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STUDY OF LASER-PRODUCED PLASMAS FROM BORON, CARBON AND BORON-CARBIDE TARGETS

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Dedicated to Professor Kseno Ilakovac on the occasion of his 70th birthday

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Spectroscopic investigations were made on plasma clouds created by 20 ns, 3 J ruby laser pulses impinging perpendicularly onto targets of boron carbide, carbon and boron. The irradiance on the targets was about 132 GW cm^{-2}. Time-resolved spectra of plasmas in the region of wavelength from 16 to 32 nm were observed at a distance of 1 mm from the targets. The maximum electron temperatures were about 60 eV in the case of carbon and boron targets, and about 45 eV in the case of boron-carbide target. Laser evaporation from carbon occurred directly from the solid state (sublimation), and in the case of a boron and boron-carbide melting was observed as an intermediate state.

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1. *Introduction*

Boron carbide (B_4C) is an interesting ceramic because of its useful properties: it is a hard, lightweight material used in the nuclear and aerospace industries (abrasive flow control, grit blast and sandblast nozzles, armour, neutron absorbers, etc.). After diamond, cubical boron nitride and boron oxide, boron carbide is the hardest known material at room temperature [1]. Most of studies were made about its characteristics as a deposition material (thin films). This work is oriented towards a basic study: plasma behaviour and a comparison with plasmas of its pure constituents – pure boron and pure carbon plasmas produced under the same conditions, as well as the microscopic observations of the surfaces. Laser-produced plasmas from boron-carbide targets have previously been studied (see Refs. [2] and [3]). Similar, but more extensive investigations have been done with boron-nitride plasmas (see, e.g., Ref. [4]).

2. *Experimental set-up*

Plasmas were produced by a ruby laser (KORAD K1 laser and K1500 amplifier) with a pulse energy of 3 J and pulse duration up to 20 ns. The laser beam was focused perpendicularly onto the flat surfaces of the targets placed in a vacuum chamber (10^{-5} mbar). Each laser pulse impinged always onto a position place on the target surface. The plasma clouds were observed side-on with a VUV flat-field grazing incidence spectrograph, equipped with a flat gateable microchannel plate (MCP) and a CCD camera [5].

Experimental set-up and equipment are shown on Fig. 1. Cross-section of the laser beam on the target was 2.0×10^{-3} cm^2. The laser fluence was about 1.5 kJ cm^{-2} and the irradiance about 132 GW cm^{-2}.

Technical properties of targets are shown in Table 1.

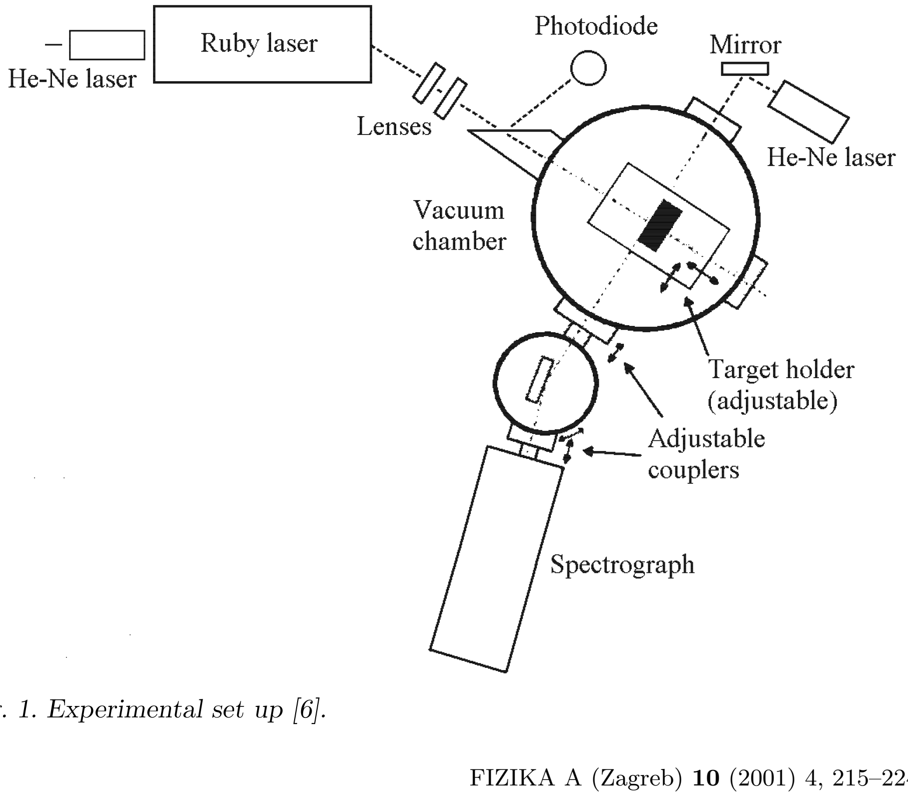


Fig. 1. Experimental set up [6].

