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ABSTRACT

Boron thin films were produced using femtosecond pulsed laser deposition aiming at the development of a neutron detector. As neutrons have no charge, in order to detect this particle converter materials are applied, promoting nuclear reactions that result in the emission of charged particles, allowing the neutrons presence to be indirectly inferred. Among the possible conversion materials, ^{10}B has a considerable cross section for thermal neutrons and accessible cost. Furthermore, the nuclear reaction produces easily detectable alpha particles, making it a rational option to develop a low cost and portable neutron detector. The boron ablation threshold fluence has been measured by the Diagonal Scan (D-scan) technique that resulted in the minimum laser energy value of 17.7 (6) μJ and fluence of 5.63(19) J/cm^2. Boron deposition was performed varying the pulse energy and deposition duration. The growth rate, morphological and physical aspects of the boron pulsed laser deposition were characterized by a Scanning Electron Microscope and an optical profilometer. The films surfaces have a flaky aspect with eventual droplets which had decayed overtime to a more smooth surface. The studied parameters allowed producing a boron coating with the optimal thickness in order to minimize self-absorption effect in the film, thus increasing efficiency.

Keywords: Femtosecond laser, low coherence, laser ablation, D-Scan technique, boron thin film, neutron detector.

1. INTRODUCTION

The development of a low-cost thermal neutron detector can be achieved using a boron thin film coupled to a silicon photodiode. The use of silicon photodiode offers several advantages[1], such as: can be easily obtained commercially, operational at room temperature and has a compact electronics associated. Thermal neutron detectors using boron or boron compounds as converter materials associated with diodes are already available[2], including a portable model[3] for individual monitoring.

The neutron reaction[4] ^{10}B(n,α)^{7}Li, illustrated in Figure 1, produces an alpha particle and a lithium nuclei. The use of ^{10}B as the neutron converter has some advantages[3,5,6], such as high cross-section to thermal neutrons (~3840b at 0.026 eV), high isotopic abundance (19.8%), besides being solid state and chemically inert, 100% probability of alpha particle emission and is available commercially.

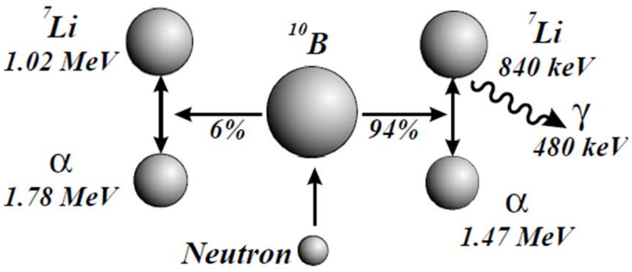

Figure 1. Reaction channels from ^{10}B(n,α)^{7}Li.

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However, boron foils manufactured[2] from ^{10}B are not readily available due to the increased cost and the material preparation process for creating the foils, which is difficult considering that the boron is a brittle material. Furthermore, it has a high melting point[5] to prepare by an ordinary evaporation method. An alternative solution for this situation is the use of the pulsed laser deposition technology (PLD), which has the ability[5] to produce a stable boron film that has stable pulse energy intensity

The PLD technique has already been performed for boron and boron compounds thin films fabrication using excimer[7], Nd:YAG[5,8–11], and femtosecond lasers[12,13]. Pulsed laser deposition using femtosecond lasers has some advantages in relation to Nd:YAG (nanoseconds), for instance, the decrease of the threshold ablation fluence[14], due to rapid energy[15] deposition, the avoidance of laser-plume interaction and heat-affected zones in the irradiated targets are strongly localized with minimal residual damage. In addition, boron thin films[12] made using femtosecond pulses present a lower stress and higher adhesion to the substrate, and also tend to be more efficient as the boron ejects are more atomized rather than the clustered aspect from ns lasers that results in material losses.

The aim of this work was to measure the boron ablation threshold fluence and the deposition rate together with the film morphological and physical aspects aiming the development of a low cost neutron detector.

