

## Plasma plume analysis and the effect of terawatt laser system irradiation on C, W layers

C. P. Lungu<sup>1</sup>, C. M. Ticos<sup>1</sup>, C. Porosnicu<sup>1</sup>, I. Jepu<sup>1</sup>, M. Lungu<sup>1</sup>, P. Dinca<sup>1</sup>, O. Pompilian<sup>1</sup>, B. Butoi<sup>1</sup>, D. Ursescu<sup>1</sup>, R. Banici<sup>1</sup>, G. Cojocaru<sup>1</sup>, R. Ungureanu<sup>1</sup>, C. Luculescu<sup>1</sup>, A. Marcu<sup>1</sup>, A. Marin<sup>2</sup>, P. Osiceanu<sup>2</sup>, I. Feraru<sup>3</sup>, C. Grigorescu<sup>3</sup>

<sup>1</sup>National Institute for Lasers, Plasma and Radiation Physics, Magurele, 077125, Romania

<sup>2</sup>"Ilie Murgulescu" Institute of Physical Chemistry, Bucharest, 060021, Romania

<sup>3</sup>National Institute R&D for Optoelectronics INOE 2000, Magurele, 077125, Romania

The plasma plume produced by a terawatt laser system (TEWALAS) was analyzed as well as the structural and morphological modifications of the tungsten and carbon layers (C films coated on top of 200 nm tungsten films) deposited on fine grain graphite substrates using thermionic vacuum arc (TVA) method.

### 1. Introduction

Fusion plasma has become in our days the most desired goal of an extensively explored research domain for energy production. An important problem still unsolved is the material composition of the reactor main chamber, aimed to resist at high energy fluxes of 10-100 MW/m<sup>2</sup>. Thus, it was proposed to be used tungsten (W) plates in the divertor region for wall protection, knowing it has the desired properties, i.e. low sputtering rate and high melting temperature. Other materials are carbon (C) as CFC - carbon fiber composite, and beryllium (Be), a light element with a relative high melting temperature: 1551 K. However, the behaviour of Be, C and W pure materials and mixed films at high flux energies is not fully understood yet and it is widely currently studied [1-2], e.g. carbon films using ns-laser irradiation [3]. We study the behaviour of a C, W multilayer deposited on fine grain graphite substrates using the original thermionic vacuum arc (TVA) method [4-5], in interaction with single or multiple ultra short, TW power pulsed laser beams. Terawatt laser (TEWALAS) facility is a multi-terawatt laser amplifier system, 0.025 - 360 x 10<sup>-12</sup> s pulse duration, up to 400 mJ pulse energy, 10 Hz maximum repetition rate [6]. 10<sup>12</sup> - 10<sup>14</sup> W/cm<sup>2</sup> power density laser beams were focused on the prepared pure and mixed layers in vacuum (< 10<sup>-3</sup> Pa).

### 2. Experiment

**2.1. Preparation** of the carbon and tungsten films was achieved using the TVA method developed at NILPRP that uses an electron beam emitted by an

externally heated cathode (a grounded tungsten filament) accelerated by a high anodic voltage. The thickness of the films was in situ measured with a quartz micro-balance. Deposition temperature was 600<sup>o</sup> C and coating rates were ~ 0.5 Å/s for W and ~ 11 Å/s for C.

**2.2 Laser irradiation** was performed using the TEWALAS laser system at 800 nm wavelength. The exposure was performed in high vacuum at a residual pressure of 1.3 x 10<sup>-4</sup> Pa. Sample surface was irradiated with a train of long laser pulses, followed by a short laser pulse. Long pulse line length was 2.8 mm (~ 0.17 μm<sup>2</sup>, the focal spot area) and the pulse energy was 112 mJ whereas the short pulse line length was 3.2 mm and the laser pulse energy was 151.2 mJ (~ 0.33 μm<sup>2</sup>, the focal spot area). A schematic diagram presenting the laser irradiation geometry of C, and W layers deposited on graphite substrate is presented in Fig.1.

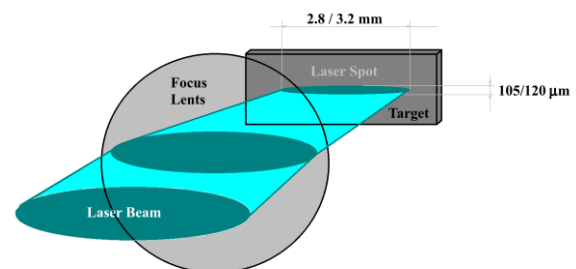


Fig. 1 Schematic diagram of the laser irradiation.

