



Evaluation of the application of Li₂B₄O₇:Cu,Ag optically stimulated dosimeter for sterile insect technique

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ABSTRACT

The Sterile Insect Technique, or SIT, is an environment-friendly insect pest control method and uses gamma rays or X-rays to sterilize insects, remaining sexually competitive but cannot produce offspring. The dose control during the SIT procedures is essential for the quality of the irradiated product or material. Insects that receive too low a dose are not sufficiently sterile and those that receive too high a dose may be uncompetitive. There are few dosimetric systems used for the dose control during the irradiation procedure. The aim of this paper is to characterize the Optically Stimulated (OSL) response of Li₂B₄O₇:Cu,Ag, synthesized by the Nuclear Energy Department of the Federal University of Pernambuco, and the Thermoluminescent (TL) response of MTS-N dosimeters to be used for SIT dosimetry and applied in Moscamed Brazil's pest and vector control programs. These dosimeters were calibrated with ⁶⁰Co source and to compare the response of the luminescent dosimeters with the alanine, that is a reference dosimeter, a holder was used. Student's t-test was applied to evaluate the hypothesis that the results obtained with the three types of dosimeters are similar and the data showed that with 99% confidenciability the hypothesis is accepted, that is, the results of the three types of dosimeters are similar. It is possible to conclude that the OSL Li₂B₄O₇:Cu,Ag, and the TL MTS-N can be used for dose control during the SIT irradiation procedures.

Keywords: Sterile Insect Technique, Optically Stimulated Luminescence, Thermoluminescence

ISSN: 2319-0612 DOI: https://10.15392/bjrs.v10i2A.2038 Submitted:2022-03-09 Accepted: 2022-06-14



1. INTRODUCTION

The Sterile Insect Technique, or SIT, is an ecological method of pest control that consists of the massive rearing of male insects of a given species, followed by their sterilization and systematic release in defined areas. Thus, mating between these sterile males and wild females is unable to produce offspring, resulting in a declining pest population [1]. This technique is considered environmentally safe, as it does not generate toxic waste, and promotes environmental protection by reducing the use of insecticides [2,3].

The sterilization of insects is generally induced by X-rays or gamma rays, from the biological effects produced in the germinal cells of the irradiated males, without significantly compromising their degree of competitiveness [4]. Thus, the ideal radiation dose to induce sterility must be associated with a balance between competitiveness and effective sterilization. The dose control during the SIT procedures is essential for the quality of the irradiated product or material. Insects that receive too low a dose are not sufficiently sterile and those that receive too high a dose may be uncompetitive [5,6]. A dosimetry system that allows placing dosimeters in different positions within the canister is important to have information about the dose distribution and the dose received by the insects. According to ISO/ASTM 51940:2013 [7], the alanine dosimeters can be used as standard. But to read alanine dosimeters, Electron Paramagnetic Resonance (EPR) equipment is needed, which is expensive and rarely available in facilities that perform the SIT technique. For this reason, routinely Gafchromic® film systems [8] are used, due to their low-cost and simple measurement technique [2,9,10,11]. However, the performance of this dosimetric system is affected by environmental factors, such as temperature and analysis time [8]. Recently, studies are performed to evaluate the possibility to use luminescent materials for the dosimetry of SIT. In this context, the phosphates are important compounds for thermoluminescence (TL) and optically stimulated luminescence (OSL) due to the advantage of being doped with rare earths as well as stable chemical and physical proprieties [12]. Obryk's group reported in 2007 that LiF:Mg,Cu,P can measure up to 1 MGy dose [13]. The development of reusable luminescent dosimeters with high sensitivity and a wide linear range for high-dose detecting is still in investigation. In this scenario, the lithium tetraborate (Li₂B₄O₇) is a material of interest for dosimetry due to its effective atomic number (Zeff =7.3) which is very close to that of the biological tissue (7.4); high sensitivity; low detection limit and simple annealing procedure [14]. The TL properties of Li₂B₄O₇ with different dopants and co-dopants have been investigated by several authors [15-18], however, there are few works in the literature on the OSL properties and applications for this material [19]. Heman et al. [20], for example, studied OSL characteristics of Li₂B₄O₇ doped with different concentrations of Cu, Ag and co-doped with Cu, Ag. Their results showed that Ag as a co-dopant plays a role of increasing the sensitivity of the host material when doped with Cu and gives rise to increase the overall emission.

So, the aim of this paper is to characterize the optically stimulated responses of lithium tetraborate co-doped with copper and silver (Li₂B₄O₇:Cu,Ag), synthesized by the Nuclear Energy Department of the Federal University of Pernambuco, to be used for SIT dosimetry applied in Moscamed Brazil's pest and vector control programs, besides comparing its response with the thermoluminescent dosimeters LiF:Mg,Ti (MTS-N detectors), a usual TLD dosimeter, and the EPR-Alanine dosimeters, that is the reference dosimeter for this type of radiation application [7].