TABLE 1. Technical properties of targets (Refs. [1,7]).

	Boron	Glassy carbon (Sigradur G) Carbon	Boron carbide (hot-pressed) B_4C
Density	$2.34 - 2.37$ g cm^{-3}	1.42 g cm^{-3}	2.45 g cm^{-3}
Porosity	0%	0%	0%
Melting point	2180 °C	3000 °C	2450 °C
Boiling point (normal pressure)	3700 °C	(4827 °C)	3000 °C

3. *Spectral analysis*

Time-resolved spectra of the plasma radiation were observed with a time resolution ($\Delta\tau$) of about 10 ns, with variable time delay after the pulse, in the spectral region 16 to 32 nm, with an instrumental resolution (FWHM) of about 0.1 nm. The plasmas were observed at (1.0 ± 0.2) mm distance from the target surface. Same observations were done quite near the surface, at about 0.5 mm.

Figure 2 presents an example of a spectrum recorded at the time of maximum

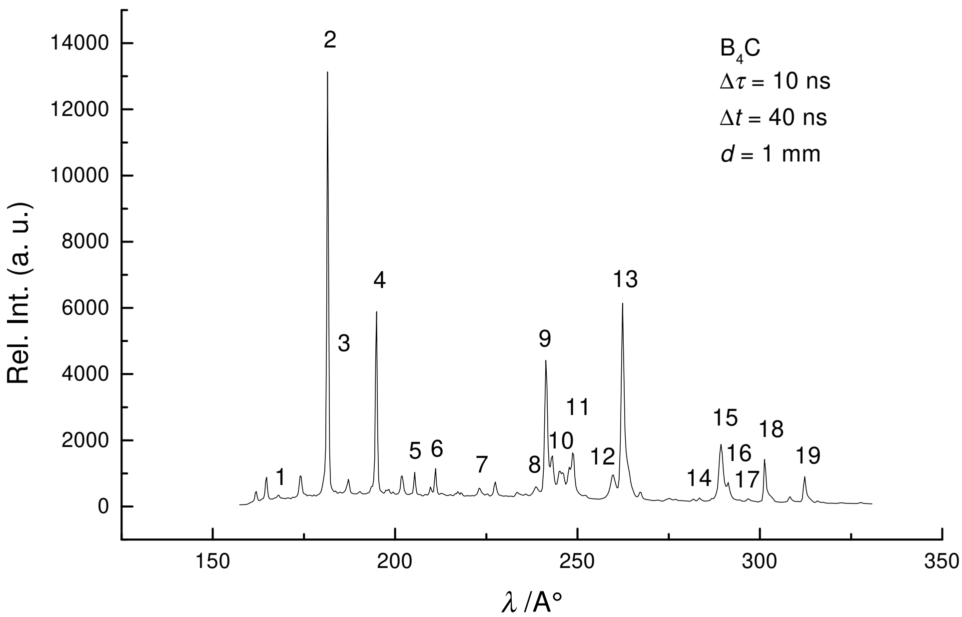


Fig. 2. Example of spectrum emitted by a B_4C *plasma.*

TABLE 2. Spectral lines λ_{th} Refs. [8,9].

No.	λ_{th} (Å)	Atom (ion)	Transition
1	5 × 33.737	C VI	1s – 2p (Lα)
2	3 × 60.3144	B IV	$1s^2$ – 1s2p
3	182.097	C VI	2s - 3p
	182.230	C VI	2p – 3d (H$_\alpha$)
4	4 × 48.586	B V	1s – 2p (Lα)
5	6 × 34.973	C V	$1s^2$ – 1s3p
6	4 × 52.682	B IV	$1s^2$ – 1s3p
7	222.79	C IV	$1s^22s$ – $1s^25p$
8	238.23	C IV	$1s^22p$ – $1s^27d$
9	4 × 60.3144	B IV	$1s^2$ – 1s2p
10	5 × 48.586	B V	1s – 2p (Lα)
	244.91	C IV	$1s^22s$ – $1s^24p$
	7 × 34.973	C V	$1s^2$ – 1s3p
11	248.71	C V	1s2p – 1s3d
12	259.52	C IV	$1s^22p$ – $1s^25d$
13	262.60	C IV	$1s^22p$ – $1s^25s$
14	283.60	C IV	1s2s2p – 1s2s3d
15	289.20	C IV	$1s^22p$ – $1s^24d$
16	6 × 48.586	B V	1s – 2p (Lα)
17	296.92	C IV	$1s^22p$ – $1s^24s$
18	5 × 60.3144	B IV	$1s^2$ – 1s2p
19	312.43	C IV	$1s^22s$ – $1s^23p$