2. MATERIALS AND METHODS

A femtosecond pulsed laser system was used for the determination of the ablation threshold of the target, and also for the boron thin film deposition. This system consists in a CPA Ti:Sapphire laser (Femtopower Compact Pro CE-Phase HP/HR from Femtolasers), continuously generating 25 fs (FWHM) pulses centered at 775 nm with 40 nm of bandwidth (FWHM), at a maximum repetition rate of 4 kHz. The target is a boron disc (diameter: 25.6 mm, thickness: 2 mm) from American Elements (Figure 2).

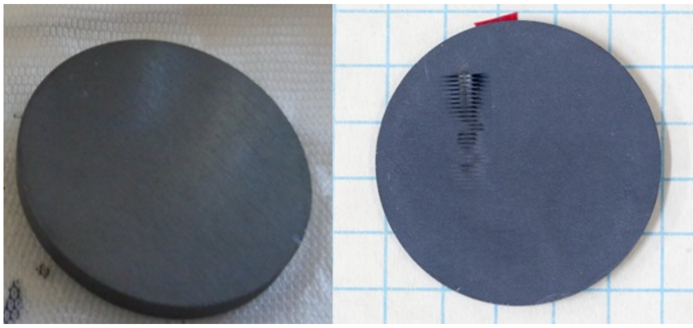

Figure 2. Metallic enriched boron before (left) and after the determination of ablation threshold (right)

The boron ablation threshold fluence has been measured to be 0.4 J/cm^2 (for 100 pulses) using the D-Scan technique[16], in which a surface damage (Figure 3) is made by exposure to a laser beam and the sample is translated in the +z and +y directions simultaneously[17].

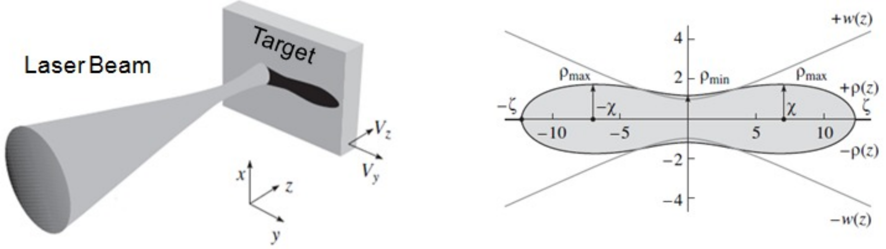

Figure 3. Surface damage threshold determination under exposure to laser beam (left) and profile etched on the surface of a sample by the D-Scan method (right)

The profile etched on the surface of a sample is shown in figure 3, which offers the half of the ablation profile maximum transversal dimension ($\rho_{max)}$ value, which allows to determinate the fluence (F_{th}) and the number of pulses (N) using equations 1 and 2[16], where E_0 is the pulse energy, V_y is the transversal velocity, ϑ_3 is the Jacobi theta function and f is the laser repetition rate.

$$F_{th} \cong 0.117 \frac{E_0}{\rho_{\max}^2} \quad (1)$$

And

$$N = \vartheta_3\left(0, e^{-\left(\frac{V_y}{f\rho_{máx}}\right)^2}\right) \quad (2)$$

The experimental setup for boron film deposition (Figure 4) consists of a vacuum chamber, in which the laser beam is aligned through a 15 cm focusing lens. This beam is direct at the target (boron) having its varied incidence position using a beam oscillator. Inside this chamber occurs the ablation of the target that is rotated by a motor, and the plume is formed going towards the substrate (microscope slide), thus forming the film on its surface. The vacuum was generated by a vacuum pump reaching the pressure of 7.5×10^{-2} mTorr, and the distance from the target to the substrate was fixed in 3.5 cm. Boron thin films were made on the surface of microscope slides in triplicate, using pulse energies of: 37(6), 177(6) and 530(7) μJ, varying the deposition time.