2. MATERIALS AND METHODS

For this study, dosimeters of lithium tetraborate co-doped with copper and silver (Li₂B₄O₇:Cu, Ag), prepared by the Nuclear Energy Department of the Federal University of Pernambuco (DEN/UFPE), were used. The samples were produced via the solution combustion synthesis method [21] by mixing stoichiometric amounts of LiNO₃ (98% purity, Sigma-Aldrich), H₃BO₃ (99,5%, Nuclear), glycine (C₂H₅NO₂ as fuel, 98,5%, Neon), plus 0.4% mol of Cu(NO₃)₂.3H₂O (98%, Dinâmica) and 0.1% mol of AgNO₃ (99,5%, Química Moderna). The mixture was placed in a beaker and heated on a hot plate for a few minutes until obtaining a thick and transparent gel and then transferred to a muffle furnace (pre-heated to 450 °C), where spontaneously ignited after one to two minutes. After the combustion, the powder produced was heated in a muffle furnace at 850 °C for one hour. The resulting powder was cold-pressed in pellets with 4 mm diameter and 1 mm thickness.

The OSL response of the Li₂B₄O₇:Cu,Ag dosimeters was measured using a LumiDeteck3000 OSL reader, a homemade reader, under constant illumination intensity mode (CW) with blue LEDs with peak emission at 470 nm using a U340 filter during 40 s and a channel time of 0.1 s. This type of stimulation is indicated as BSL response. Figure 1 shows the BSL decay curve of Li₂B₄O₇:Cu,Ag

irradiated with ⁶⁰Co gamma radiation with 65 Gy. The area of the Blue OSL (BSL) decay curve was calculated and associated with the radiation dose received by the dosimeter.



Thermoluminescent (TL) dosimeters of MTS-N (LiF:Mg,Ti) produced by the Institute of Nuclear Physics (INP) in Kraków, Poland [22], and the alanine/EPR dosimeters [23] obtained from Aérial, France were used to validating the response of the Li₂B₄O₇:Cu,Ag dosimeters.

The TL dosimeters were read in a Harshaw 3500 TL reader with a heating rate of 15 °C/s and a maximum temperature of 300 °C. Figure 2 shows the glow TL curve of this dosimeter irradiated with ⁶⁰Co gamma radiation with 65 Gy. The area of the glow curve was calculated to estimate the radiation dose received by dosimeters.



Figure 2: Glow TL curve of MTS-N dosimeter.

The alanine measurements were conducted with a Bruker Magnettech ESR 5000, EPR spectrometer, equipped with a standard cylindrical microwave resonator cavity operating in the X-band. The microwave power and modulation amplitude applied were 10 mW and 0,7 mT, respectively. The modulation frequency was 100 kHz. Figure 3 shows the EPR signal of alanine. The alanine-EPR response was determined using the peak-to-peak amplitude (h) of the most intense EPR peak, which corresponds to the amplitude of the center line of the EPR spectrum. This peak-to-peak amplitude is proportional to the alanine-derived free radical concentration in the alanine dosimeter [24].



The irradiated material was males of *Aedes aegypti* mosquitoes produced in mass rearing and used in the Aedes Sterile Project carried out by Moscamed Brazil, in Recife and Juazeiro cities [25]. For transport and irradiation purposes, the insects were packed in cubes (100 insects/cm³). The target dose used for sterilization of males was 65 Gy. This sterilizing dose was defined in a previous study considering the minimum radiation dose applied to ensure sterility levels above 99% without compromising the performance of sterile males [26].

2.1. Calibration of the dosimeters

For calibration of the dosimeters, they were irradiated in an MDS Nordion Gammacell, model 220, with a ⁶⁰Co source, previously calibrated with an ionization chamber, with doses in the range from 20 Gy to 80 Gy. Three pellets of each type of dosimeter (MTS-N, Alanine and Li₂B₄O₇:Cu,Ag) were irradiated together and inside a PMMA cylindrical holder with a wall thickness of 4 mm. This thickness was selected to provide the optimum amount of material for achieving electron equilibrium for ⁶⁰Co gamma rays. The Li₂B₄O₇:Cu,Ag dosimeters were sealed in black plastic to avoid the incidence of room light, which affects the OSL response.

After calibration, measurements were performed to compare the response of the OSL $(Li_2B_4O_7:Cu,Ag)$ and TL (MTS-N) dosimeters with the reference dosimeter, that is alanine. For that, the holder, shown in Figure 4, was used. This support has three parts (I-upper, II-middle, and III-lower) and in each one, it is possible to include three dosimeters. With this setup it is possible to obtain the values of the radiation doses along the central axis of the canister and verify the uniformity of the radiation field.

The holder was coated with black plastic, to avoid the incidence of light, and was positioned in the center of the irradiator, being irradiated with 65 Gy, which is the dose, to water, used to irradiate and sterilize the insects [26]. The dosimeter holder was located in the irradiation chamber, so that the dosimeters were at the center of the radiation field, as shown in Figure 5.



Figure 4: Image of the holder used for the irradiation of the dosimeters.