of the B_4C plasma emission. The time period between the maximum of laser irradiation and the time when the spectrum was recorded was $\Delta t = 40$ ns.

The lines of the emission spectra were from the hydrogen-like and helium-like ionization stages of boron (B V and B IV) in boron and boron-carbide plasmas. The carbon emission lines in pure carbon and boron-carbide plasmas were in the hydrogen-like, the helium-like and the lithium-like ionization stages (C VI, C V and C IV). The significant lines in all of the analysed plasmas are presented in Table 2.

From the highly-ionized stages of atoms in plasmas, we can deduce electron temperatures of close to 50 eV (1 eV corresponds to 11600 K) for all observed plasmas. The temperatures were estimated assuming the upper levels of the lines were in the partial local temperature equilibrium (PLTE) with the ground state of the next ionization stages and both ionization stages were in coronal equilibrium, using the formula [10]

$$R = \frac{i'}{i} = \frac{\omega' A' g'}{w A g} \exp\left(\frac{E'_\infty - E' - E_\infty + E}{kT}\right) \frac{G_i S}{g'_i \alpha}, \qquad (1)$$

where the relative intensities i and i' denote lines of two successive ionization stages. In this way, we have two sets of parameters in which ω stands for the frequency, A for the transition probability, g for the upper-level statistical weight, E for the excitation energy, E_∞ for the ionization energy of the lower ionization stage and g_i for the ground-state statistical weights of the next ionization stage (for example C V and C VI). The ionization and recombination coefficients (S, α) are considered as those for the "ion" (ionization of, for example, C V and recombination leading into C V from C VI) [10].

From the relative intensity ratio of two spectral lines of successive ionization stages, $\lambda_{\mathrm{C\,V}} = 24.87$ nm, 1s2p ^{3}P^0 → 1s3d ^{3}D, and $\lambda_{\mathrm{C\,IV}} = 28.92$ nm, 1s^{2}2p ^{2}P^0 → 1s^{2}4d ^{2}D, at 1 mm distance from the target surface, we estimated the following electron temperatures: $T_e \approx 60$ eV in pure carbon plasma, and $T_e \approx 45$ eV in boron-carbide plasma. In the first period of target ablation (the delay of about 4 ns) and in the last period (the delay of about 60 ns), the temperatures were lower. Temperatures reached maximum at the time when the emission was maximal (at about 30 ns). The obtained temperature in pure carbon plasma is in good agreement with the earlier results of similar measurements [11].

The recorded spectra reveal that the excitation of boron ions in boron-carbide plasmas is significantly stronger than the excitation of carbon ions (see Fig. 2). This is due to molecular configuration of B_4C as well as the different energies needed for the excitation of boron and carbon atoms. Observed lines indicate that the electron temperature is closer to 45 eV when B IV ions have the maximum concentration, than to 60 eV when the concentration of C V ions is at maximum.

From the observed equal maximal intensities of the Lyman-alpha lines in pure carbon and pure boron plasmas, we can deduce that the electron temperatures of both plasmas are comparable, i.e. about 60 eV.

We could not see any significant difference of the excitation of boron ions in boron-carbide and pure boron plasmas, but it was seen in the excitation of carbon ions. The line intensities of carbon lines were much weaker in boron-carbide plasmas than in carbon plasmas. An example is shown in Fig. 3.

Figure 4 shows a comparison of the time dependence of relative intensities of several ionic lines (C V, 1s2p → 1s3d; C IV, 1s^{2}2s → 1s^{2}3p; B IV, 1s^2 → 1s^{2}p) in boron-carbide plasmas. Ions of lower ionization stages, especially strong B IV, show slower intensity decrease than ions of higher ionization stages due to the effective recombination of the latter.

From the spectral analyses at two different distances from the target surface, we estimated the velocity of plasma expansion from about 7×10^6 cm/s to about 10×10^6 cm/s in all measurements.

