

High Dosage Thermoluminescence Diamond Dosimeters

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Abstract—Diamond thin films have been irradiated with high doses (up to 12.8 kGy) of ⁹⁰Sr beta particles. The diamond thin films have been synthesized from commercial Tequila as a precursor using Pulsed Liquid Injection by the Chemical Vapor Deposition (PLICVD) technique reported recently. Thermoluminescence (TL) phenomena at these doses exhibit peak curve shift to higher temperatures (from 370 to 440 K) in the glow curve and the integrated TL curve show a linear behavior. Therefore, it has been considered that diamond thin films could be used as high doses dosimeters.

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1. INTRODUCTION

Until today the TL phenomena are used to determine the interaction of the matter and radiation, a technique to determine the radiation dose was first proposed by Farrington Daniels in 1950s [1, 2]. Diamond thin films synthesized by different chemical vapor deposition (CVD) techniques have been investigated as thermoluminescent materials for medical application [3–5].

Moreover, diamond is nontoxic and has a tissue equivalent atomic number desirable in clinical dosimetry. However, there are unresolved problems concerning the reproducibility and homogeneity of samples [6, 7]. Successful synthesis of diamond films from organic compounds by PLICVD using different liquid precursors as acetone, ethanol, and methanol was reported by Morales et al., [8]. The PLICVD method involves vapor phase reaction driven by flash evaporation phenomena to produce reactive products at the reaction chamber. It allows a uniform deposition over large area, selective deposition, and high reproducibility [7–9].

A careful C–O–H relationship is necessary to produce pure diamond phase in a CVD type system. Tequila composition contains suitable C–O–H proportion to get diamond thin films by a PLICVD technique. The liquid precursor basically consists of water and ethanol (C₂H₅OH + H₂O), combination important to form bonding energies of C–C, C–H, C–O, and H–OH, since the ethanol molecules will be dissociated to supply carbon atoms with hybrid bond (sp³), whereas water provides an excess of hydrogen to produce other allotropes [10].

In this work diamond film synthesized by the PLICVD method using tequila as precursor were irradiated with high doses (up to 12.8 kGy) of ⁹⁰Sr beta particles. These films present a linear behavior of the integrated TL as a function of the irradiation dose. So, diamond thin films synthesized by the PLICVD method could be used as high dose dosimeters.

2. EXPERIMENTAL

Small pieces of Si (100) wafer used as substrates were fixed to holder through silver paste. Temperature was controlled at 850°C by means of an automated temperature controller. Reactor pressure varied from 4.76 up to 4.99 Torr during injection processes and ash evaporation. Carriers and reaction gases flux were fixed at 0.8 and 0.1 L/min respectively. The Tequila precursor was injected into the reactor trough a precise injection sys-

tem using a microdose of 6.26×10^{-3} mL per pulse. Temperatures in the evaporation zone and along the vapor transport line were fixed at 280°C [9].

The surface morphology and structure of the diamond films were studied using an Atomic Force Microscope (AFM) model AutoProbe CP and a Dilor microRaman spectrometer with 20 mW and 632 nm He–Ne laser equipped with a confocal microscope. TL measurements and beta irradiation were performed on a Riso TL/OSL model TL/OSL-DA-20 at room temperature ($\sim 22^\circ\text{C}$). The device is equipped with a ^{90}Sr beta radiation source using a 5 Gy/min dose rate. The TL readouts were carried out in a N_2 atmosphere at a heating rate of 5 K/s.

3. RESULTS AND DISCUSSIONS

Nanosized diamond grown onto the Si substrate is shown in atomic force microscopic image in Fig. 1. Raman spectroscopy allows one to perceive between different phases of a given material, since the Raman scattering efficiency for graphite is over 50 times greater than that for diamond. Figure 2 shows microRaman spectrum of a nanodiamond film with its sharp band located close to 1332 cm^{-1} , which is characteristic of a good quality diamond [11].

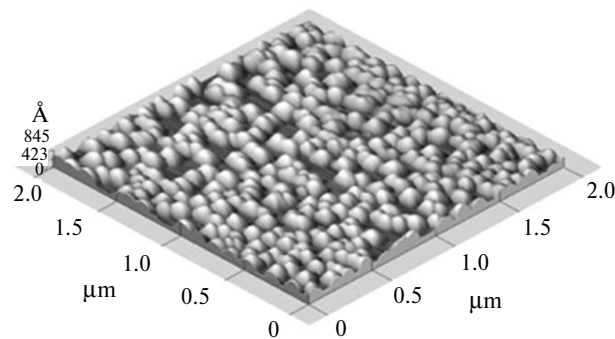


Fig. 1. AFM image of the diamond thin film surface on a Si substrate [9].

