Among these applications are laser-induced ablation spectroscopy (LIAS), laser-induced desorption spectroscopy (LIDS), and laser-induced breakdown spectroscopy (LIBS), in which laser-based methods are used in combination with spectroscopy for analytical purposes. Recently, these three methods have been considered for use as in-situ diagnostic tools for plasma-facing materials within the International Thermonuclear Experimental Reactor (ITER), which is presently under construction [5–11].

Due to the occurrence of several interaction processes between the edge plasmas and inner walls of fusion device chambers and divertors, the wall materials are subjected to harmful mechanisms, such as erosion, re-deposition and fuel retention. These can affect overall system performance and can also raise safety issues, thus necessitating strict control of steady-state and transient wall power loads to within technically acceptable limits, as well as the control of in-vessel tritium inventories. Hence, the ejected wall materials should be properly monitored via the methods consisting with the harsh conditions expected in ITER as well as in other fusion tokamaks. Although the suggested laser-based methods of LIAS, LIDS, and LIBS seem to be capable for this purpose, Huber et al. [12] advocated that additional research on the basic processes that support the special conditions inherent to fusion devices is required before applying these methods in ITER.

Among these laser-based diagnostic methods that may be used in ITER, LIBS can be performed between discharges, in which the complicated interactions between the edge plasma, laser-produced plasma, and magnetic field effects do not need to be considered. To avoid these complexities, our research team focused its study on LIBS, in which only the vacuum condition makes it compatible with the special conditions found in fusion devices.

Due to its hardness and high melting point, W is a suitable element for the ITER divertor upper baffle and dome and thus could be used in either of two ways in high-power divertor areas [12–16]. Therefore, knowledge of ablation due to laser irradiation, plasma plume generation and expansion, and light emission is crucial to the design and optimal setup of LIBS for its in-situ diagnostics. Moreover, an analogy has recently been found in regimes of W erosion within ITER-like reactors and erosion due to laser ablation [17]. This investigation was thus motivated by the quest of investigative findings related to ITER plasma–wall interactions associated with environments characterized by W laser-plasma generation and expansion (in a vacuum) at different laser–pulse irradiation levels.

There have been several model-based studies on laser ablation (and its related processes) over the years for a number of candidate metallic targets. While many problems such as heat conduction through the target, as well as plasma formation, expansion, shielding, and emission must be considered, one of the most crucial challenges is the actual

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http://dx.doi.org/10.1016/j.sab.2016.05.008

phenomenon of ablation itself. Different mechanisms associated with surface evaporation and phase explosion (also known as explosive boiling or volumetric boiling) have been considered for laser ablation. In 1968, Anisimov [18] developed a model for evaporation based on treating the Knudsen layer region as a "gas dynamic discontinuity" across which certain jump conditions applied; as a result, a full expression of conservation of mass, momentum, and energy principles followed. Anisimov employed this model only for strong vaporization limit scenarios, wherein the flow just outside the Knudsen layer was sonic. Over subsequent years, jump conditions for subsonic expansions and the Knudsen layer with recondensation boundary conditions were developed by many researchers [19–24], and particularly through the more recent method proposed by Gusarov and Smurov [25]. Several other researchers have also used the evaporation mechanism and the jump conditions of the Knudsen layer to model the nanosecond laser ablation process (for examples, see Refs. [26–32]). At higher laser fluences, phase explosion (or explosive boiling) has regularly been considered the primary mechanism behind the ablation process. This mechanism, proposed by Martynyuk [33], has likewise also been investigated by numerous researchers. Experimentally, a sudden increase in crater depth or dramatic changes in other physical properties have been attributed to phase explosion (for example, see Refs. [34–37]). Experiments with different materials have yielded different delay times for phase explosion [35,36,38], many of which have lasted up to the ends of the given laser pulse periods and some of which have transcended down into the microsecond level. The delay time has conventionally been attributed to the time lag necessary for the formation of homogeneous nuclei [19]. Recently, a multiphase collisional radiative model was developed by Autrique et al. in which the entire target and plume system was described by a one-dimensional hydrodynamic model that adequately accounts for energy conservation, mass conservation, and pressure relaxation [20,39]. A review on radiative models was provided by Gornushkin and Panne [40].