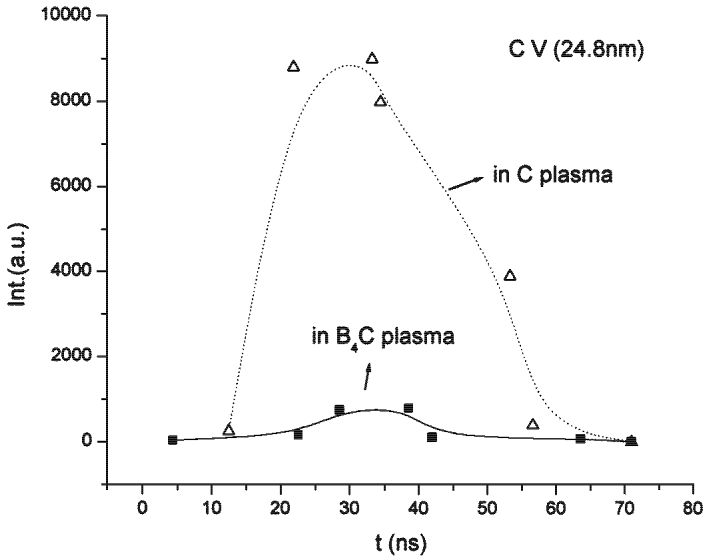


Fig. 3. Comparison of carbon line intensities in pure carbon and in boron-carbide plasmas.

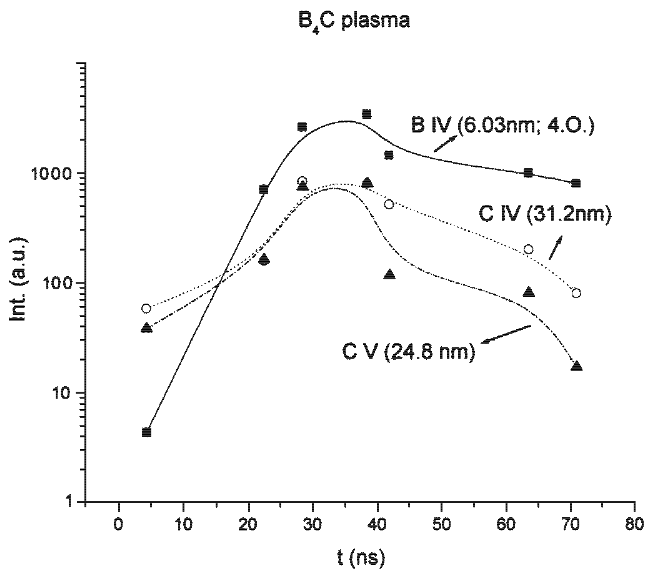
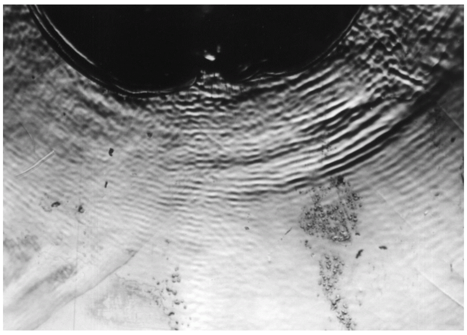


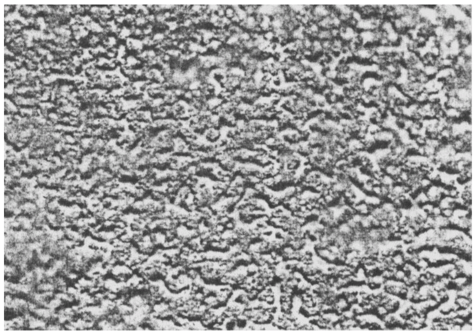
Fig. 4. Time dependence of relative line intensities for boron and carbon in boron-carbide plasma.

4. Light microscopic observations

Target surfaces were observed by a metallurgical light-microscope (Leitz – Aristomet). On the surface of carbon (sigradur) craters with sharp edges were observed (Fig. 5). The central part of the craters was the result of the sublimation process of carbon. Average radius of the craters is 1 mm. Also, periodical ripples are visible closed to the edge of the crater. They are the result of a plastic deformations due to a high pressure shock caused by the laser beam. The measured period of the ripple structure is about 30 μm, what is much larger then the laser wavelength.