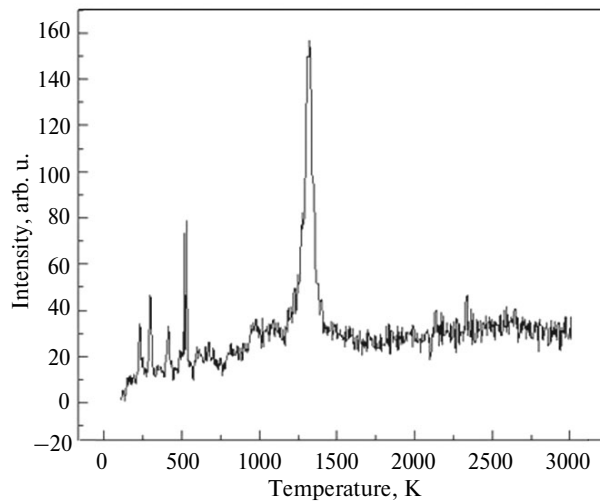


Fig. 2. MicroRaman spectrum indicates sharp band characteristic of good quality diamond structure [9].

Diamond films exposed at ^{90}Sr beta radiation at an absorbed dose of 500 Gy show a glow curve with one peak at 440 K, as displayed in Fig. 3. The reusability tested in 16 cycles using the same dose shows the instability less than 5% in last 10 cycles. This indicates that the repeated use of the diamond film should not change the sensitivity of its glow curve. This is highly relevant because most materials suffer from radiation damage at high dose (>100 Gy). Figure 4 shows readings of successive reuse cycles. The minimum detectable dose with value 3.2 Gy was calculated [12].

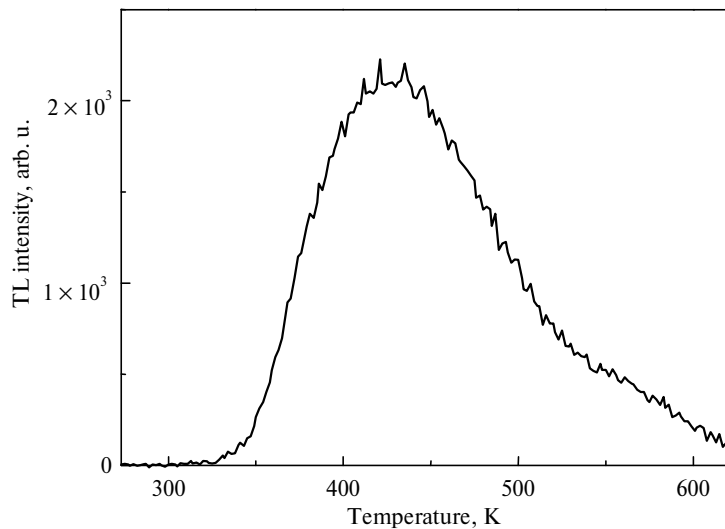


Fig. 3. Glow curve corresponding to an exposure dose of 500 Gy.

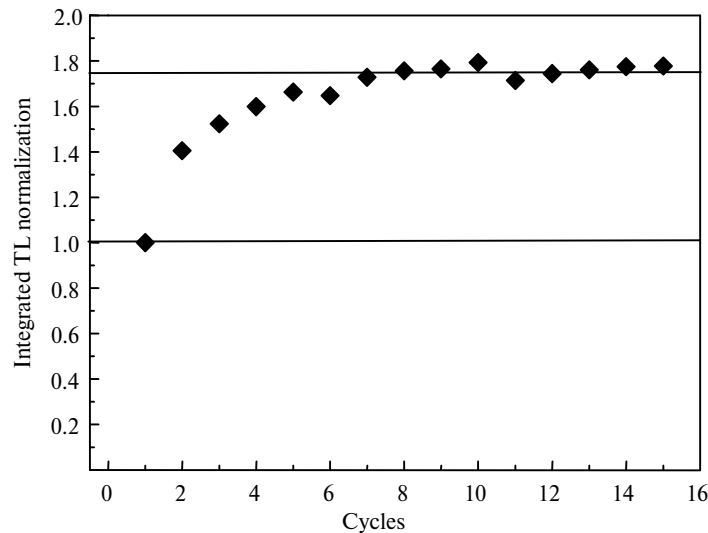


Fig. 4. Repeatability of TL response over 16 cycles.

