Journal of Nuclear Materials 390–391 (2009) 144–147

Contents lists available at ScienceDirect

Journal of Nuclear Materials

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

Characterization of carbon and tungsten micro-particles mobilized by laser irradiation in order to develop an ITER dust removal technique

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A B S T R A C T

Due to the safety problem of dust generation in fusion facility, the preliminary step of this work is to demonstrate the ability of laser technique to eject carbon and tungsten particles from Tokamak surfaces. The laser-induced ejection mechanisms will be discussed as a function of absorption properties of dust and surface. Dynamics of mobilized particles have been investigated in order to determine an appropriate technique to collect them. For both carbon and tungsten dusts, we achieved measurements of ejection velocity as a function of laser fluence and ambient pressure. The results demonstrate the ability of the laser technique to eject dusts with very high velocities, even under few Pascal of gas pressure. They give us a better knowledge of the laser interaction mechanisms with carbon and tungsten particles and are very promising for the development of a collection technique.

1. Introduction

During ITER lifetime, interactions between plasma and plasma facing components will induce dust production [1]. These dusts will be mainly composed of carbon, tungsten and beryllium. They could modify the plasma operation and represent a potential safety issue in case of accidental release or air ingress. Then, limits have been defined in the framework of ITER licensing. In particular, the amount of particles of each material, with sizes ranging from 10 nm to 100 μm, must not exceed 6 kg on hot surfaces [2]. As a consequence, a dust collection device has to be developed to keep these inventory guidelines [3].

The first step to realize such a cleaning process is the particle mobilization. Indeed, before their collection, dusts must be moved away from the surface to reduce the surface-particle adhesion force. Laser processes have already been successfully used to clean surfaces in the field of nuclear decontamination [4], optics and microelectronics industries [5]. This technique has also been proposed in the ITER context to detritiate co-deposited carbon layer [6]. Studies have also been performed to develop a laser process for particle removal. These experimental [7,8] and theoretical [9,10] works demonstrate that dry laser cleaning technique can be considered as a very promising solution for this application. However, most of these studies have been done in the frame of microelectronic applications and concern dielectric particles on silicon substrate. The development of such a technique for dust removal in Tokamak environment requires specific studies to understand the ejection mechanisms of C, W and other metallic particles. It is also necessary to study and optimize the dynamic characteristics of the ejected particles to develop an appropriate collection system.

In this purpose, the removal efficiencies of graphite and tungsten particles deposited on various substrates have been measured for different fluence conditions. Then, measurements of velocity and ejection distance have been done as a function of the laser fluence and pressure.

2. Experiments

All experiments were carried out with a XeCl excimer laser source (CILAS, Model UV635) operating at 308 nm wavelength with pulse duration of about ≈50 ns. A pulsed laser is chosen because it allows the irradiation of surfaces with high peak power and the limitation of the residual thermal effects in the substrate.

Carbon and tungsten particles have been generated to simulate fusion reactor dust in our experiments. Those particles were created by laser ablation of graphite and tungsten targets in ambient air and were collected on a substrate set perpendicularly to the target (Fig. 1(a)). Substrates with different optical and thermal properties were used: SiO_2, Si and W. Their surfaces were pre-cleaned by using an ultrasonic bath. The focal point scans the target surface, using a translation stage, to obtain a uniform layer of isolated particles. The laser repetition rate was 40 Hz. With this technique, the particles formation processes are very similar to those occurring in a fusion reactor. Indeed, in a Tokamak, particles are created by the interactions between the plasma and the plasma facing

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0022-3115/$ - see front matter © 2009 Published by Elsevier B.V.
doi:10.1016/j.jnucmat.2009.01.151

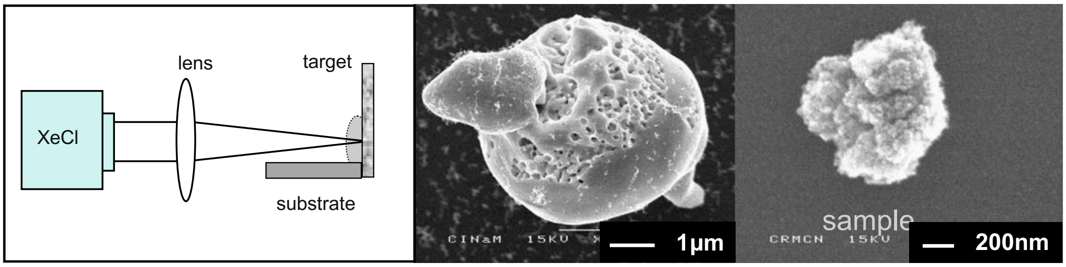

Fig. 1. (a) Setup of particles creation by laser, (b) SEM image of tungsten created by laser deposited on Si substrate, and (c) SEM image of carbon particles.