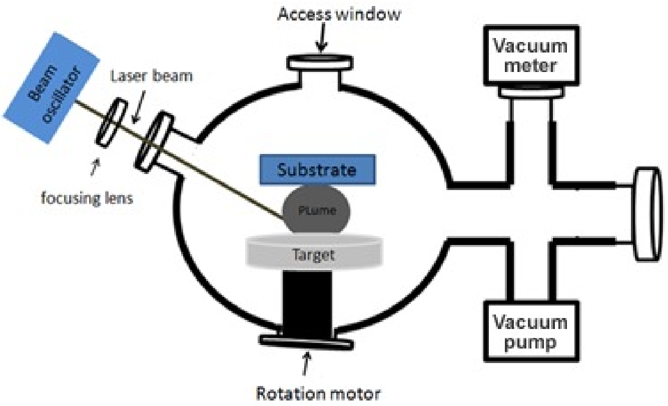

Figure 4. Experimental setup of boron deposition by femtosecond pulsed laser. The focusing lens has 150 mm of effective focal length.

3. RESULTS AND DISCUSSION

The boron ablation threshold fluence dependence on the pulses superposition obtained by the D-Scan technique is shown in Figure 5. This graph allowed determining the boron target ablation threshold to be 0.89(5) J/cm^2, which in our setup (15 cm focusing lens) corresponds to a pulse energy of 17.7(6) μJ and fluence of 5,63(19) J/cm^2. Aiming to test different situations for boron deposition on the microscope slides the pulse energies used were 37(6), 177(6) and 530(7) μJ, resulting in the following fluences: 11.8 (19), 56.3(19) and 168.7(22) J/cm^2.

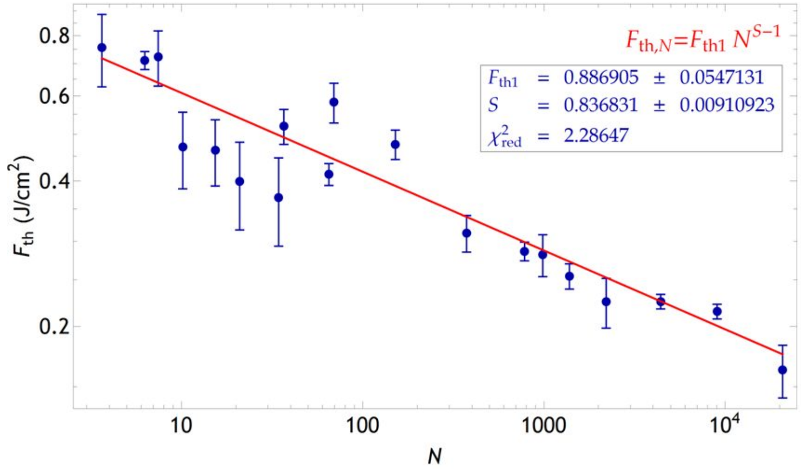

Figure 5. Boron ablation threshold fluence (F_{th}) dependence on the pulses superposition (N) obtained by D-Scan technique

The morphological and physical aspects of the boron films were analyzed using a Scanning Electron Microscope (SEM) model TM300 from Hitachi. The films did not present drastical morphology changes between different deposition times. Figure 6 presents four films taken 18 months after the deposition, and they differs greatly from the aspect of it from images taken few days after the deposition, Figure 7, pointing that the film may have reacted with air and changed its appearance, however for the aimed application this is not an issue. From this results is possible to note that the films surface present granulation and droplets, which already was been reported[18] due to the fact that the high laser energy of the microscopic and macroscopic particles[19] that are ejected from the target.