Figures 5 and 6 show glow curves of diamond films exposed to doses in the range from 0.01 to 12.8 kGy. It can be seen that the peak curve shifts to higher temperatures (from 370 to 440 K) and raises TL intensity without saturation as the dose increases. A low dose range (<100 Gy) provides clinical applications and its complement is helpful for irradiation space and food. An important TL dosimeter property is that it presents linearity between TL intensity and absorbed dose, in its full interval. This is exhibited in Figs. 7 and 8, where the integrated TL as a function of the irradiation dose indicates a linear dependence. Hence, the PLICVD diamond film is promising for the development of a high-dose dosimeter.

4. CONCLUSIONS

Diamond films under study present very promising features to be developed as TL dosimeters for applications involving high doses of radiation (>10 Gy). TL response of nanodiamond as a function of a dose has a linearity. Its materials can be reused several times. Even more, its material can be used in vivo measurements for radiotherapy applications, dosimetry special, in food and industry. A Tequila-based diamond film as a dosimeter can be applied in different areas, where a radioactive control is used.

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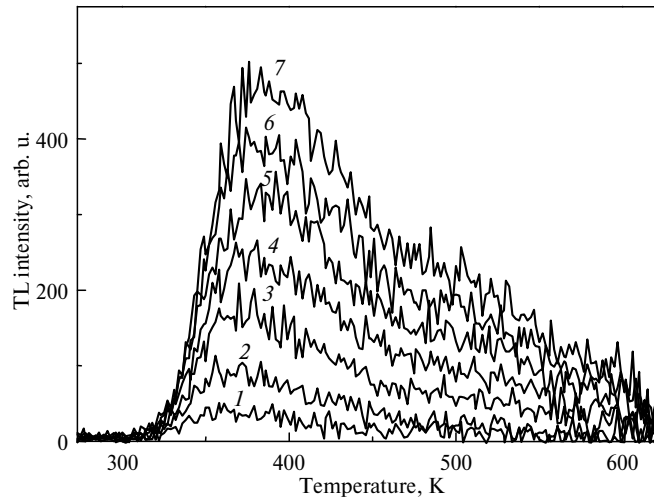


Fig. 5. Characteristic glow curves using exposure doses of 10 (1), 25 (2), 50 (3), 75 (4), 100 (5), 125 (6) and 150 (7) Gy.

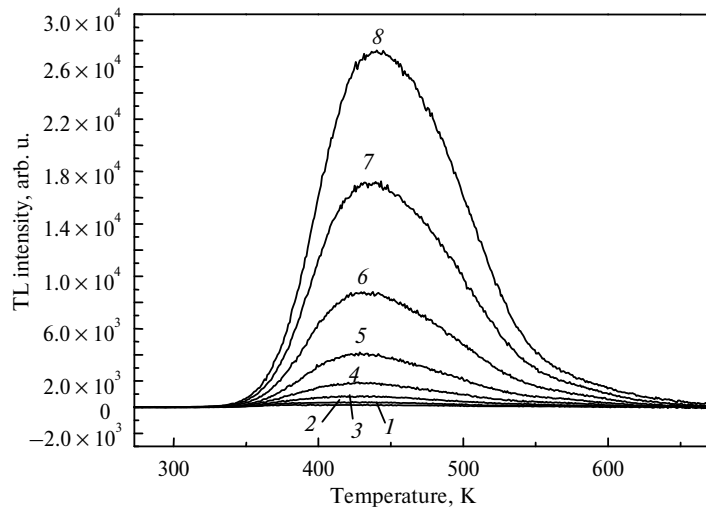


Fig. 6. TL glow curves of PLICVD diamond film excited with ⁹⁰Sr radioisotope at high dose interval, of 100 (1), 200 (2), 400 (3), 800(4), 1600 (5), 3200 (6), 6400 (7) and 12800 (8) Gy.

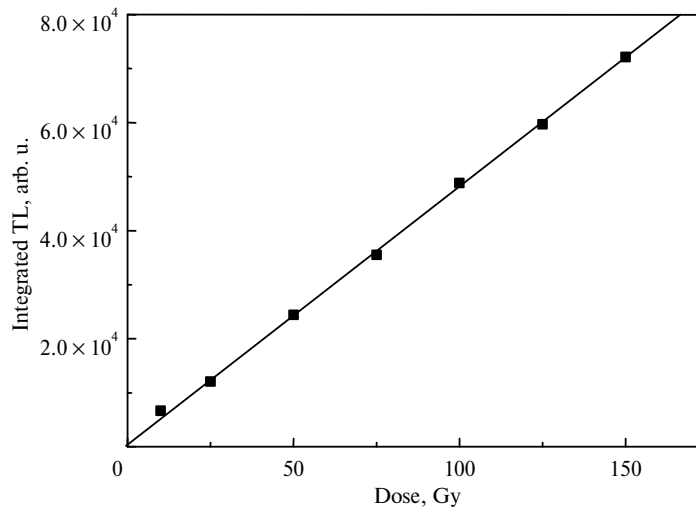


Fig. 7. Linearity of the integrated TL as a function of absorbed dose of the diamond thin film in the low dose range.