components (PFC). These interactions induce erosion involving the injection of supersaturated vapor in the edge plasma. Next, the resulting condensation leads to the formation of clusters which gives rise to solid particulates by further accretion [11]. Afterwards, these dusts are deposited on the PFC. The average size of carbon particles created with laser ablation is about 800 nm diameter. These particles have an aggregate shape or a so-called cauliflower shape which is characteristic of the particles collected in a fusion reactor (Fig. 1(c)) [12]. The average size of the tungsten particles is about 5 µm and they have a droplet shape (Fig. 1(b)).

For the laser irradiation experiments, a metallic 4.5×4.5 mm^2 mask is imaged by a f = 250 mm lens to obtain a 1×1 mm^2 near-uniform irradiation of the substrate. The laser pulse energy was varied with the aid of a manually operated beam attenuator (Optec, AT4030) to vary the laser fluence from 15 mJ/cm^2 to 1.8 J/cm^2. The particle removal efficiency (PRE) measurements were performed in ambient air with five shots. The sample is fixed on a long-range motorized translation stage that allows a precise repositioning of the irradiated zone in front of an optical microscope. Then, the particles are counted (imaging software), before and after the laser irradiation, to determine the proportion of removed particles. For the studies of the particle ejection dynamics, the samples were placed in a customized chamber to control the gas pressure. The laser irradiation induces the ejection of excited particles, and their fluorescence emission is recorded with a fast intensified CCD camera (Princeton Instrument, model 576/RB-E). The time delay between the laser pulse and the observation gate was varied with the aid of a delayed pulse generator [13].

3. Results and discussion

3.1. Ejection mechanisms

In order to understand the respective role of the substrate and dust in removal mechanisms, we studied the removal efficiency of carbon particles on various substrates, with different absorption properties (SiO_2 is a transparent material at 308 nm, α_S = 1.46 E^6 cm^{-1}, α_W = 1 E^6 cm^{-1}) and heat conductivity (K_{Si} = 123 Wm^{-1} K^{-1}, K_{SiO_2} = 1.3 Wm^{-1} K^{-1}, K_W = 174 Wm^{-1} K^{-1}). Fig. 2(a) shows the PRE curves for carbon as a function of the laser fluence. The particle removal starts at very low fluences. On silicon substrate, the cleaning effect appears at F_{las} = 60 mJ/cm^2, and the efficiency increases up to almost 100% (95% at 350 mJ/cm^2). However, the slopes of these curves are less sharp than for dielectric particles [14] which could reveal a less deterministic process. The PRE curve obtained for silica substrate is very similar which evidences that the ejection mechanism is not due to substrate motion or ablation [15]. The carbon strongly absorbs the 308 nm wavelength, and it is reasonable to think that the removal process is due to the ablation of the particle [16]. SEM analyses of the silicon substrates after the laser irradiation show some local thermal damages of the surface induced by the heat diffusion from the particles. Then, the removal mechanism seems to be the thermal ablation of the carbon particles. The difference observed between the curves obtained with Si and SiO_2 substrates is attributed to a higher thermal conductivity of Si leading to more significant losses in particle heating by diffusion towards the substrate. On tungsten substrate, the complete dust removal cannot be achieved in the same range of laser fluence and the slope of the curve is even slower than the two other ones. Even, if the Tungsten heat conductivity is slightly higher than the silicon one, it is doubtful that such a small change of the heat diffusion coefficient value can lead to a so significant modification of the PRE curve. These differences could also be due to higher adhesion forces between C particles and W surfaces, but further calculations are needed to quantify the relative importance of these mechanisms.

Fig. 2(b) shows the removal efficiency for carbon and tungsten particles on silica substrates. We can observe that the influence of the dust properties on cleaning process is much more significant than the influence of the surface properties. Even, if both particles strongly absorb the laser energy, the efficiency of W removal is limited to 50%. The saturation of this curve, for fluences higher than 1.2 J/cm^2, can be attributed to the melting of silicon surface. Previous experiments have already shown the difficulty of W particle removal compared to gold or copper particles [17]. In our situation, the removal process is certainly due to thermal ablation of