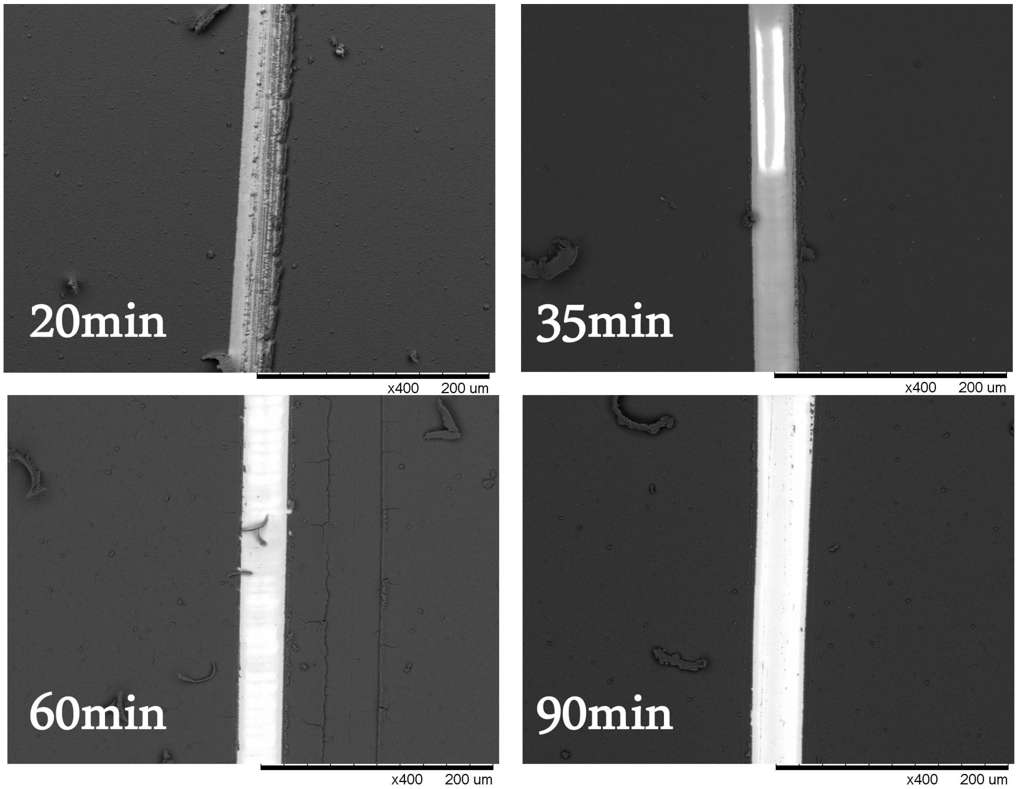

Figure 6. Boron films with different time deposition with their respective surface images obtained by SEM.

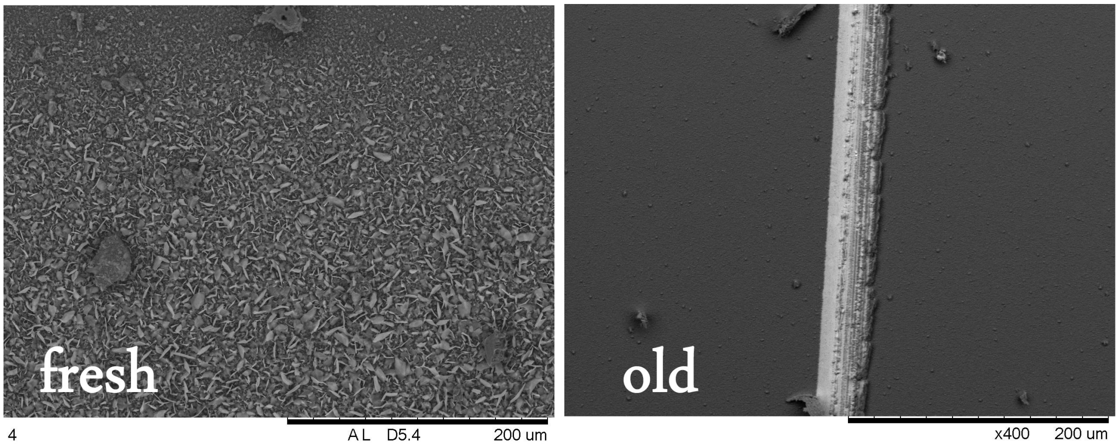

Figure 7. Boron films images taken few days after the deposition (left) and 18 months after deposition (right).