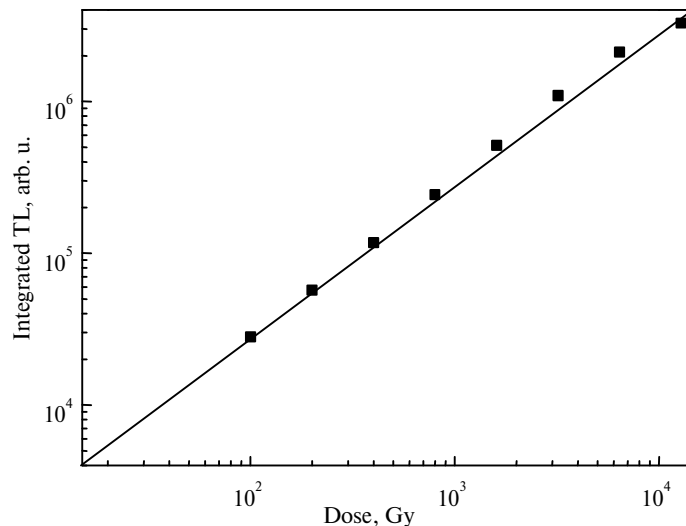


Fig. 8. Integrated TL as a function of irradiation with a high dose obtained from diamond Im subjected to beta particle irradiation.

REFERENCES

1. McKeever, S.W.S., *Thermoluminescence of Solids*, Great Britain, Cambridge: Cambridge University Press, 1985.
2. Chen, R. and McKeever, S.W.S., *Theory of Thermoluminescence and Related Phenomena*, Singapore: World Scientific, 1997.
3. Rêbisz, M., Voss, B., Heinz, A., Usenko, E., and Pomorski, M., CVD Diamond Dosimeters for Heavy Ion Beams, *Diamond Relat. Mater.*, 2007, vol. 16, nos. 4–7, pp. 1070–1073.
4. De Angelis, C., Bucciolini, M., Casati, M., Lovik, I., Bruzzi, M., Lagomarsino, S., Sciortino, S., and Onori, S., Improvements in CVD Diamond Properties for Radiotherapy Dosimetry, *Radiat. Prot. Dosimetry*, 2006, vol. 120, nos. 1–4, pp. 38–42.
5. Guerrero, M.J., Tromson, D., Descamps, C., and Bergonzo, P., Recent Improvements on the Use of CVD Diamond Ionization Chambers for Radiotherapy Applications, *Diamond Relat. Mater.*, 2006, vol. 15, nos. 4–8, pp. 811–814.
6. Descamps, C., Tromson, D., Guerrero, M.J., Mer, C., Rzepka, E., Nesladek, M., and Bergonzo, P., Nitrogen-Doped Diamond: Thermoluminescence and Dosimetric Applications, *ibid.*, 2006, vol. 15, nos. 4–8, pp. 833–837.
7. Benabdesselam, M., Serrano, B., Iacconi, P., Wrobel, F., Lapraz, D., Herault, J., and Butler, J.E., Thermoluminescence Properties of CVD Diamond for Clinical Dosimetry Use, *Radiat. Prot. Dosimetry*, 2006, vol. 120, nos. 1–4, pp. 87–90.
8. Morales, J., Apátiga, L.M., and Castaño, V.M., Synthesis of Diamond Films from Organic Compounds by Pulsed Liquid Injection CVD, *Surf. Coat. Tech.*, 2008, vol. 203, nos. 5–7, pp. 610–613.
9. Morales, J., Apátiga, L. M., and Castaño, V.M., Growth Diamond Films of Tequila, *Rev. Adv. Mater. Sci.*, 2009, vol. 21, pp. 14–34.
10. Morales, J., Bernal, R., Cruz-Vazquez, C., Salcido-Romero, E.G., and Castaño, V.M., Thermoluminescence of Tequila-Based Nanodiamond, *Radiat. Prot. Dosimetry*, 2010, vol. 139, no. 4, pp. 580–583.
11. Azevedo, A.F. and Ferreira, N.G., Nanodiamond Films for Applications in Electrochemical Systems and Aeronautics and Space Technology, *Química Nova*, 2006, vol. 29, pp. 129–136.
12. Pagonis, V., Kitis, G., and Furetta, C., *Numerical and Practical Exercises in Thermoluminescence*, U. S. A.: Springer Science + Business Media Inc., 2006.