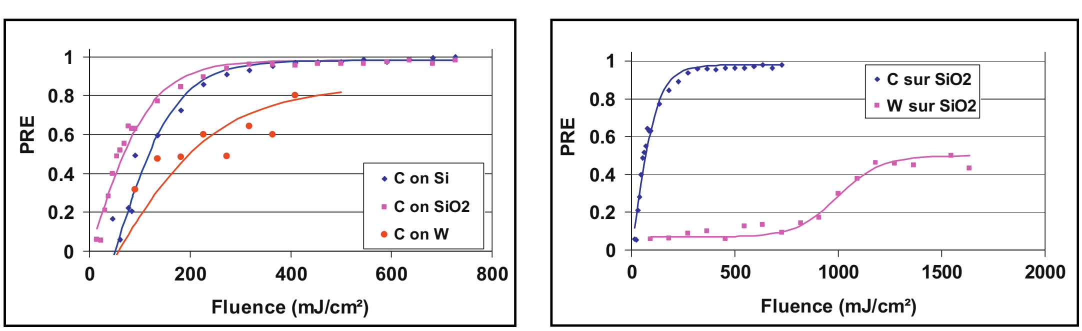

Fig. 2. (a) PRE of Carbon particles on various substrate, and (b) PRE of tungsten and carbon particles.

dust in both cases, but the particle sizes are different and, as shown in Fig. 2(a), the adhesion forces between tungsten and other materials could play a significant role. Numerical simulations are under development to have a better understanding of the cleaning process.

3.2. Ejection dynamics

Characteristics of the ejected particles must be determined to develop an appropriate collection technique. For this purpose, their ejection dynamic has been investigated. Images of the ejected dust have been taken for different delays after the laser irradiation. From these pictures we deduced the travel length as a function of time, and then the particle velocity. Fig. 3 shows a comparison between silicon and silica substrates for carbon particle removal. These ejection velocity measurements are in agreement with the PRE results previously discussed. First, the particles ejected from a silica substrate can be observed during a longer time than the particles coming from a silicon substrate which indicates a higher level of excitation. We also noticed that the mean velocity of particles ejected from silica (calculated over a 3 µs period: $v = 4240\ \mathrm{m\,s^{-1}}$) is higher than the mean velocity of particles ejected from silicon (calculated over a 1.5 µs period: $v = 3800\ \mathrm{m\,s^{-1}}$). This small difference of velocity, as well as the higher excitation of C particles on SiO_2 substrate can be explained by the difference of particle temperature. This confirms that temperature losses of particles by diffusion toward the silicon substrate play a role in the intensity of the process, but do not modify the origin of ejection process.

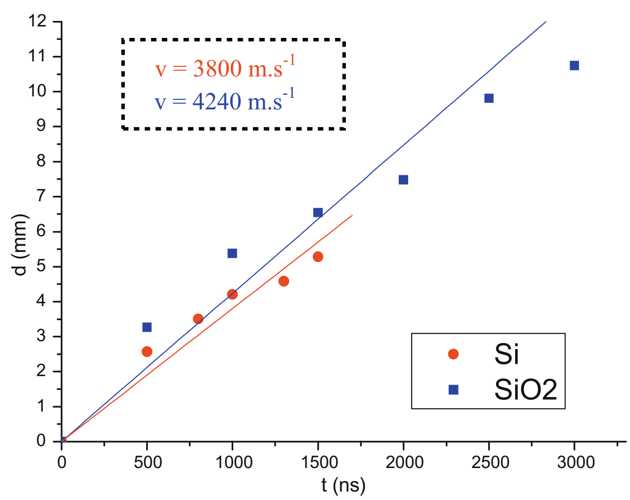

Fig. 3. Distance as a function of times of C particles at 0.1 Pa, with $F = 1.6\ \mathrm{J/cm^2}$, deposited on various substrate.

Fig. 4 shows the ejection velocity measurements released for different fluences for tungsten (Fig. 4(a)) and carbon (Fig. 4(b)) particles deposited on silicon substrate. The fluence of laser irradiation has a significant effect on the velocity and thus the distance of ejection of the particles. After 3 µs, W particles are 13 mm away from the surface for a 1.6 J/cm^2 irradiation and traveled only on 9.5 mm for an irradiation at 700 mJ/cm^2. An increase of the laser fluence induces a higher energy deposition on the particles, which become more excited and then have a higher kinetic energy and velocity. We must notice that in our situation it is difficult to eject the particles farther than 10 mm away from the surface. However, our measurement technique allows only the detection of emissive species. At 0.1 Pa, when these fluorescence emissions stop, the particle velocities are still almost constant, and we can imagine that they could continue their travel after the desexcitation processes. Further investigations with this imaging technique will be done with a continuous probe laser beam to detect, by diffusion, the light of all the particles, and not only of the emissive ones.