The growth rates of the boron deposition were characterized varying the deposition duration and laser energy, and the films were made in triplicate for each deposition time. The boron films thickness were measured using a profilometer (ZeGage - Zygo). The relation between the average thickness and time deposition is shown in Figure 8 in function of pulse energy, considering the region with the highest values of thickness.

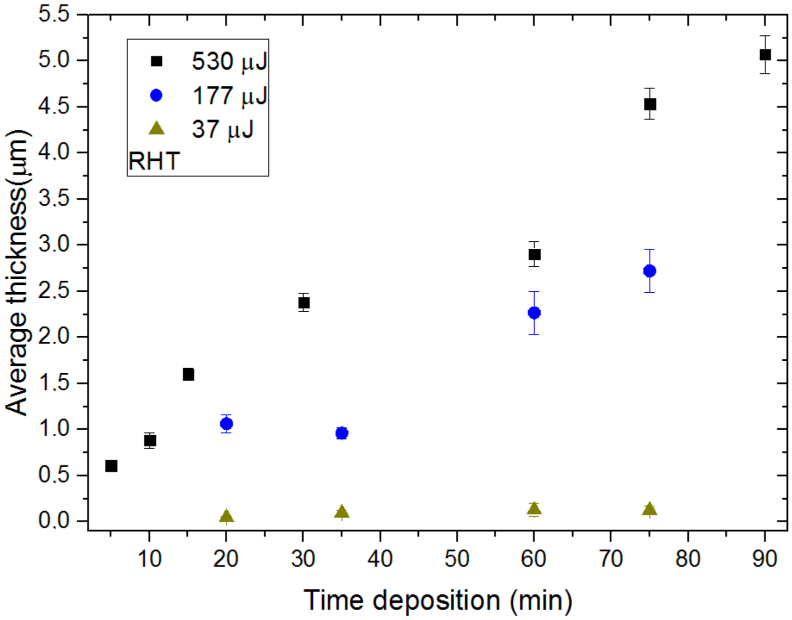

Figure 8. Average thickness in function of time deposition region with the highest values of thickness (RHT)

4. CONCLUSIONS

The film thickness is directly related with the energy pulse and deposition duration (Figure 6). The film thickness is directly proportional to the deposition duration, for all pulse energies used. The time deposition for 60 and 75 min allowed a better comparison of the relation between the film average thicknesses and the pulse energy, which is also directly proportional.

For neutron detection, the fact that the surface of the boron film presented flaky aspect does not affect the detector operation, however, this appearance changed over time. The nuclear reaction (Figure 1) will occur, and, if the film thickness is at least smaller than the range of particulates produced, this film can be used for the detection of neutrons independently of its irregular morphology surface.

The relation between the film thicknesses with time deposition as a function of the energy of the pulse allowed fabricating a boron coating with the optimal thickness of 4.0 μm, the theoretical value at which the charged particles can be detected in the photodiode with minimized self-absorption in the film, this way increasing the neutron detection efficiency.

REFERENCES

[1] Manfredotti, C., Lo Giudice, A., Fasolo, F., Vittone, E., Paolini, C., Fizzotti, F., Zanini, A., Wagner, G. and Lanzieri, C., "SiC detectors for neutron monitoring," Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip. **552**(1–2), 131–137 (2005).

[2] Nelson, K. A., Edwards, N. S., Hinson, N. J., Wayant, C. D. and McGregor, D. S., "A suspended boron foil multi-wire proportional counter neutron detector," Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip. **767**, 14–18 (2014).

[3] Tomov, R., Venn, R., Owens, A. and Peacock, A., "Development of a portable thermal neutron detector based on a boron rich heterodiode," Opt. Photonics Counterterrorism Crime Fight. IV **7119** (2008).

[4] McGregor, D. S., Klann, R. T., Gersch, H. K. and Sanders, J. D., "Designs for Thin-Film-Coated semiconductor thermal neutron detectors," IEEE Nucl. Sci. Symp. Med. Imaging Conf. **4**, 2454–2458 (2002).