For the ablation of W in vacuum, however, equilibrium explosive boiling and normal evaporation are minimally present when superheating occurs. As the surrounding pressure is very low, atoms that are released from lattices expand freely. Moreover, the formation of large particles, ejected outward from a target surface at constant velocities on the order of a few meters per second, can properly be explained using a partially superheated sublimation mechanism. Note that due to equilibrium phase transitions such as homogeneous boiling, the ejected liquid (in its entirety) is expected to be converted into vapor. Although some small clusters are likely to be formed by equilibrium boiling, the generation of stable particles on the order of a micrometer in size (as what one can typically expect from nanosecond laser ablation of W in a vacuum), is not probable. Similar to heterogeneous and homogeneous boiling, normal evaporation thus cannot be considered as an ablation mechanism. Moreover, the commonly used Clausius–Clapeyron relation for the calculation of saturated pressure on target surfaces leads to a value that is almost four times greater than the critical pressure at the critical temperature of W. Therefore, even at low laser fluences, the formulations for evaporation and the Knudsen layer developed in Refs. [18–24] are not adequate for W. Hence, in this paper, the partial sublimation of the superheated zone is proposed as the mechanism of the nanosecond laser ablation of W under vacuum conditions.

For the modeling of ablation in vacuum conditions, the most consequential issues relate to the boundary conditions of the plume. At the vacuum edge, the media is not in a continuum-mode; thus, associated hydrodynamic equations cannot be established. Any temptation to use a very low concentration ambient gas for the vacuum boundary and to solve the subject equations using the Godunov scheme or the Riemann solvers leads to a strong fictitious shockwave, which in reality can never occur in a vacuum [41].

In this investigation, a model was developed and evaluated for the nanosecond laser ablation of W based on the more likely phenomenon of explosive partial sublimation of a superheated irradiated zone and resulting formation of a rarefaction wave that propagates through the vacuum. Plasma shielding and target heating by a laser beam tail were also evaluated. However, if targets heat to temperatures higher than material melting points during this process, the pressure in front of the target surface (generated from explosive sublimation) will prevent atoms from flying freely, which will then likely result in a homogenous boiling (volumetric vaporization) condition. Target heating by interactions with adjacent plasma, and via irradiation by plasma, were overlooked. The research team used both experimental and computational methods for this study (1) to verify the applied model and (2) to achieve a clear representation of the involved complex phenomena.

Section 2 below is devoted to explaining the implemented model and to reviewing the basic theories behind the modeling of laser ablation. Section 3 explains the experiments performed for checking the accuracy of the subject model. The associated results are accordingly presented and discussed in Section 4, with final conclusions being provided in Section 5.

2. Model and theory

We constructed its model on the basis of experimental observations through which the laser ablation of W under vacuum conditions produces many large particles (Fig. 1). Panel (a) of Fig. 1 shows that at a delay time of 1 μs, the plume is split into two parts. The front part depicts the expanding plasma, while the hot part near the surface produces particles at later delay times (panel (b)). The particle speed is ultimately dependent on the laser fluence and typically falls within the range of a fraction of a meter per second to a few meters per second. This phenomenon can be explained easily if one supposes that an irradiated target zone is explosively fragmented into many atoms and particles. The number of atoms is always expected to be much higher than the number of particles. Atoms experience many collisions and form a high-pressure vapor plume, while particles typically only undergo a few collisions that scatter them into a solid angle of 2π in front of the target surface. The pressure of the plume may also be increased by absorption of laser energy through an inverse Bremsstrahlung phenomenon. Therefore, the driving force for atoms is generally much higher than the driving force for particles. While vapor plumes can expand to approximately 1 cm away from a target surface, particles with a constant speed of 1 nm/ns can only travel 1 μm during 1 μs (see Fig. 1(a)). As a result, the plume is split into two parts, as is evident in Fig. 1(a).

These phenomena can be modeled by a partial sublimation of a superheated irradiated zone adjacent to a vacuum. Fig. 2 schematically represents a one-dimensional hydrodynamic model. A target zone reaches temperatures higher than the sublimation temperature of W. Subsequently, this superheated zone is explosively fragmented for

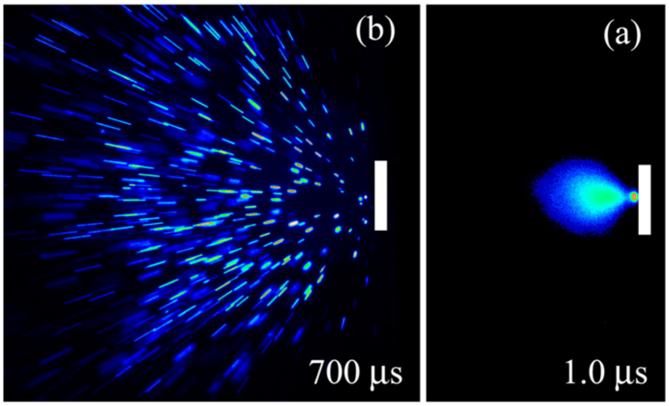

Fig. 1. Laser ablation of W under vacuum conditions by Nd:YAG laser, $\lambda = 1064$ nm, FWHM = 7 ns, and laser fluence of 131 J cm^{-2} at (a) 1 μs delay time and (b) 700 μs delay time.