**2.3 Plasma plume analysis** was in-situ performed, by spectra emission recording in the 10-22 nm range. The results are presented for the first 10 (long, 360-ps duration) successive pulses in the same spot on W film coated on graphite substrate.

As shown in Fig. 2, there is a band emission centred in the 14-18 nm interval fluctuating with the number of laser pulses. Considering the big differences in the number of electrons between the two elements, respectively W (74) and C (6) respectively, the band emission should belong to W while C should rather give distinct emission lines as visible in Fig. 2 (e.g. pulse 1). By assuming that indeed the emission belongs to W, we could conclude that we actually ablate the W layer starting from the 3rd laser pulse for the following 3-4 pulses. Using this assumption, we have no band emission before and after this interval, while the carbon layer and respectively substrate (graphite) are being ablated.

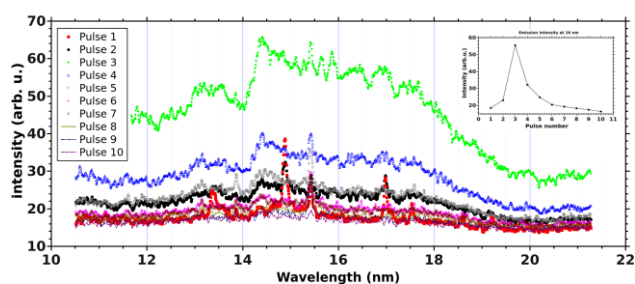


Fig. 2. The emission in a 10-22 nm range for 10 successive long pulses. Inset, spectral emission intensity in the middle of the recorded emission band at ~16 nm, assigned to tungsten.

We assume that the ablation of the carbon layer is made in the center of the spot by the first few pulses; during the following several pulses the W layer is ablated and after that, the laser pulse is interacting with the graphite substrate. However, due to the Gaussian energy profile of the beam it leads to weaker interactions on the side zones.

### 3 Results

**3.1 SEM investigations** gave us information about the laser-matter interaction in our experiment as reflected in the morphology changes. The laser beam starts producing ablation from the centre of the irradiated area where the fluence approaches 2 J/cm<sup>2</sup>.



Fig. 3 SEM details of central irradiated area surface for the C/W/graphite structure.

The sample is ablated starting from the first laser pulse and the top carbon layer coated on W film is initially completely vaporized from areas of about 100 mm wide and about 2 mm long (Fig. 3) yielding a minimal laser fluence of about 1.7 J/cm<sup>2</sup>.

**3.1 The EDX investigations** confirmed that the white zone visible in the SEM images (Fig. 3) is the tungsten layer uncovered from the carbon layer. This tungsten layer (about 200 nm thick) is also ablated from areas of about 10 μm wide corresponding to laser fluencies close to the maximal one (~ 2 J/cm<sup>2</sup>) of the very first pulse. For laser-induced structural modifications we roughly notice 3 zones in the investigated areas. An intensely irradiated zone in the centre of the beam, where the ablation process is centred, an average irradiated zone assimilated with the limits of the measured ablated zone, and a low power density zone outside the traces of the ablation process, where there are no morphological modifications, although the beam energy density is still not null. We started the structural exploration with the intense irradiated zone.

**3.2 Micro-Raman investigations** in its centre has pointed the unexpected presence of diamond crystals (1330 cm<sup>-1</sup>) [7], as can be observed in Fig 4.

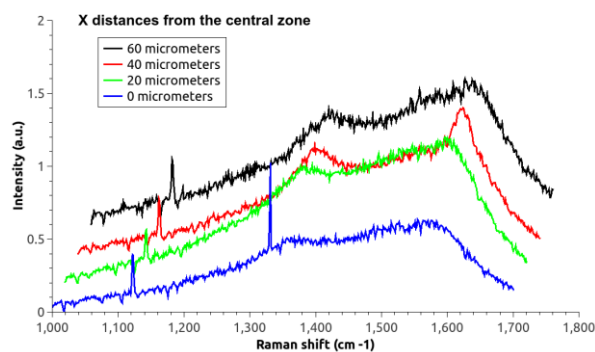


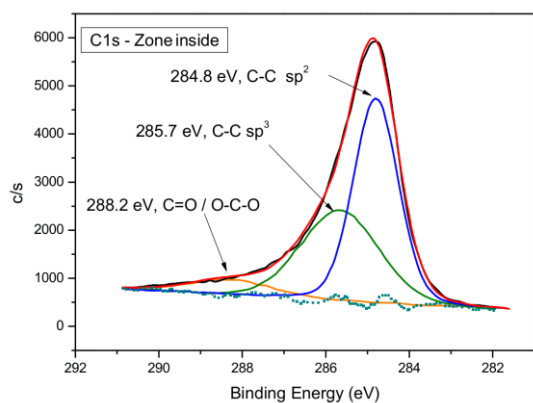
Fig. 4 Diamond signal in Raman measurements in the centre of '5 pulses' irradiation zone