After the laser irradiation, rather uniform formations are visible on the boron surface (Fig. 6) due to the melting, fast cooling and subsequent crystallization. Average radius of these formations is 1 μm. The spheroids merge and form open-cell formations because of the very high surface temperature. Interaction with the laser beam leaves the boron-carbide surface with cracks (due to cooling) and small craters (diameter up to 10 μm). This surface is shown in Fig. 7. The edges of the craters can be observed as the vortex formations, simple





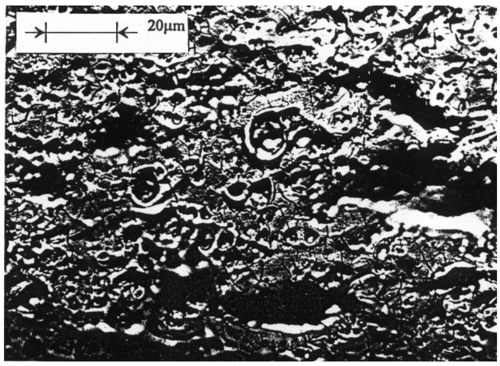


Fig. 5 (left). C surface with craters irradiated by ruby laser (magnification 600×*).*

Fig. 6 (right). B surface irradiated by ruby laser (magnification 600×*).*

Fig. 7 (bottom). B_4C *surface irradiated by ruby laser (magnification* 600×*).*

ring-type that could be compared with analysis of tantalum surfaces irradiated with the nanosecond laser pulses [12]. Plasma evaporations developed from these craters by microexplosions.

5. *Conclusions*

The presented results lead us to the following conclusions:

(i) Emission spectra from B_4C plasma plumes have been recorded. With a laser irradiation of 132 GW cm^{-2}, produced by a ruby laser, emission of C IV to C VI lines as well as of B IV and B V lines in the spectral region 17 to 32 nm was observed. In previous similar measurements [13], made with lower laser intensities of about 1.25 GW cm^{-2}, only emission of B I, B II and C II lines (in the visible region of spectrum) was observed. In both cases, the relative intensity of carbon lines in B_4C plasma was drastically weaker than the intensity of boron lines.

(ii) When we irradiated a carbon target with the 132 GW cm^{-2} laser intensity (ruby laser), spectral lines from C IV to C VI were observed. Similar investigations had been performed earlier with the same ruby laser, but with a lower laser intensity of about 1.4 GW cm^{-2} [14]. In that case, C IV carbon spectral lines at the maximum stage of ionization were observed. Threshold for the ionization of carbon atoms was achieved at sub-GW cm^{-2} laser intensities [15].

In some previous investigations [16] made with sub-ps lasers, the intensity was about 4×10^{15} W cm^{-2} and hydrogen-like boron plasmas (soft X-ray emission) were observed, what we achieved in our work with a ns laser at a much lower intensity of about 1.32×10^{11} W cm^{-2}. Recombination is the explanation for the observed slower intensity decrease of lines from lower ionization stages (Fig. 4).

(iii) We estimated that the electron temperature in the boron plasmas was comparable with the temperature in the carbon plasmas. In the B_4C plasmas, the electron temperature was approximately 45 eV and thus about 10 eV lower than in the plasmas of the pure elements. The measurements are in good agreement with previous observations [4]. The large error of the line intensities (20%) reflects the long integration time (10 ns).

(iv) The ablation processes are different in carbon and in carbide compounds. We find laser evaporation from carbon (sigradur) directly from the solid state. Such behaviour was also observed in a previous experiment [17] which was done with sigradur and a CO_2 laser at a lower laser irradiation of about 5×10^7 W cm^{-2}. However, in the case of boron and boron-carbide, melting is observed as an intermediate state.

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ISTRAŽIVANJE PLAZME PROIZVEDENE LASEROM IZ BORA, UGLJIKA I BOROVOG KARBIDA

Izveli smo spektroskopska istraživanja oblaka plazme koju smo proizveli rubidijskim laserom pulsevima 20 ns i energije 3 J sa snopom usmjerenim okomito na mete borovog karbida, ugljika i bora. Intenzitet snopa bio je oko 132 GW cm^{-2}. Mjerili smo vremenski razlučene spektre plazme u području valne duljine 16 do 32 nm oko 1 mm nad metama. Maksimum elektronske temperature bio je oko 60 eV s ugljikovim i borovim metama, a 45 eV s borovim karbidom. Lasersko uparivanje iz ugljika dešava se izravno iz čvrstog stanja (sublimacija), dok smo opazili talenje kao međustanje s metama bora i borovog karbida.