[5] Song, Z. F., Ye, S. Z., Chen, Z. Y., Song, L. and Shen, J., "Study on boron-film thermal neutron converter prepared by pulsed laser deposition," Appl. Radiat. Isot. **69**(2), 443–447 (2011).

[6] Wang, Z. and Morris, C. L., “Multi-layer boron thin-film detectors for neutrons,” Nucl. Instruments Methods Phys. Res. Sect. A Accel. Spectrometers, Detect. Assoc. Equip. **652**(1), 323–325 (2011).

[7] Acacia, N., Fazio, E., Neri, F., Ossi, P. M., Trusso, S. and Santo, N., “Pulsed laser deposition of boron nitride thin films,” Radiat. Eff. Defects Solids **163**(4–6), 293–298 (2008).

[8] Kokai, F., Taniwaki, M., Takahashi, K., Goto, A., Ishihara, M., Yamamoto, K. and Koga, Y., “Laser ablation of boron carbide: Thin-film deposition and plume analysis,” Diam. Relat. Mater. **10**(3–7), 1412–1416 (2001).

[9] Wang, Z., Shimizu, Y., Sasaki, T., Kirihara, K., Kawaguchi, K., Kimura, K. and Koshizaki, N., “Fabrication of crystallized boron films by laser ablation,” J. Solid State Chem. **177**(4–5), 1639–1645 (2004).

[10] Wang, Z., Shimizu, Y., Sasaki, T., Kawaguchi, K., Kimura, K. and Koshizaki, N., “Catalyst-free fabrication of single crystalline boron nanobelts by laser ablation,” Chem. Phys. Lett. **368**(5–6), 663–667 (2003).

[11] Dellasega, D., Russo, V., Pezzoli, A., Conti, C., Lecis, N., Besozzi, E., Beghi, M., Bottani, C. E. and Passoni, M., “Boron films produced by high energy Pulsed Laser Deposition,” Mater. Des. **134**, 35–43 (2017).

[12] Sikora, A., Berkesse, A., Bourgeois, O., Garden, J. L., Guerret-Piécourt, C., Loir, A. S., Garrelie, F. and Donnet, C., “Electrical properties of boron-doped diamond-like carbon thin films deposited by femtosecond pulsed laser ablation,” Appl. Phys. A **94**(1), 105–109 (2008).

[13] Shimizu, T., Nakamura, T. and Sato, S., “Thin film synthesis of wurtzite boron nitride by femtosecond , pulsed laser deposition,” 1–8 (2007).

[14] Willmott, P. R. and Huber, J. R., “Pulsed laser vaporization and deposition,” Rev. Mod. Phys. **72**(1), 315–328 (2000).

[15] Bulgakova, N. M., Bulgakov, A. V, Zhukov, V. P., Marine, W., Vorobyev, A. Y. and Guo, C., “Charging and plasma effects under ultrashort pulsed laser ablation,” Proc. SPIE 7005, High-Power Laser Ablation VII **7005**, 70050C–70050C–15 (2008).

[16] Machado, L. M., Samad, R. E., de Rossi, W. and Junior, N. D. V., “D-Scan measurement of ablation threshold incubation effects for ultrashort laser pulses,” Opt. Express **20**(4), 4114–4123 (2012).

[17] Samad, R. E. and Vieira, N. D., “Geometrical method for determining the surface damage threshold for femtosecond laser pulses,” Laser Phys. **16**(2), 336–339 (2006).

[18] Kawasaki, H., Doi, K., Hiraishi, S. and Suda, Y., “Effects of cross-magnetic field on thin film preparation by pulsed Nd/YAG laser deposition,” Thin Solid Films **374**(2), 278–281 (2000).

[19] Schneider, C. W. and Lippert, T., “Laser Ablation and Thin Film Deposition,” [Laser Processing of Materials: Fundamentals, Applications and Developments], P. Schaaf, Ed., Springer Berlin Heidelberg, Berlin, Heidelberg, 89–112 (2010).