Also, a peak is observed around 1123 cm<sup>-1</sup>, whose intensity varies with the linear position on the ablated zone, i.e. stronger in the centre and weaker towards the edges. The appearance of those signals just in the centres of the ablated areas could be explained by diamond nanocrystal formation through a mechanism known to rely on the plume formation and respectively expansion, for nanosecond and picosecond laser ablation. Variations in the Raman spectra of nanocrystalline phases are generally described by relaxation of the

$q=0$  selection rule, due to uncertainty in the value of the wave vector and thus allowing for activation of phonons that would not usually be observed in the spectrum of the bulk material.

**3.3 The SAED** analysis on the edge of the laser irradiation area, allowed the identification of rhombohedral structures with lattice parameters  $a = 0.25221$  nm,  $c = 4.3245$  nm corresponding to diamond (ASTM 79-1473). The SAED pattern distances were calculated using reciprocal space.

**3.4 XPS** analysis permitted the deduction of the  $sp^3$  fraction of the films was deduced by fitting the XPS spectra for C 1s core peaks. At the outside of the irradiated zone, the surface peaks corresponding to CO-contamination (290.4 eV, O=C-O and 288.2 eV, C=O/O-C-O) and graphite (284.8 eV, C-C  $sp^2$ ) have a greater contribution than that of diamond (285.7 eV, C-C  $sp^3$ ).



a)

b)

Fig. 5 Deconvolution of the XPS C1s spectrum corresponding to the irradiated inside zone.

#### 4. Conclusions

Tungsten and carbon films with the thickness of about 2.5  $\mu$ m were coated on fine grain graphite (the C films were coated on the intermediary 200 nm tungsten films) and were irradiated using ultrashort laser pulses of as much as 150 mJ energy and  $0.1 - 360 \times 10^{-12}$  s pulse duration.

The plasma plume was characterized by optical emission spectroscopy, the structure and the morphology of the ablated area produced by the laser irradiation contain uniform distributed droplets zones as observed in the SEM images, which correspond to rhombohedral structures with lattice parameters  $a = 0.25221$  nm,  $c = 4.3245$  nm (diamond) as identified by SAED analysis. The

micro-Raman scattering measurements performed on the craters in comparison with the spectrum of a diamond tip show the  $1330$   $cm^{-1}$  wide enough to allow a discontinuous diamond structure interpretation. SEM images associated with energy dispersive spectrometry (EDS) analysis prove the existence of W particles in the diamond-graphite structure of the irradiated zones. The ratio of  $sp^3/sp^2$  bonds estimated using XPS was found larger than 60%. The Raman characterization leads to the conclusion that the films contain of NCD and NC graphite as a result of hysteresis.

#### 5. References

- [1] Ch. Linsmeier, M. Reinelt, K. Schmid, J. Nucl. Mat. 415 (2011) S212.
- [2] G.M. Wright, M. Mayer, K. Ertl, G. de Saint-Aubin, J. Rapp, J. Nucl. Mat. 415 (2011) S236.
- [3] G. Dumitru, V. Romano, H. P. Weber, S. Pimenov, T. Kononenko, M. Sentis, J. Hermann, S. Bruneau, Appl. Surf. Sci., 222 (2004) 226.
- [4] G. Musa, I. Mustata, V. Ciupina, R. Vladoiu, G. Prodan, C. P. Lungu, H. Ehrich, J. Optoelectronics & Adv. Mat. 7 (2005) 2485.
- [5] I. Jepu, C. Porosnicu, I. Mustata, C. P. Lungu, V. Kunkser, M. Osiac, G. Iacobescu, V. Ionescu, T. Tudor, Rom. Rep. Phys. 63 (2011) 804.
- [6] R. Dabu, R. Banici, C. Blanaru, C. Fenic, L. Ionel, F. Jipa, L. Rusen, S. Simion, A. Stratan, M. Ulmeanu, D. Ursescu, M. Zamfirescu, J. Optoelectronics and Advanced Materials 12 (2010) 12.
- [7] A. C. Ferrari and J. Robertson, Phys. Rev. B 63 (2001) 121405 (R).

**Acknowledgements** This work was supported by a grant of the Romanian National Authority for Scientific Research, CNCS – UEFISCDI, project number PN-II-IDPCE- 2011-3-0522