To investigate the influence of surrounding gas pressure on ejection dynamics, experiments have been done with a 10 Pa pressure in the irradiation chamber. Fig. 5 shows the traveling distance as a function of time for two pressures (Fig. 5(a)) and a typical image of dust plasma recorded for 10 Pa gas pressure (Fig. 5(b)). The ICCD picture shows two different plumes of Tungsten particles when the gas pressure is 10 Pa. At the beginning of the ejection, these two plumes have different velocities, then this difference decreases. After 7.5 µs, the distance between these plumes becomes constant ($d \approx 2.5$ mm). The first one (farther to the surface) is certainly related to interactions between the ejected products and ambient gas, and the emission of second one only comes from the ejected tungsten dust. A time and space-resolved spectroscopic analysis of this double plume should confirm this hypothesis. The curves show that the pressure has a strong effect on velocity losses of the particles. As expected, a higher gas pressure increases the number of collisions between gas and particles, which induces significant losses of their kinetic energy. At 1.6 J/cm^2 the plume of tungsten particles travels more than 13 mm under 0.1 Pa pressure,

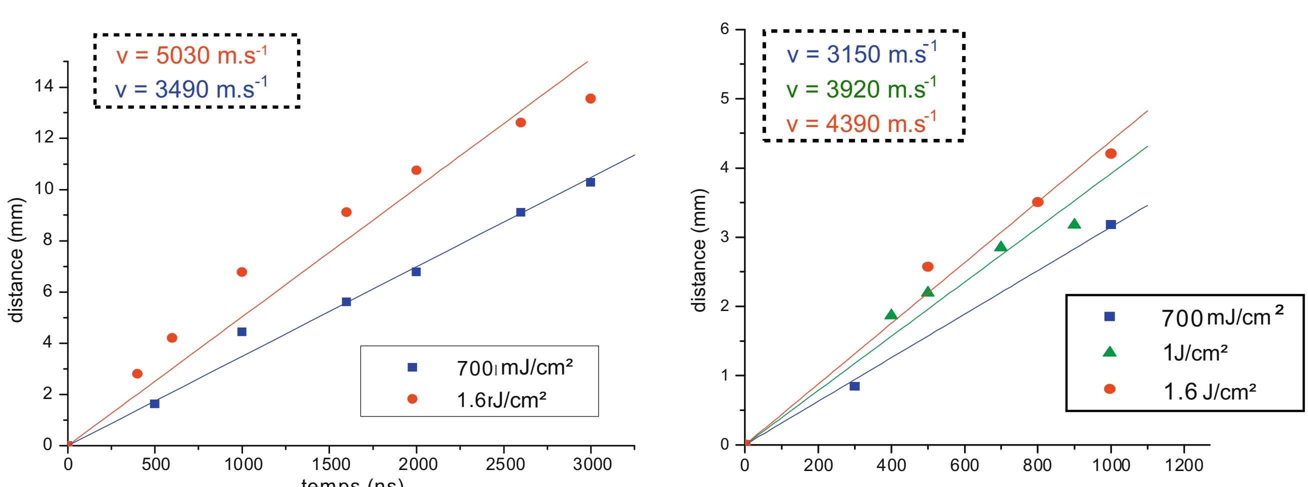

Fig. 4. Distance as a function of time for: (a) W particles on Si substrate at 0.1 Pa with various fluences, and (b) C particles on Si substrate at 0.1 Pa with various fluences. Velocity: slope of the linear regression curve.

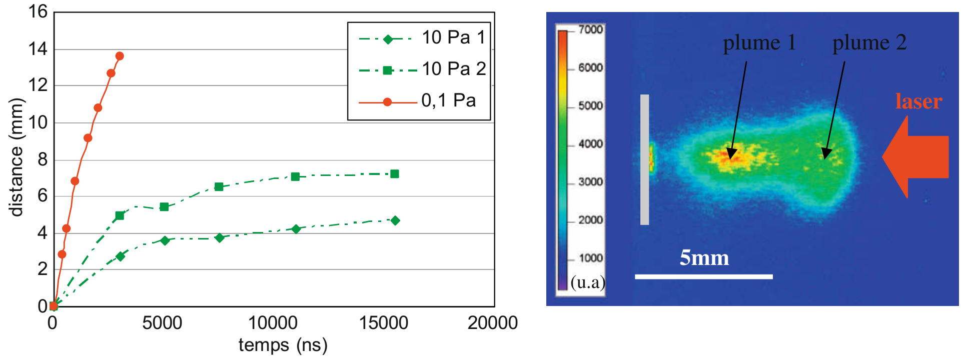

Fig. 5. (a) Distance as a function of times of W particles on Si substrate, with $F = 1.6\ J/cm^2$, under various pressure condition, and (b) ICCD picture of tungsten particles on Si substrate, with $F = 1.6\ J/cm^2$ under 10 Pa.

whereas this distance is clearly limited to 7 mm for the fastest component under 10 Pa.

4. Summary and conclusions

The main removal mechanism is the thermal degradation of the particles. Optical properties (absorptivity) of the substrate do not play any role in the ejection mechanism but some physical properties, like heat conductivity, could have an influence on the cleaning efficiency and the ejection dynamic.

The ejection distances measured with our experimental setup are around 10 mm. However these measurements took into account only the thermally excited particles. As the particles velocities are almost constant when their detection stops, we can expect a much higher ejection distance. The influence of the surrounding gas pressure is important and at 10 Pa, it is difficult to reach a travel distance greater than few millimetres. In such conditions, a collection technique based on a pumping system could be considered.

Acknowledgements

Part of this work, supported by the European Communities, was carried out within the framework of the European Fusion Development Agreement.

References

[1] J.P. Sharpe, D.A. Petti, H.-W. Bartels, Fus. Eng. Des. 63&64 (2002) 153.
[2] S. Rosanvallon, C. Grisolia, P. Andrew, S. Ciattaglia, P. Delaporte, D. Douai, D. Garnier, E. Gauthier, W. Gulden, S.H. Hong, S. Pitcher, L. Rodriguez, N. Taylor, A. Tesini, S. Vartanian, A. Vatry, M. Wykes, J. Nucl. Mater. 390–391 (2009) 57.
[3] G. Federici, R.A. Anderl, P. Andrew, J.N. Brooks, R.A. Causey, J.P. Coad, D. Cowgill, R.P. Doerner, A.A. Haasz, G. Janeschitz, W. Jacob, G.R. Longhurst, R. Nygren, A. Peacock, M.A. Pick, V. Philipps, J. Roth, C.H. Skinner, W.R. Wampler, J. Nucl. Mater. 266–269 (1999) 14.
[4] Ph. Delaporte, M. Gastaud, W. Marine, M. Sentis, O. Uteza, P. Thouvenot, J.L. Alcaraz, J.M. Le Samedy, D. Blin, Appl. Surf. Sci. 208&209 (2003) 298.
[5] W. Zapka, W. Ziemlich, A.C. Tam, Appl. Phys. Lett. 58 (1991) 2217.
[6] C. Grisolia, A. Semerok, J.M. Weulersse, F. Le Guern, S. Fomichev, F. Brygo, P. Fichet, P.Y. Thro, P. Coad, N. Bekris, M. Stamp, S. Rosanvallon, G. Piazza, J. Nucl. Mater. 363–365 (2007) 1138.
[7] D. Grojo, Ph. Delaporte, M. Sentis, O.H. Pakarinen, A.S. Foster, Appl. Phys. Lett. 92 (2008) 033108.
[8] M. Mosbacher, H.J. Munzer, J. Zimmermann, J. Solis, J. Boneberg, P. Leiderer, Appl. Phys. A: Mater. Sci. Process. 72 (2001) 41.
[9] N. Arnold, Appl. Surf. Sci. 208 (2003) 15.
[10] B.S. Luk'Yanchuk, Z.B. Wang, W.D. Song, M.H. Hong, Appl. Phys. A: Mater. Sci. Process. 79 (2004) 747.
[11] C. Arnas, C. Dominique, P. Roubin, C. Martin, C. Brosset, B. Pégourié, J. Nucl. Mater. 353 (2006) 80.
[12] J. Winter, Plasma Phys. Control. Fus. 46 (2004) B583.
[13] D. Grojo, A. Cros, Ph. Delaporte, M. Sentis, Appl. Phys. B 84 (3) (2006) 517.
[14] M. Mosbacher, V. Dobler, J. Boneberg, P. Leiderer, Appl. Phys. A 70 (2000) 669.
[15] D. Grojo, M. Boyomo-Onana, A. Cros, Ph. Delaporte, Appl. Surf. Sci. 252 (2006) 4786.
[16] D. Grojo, A. Cros, Ph. Delaporte, M. Sentis, Appl. Surf. Sci. 253 (19) (2007) 8309.
[17] C. Curran, J.M. Lee, K.G. Watkins, Opt. Laser Eng. 38 (2002) 405.