Figure 5: Setup of the holder irradiation.

After irradiation, the dosimeters were read in their respective equipment, according to the parameters described above. The results were analyzed and the mean dose value and the standard deviation were estimated by each type of dosimeter.

2.2. Application in the dosimetry of SIT procedure

For the irradiation of the insects, it was used a PMMA support with three shelves where boxes are placed and in each one was placed the insects, according to the setup shown in Figure 6.



Figure 6: (a) Setup of insect irradiation with the indication of the position of the dosimeters. (b) Sample support in cannister of irradiator.

One MTS-N, one Li₂B₄O₇:Cu,Ag, and one alanine dosimeters were encapsulated together in a plastic badge and positioned on the upper drawer of the sample support, called A, as shown in Figure 7. The badge was identified by a number, so it was possible to obtain information about the dose distribution of the insects. After the irradiation, the dosimeters were read and the results of the different types of the dosimeters were compared.



Figure 7: (a) Position of the dosimeters on upper drawer of the sample support. (b) Distribution of the dosimeters on the setup of insect irradiation.

3. RESULTS AND DISCUSSION

3.1. Calibration curves

Figure 8 shows the results of the calibration curves obtained with the MTS-N, Li₂B₄O₇:Cu,Ag, and alanine dosimeters. Each point corresponds to the average of three measurements and the bars correspond to the standard deviation. The results showed a linear response for the three types of dosimeters.



Figure 8: Calibration curves of the three types of dosimeters. (a) TLD MTS-N, (b) OSL Li₂B₄O₇:Cu,Ag, and (c) alanine-EPR dosimeters for ⁶⁰Co gamma radiation.

Table 1 presents the mean and the standard deviation values obtained with the irradiation of the dosimeters in the holder, at the same time. The t-test was employed as the statistical method to evaluate the results obtained with the MTS-N and the OSL Li₂B₄O₇:Cu,Ag compared with the standard alanine dosimeter. The results showed that there is no effective difference between the results obtained with the three dosimeters with a p-value of 0.05 [27].

Part of the Holder _	Type of Dosimeter					
	Li ₂ B ₄ O ₇ :Cu,Ag	MTS-N	Alanine			
I - upper	$64.5~Gy\pm9.1\%$	$60.0~Gy\pm4.3\%$	$66.4~Gy\pm1.2\%$			
II - middle	$54.0~Gy\pm4.1\%$	$60.0~Gy\pm1.8\%$	$63.4 \text{ Gy} \pm 0.12\%$			
III - lower	$57.0~Gy\pm5.1\%$	$63.0~Gy\pm5.1\%$	$61.9 \text{ Gy} \pm 0.12\%$			

 Table 1: The mean and standard deviation of the absorbed dose estimated with the three types of dosimeters, irradiated inside the holder at its three parts.

3.2. Application in the dosimetry of SIT procedure

Table 2 presents the results obtained with the dosimeters irradiated beside the insects during two independent procedures, performed on 23/11/2021 and 12/11/2021. The nominal dose was 65 Gy. As shown in Figure 6, these dosimeters were placed on the upper drawer.

The results show a concordance of the doses measured with the MTS-N and Li₂B₄O₇:Cu,Ag dosimeters in comparison with the measured with the reference dosimeter, alanine. In the measurements performed on 23/11/2021, for example, the difference between the mean dose values measured by the MTS-N and Li₂B₄O₇:Cu,Ag dosimeters compared to that estimated by the alanine dosimeters were 0.29% and 0.15%, respectively.

It is important to highlight that there is a systematic pattern of dose variation within the canister, and therefore not all insects receive the same dose [6]. The dose distribution inside the Gammacell irradiator varies around 5%, so that the greatest uniformity of the radiation field is obtained in the center of the canister. Based on Table 2, it is possible to verify that, due to such variations in the dose rate, higher dose values were obtained for the dosimeters placed in positions 1, 3, 7, and 9, i.e., the points furthest from the center of the drawer.

Position	23/11/2021		12/11/2021			
	MTS-N	Alanine	Li ₂ B ₄ O ₇ :Cu,Ag	MTS-N	Alanine	Li ₂ B ₄ O ₇ :Cu,Ag
A_Pos1	71.2	68.1	71.4	61.3	66.2	61.6
A_Pos2	62.7	67.2	71.1	59.6	64.6	58.7
A_Pos3	69.4	70.5	78.0	61.5	66.9	66.4
A_Pos4	66.2	66.3	62.3	61.8	64.6	53.6
A_Pos5	68.5	66.4	72.0	61.5	63.0	62.3
A_Pos6	64.1	68.5	63.4	59.9	64.8	46.0
A_Pos7	70.8	70.8	74.3	64.9	66.9	62.4
A_Pos8	67.9	68.8	51.2	60.9	64.5	43.3
A_Pos9	76.1	71.3	73.8	68.3	66.8	43.7
Mean (Gy)	68.5	68.7	68.6	62.2	65.4	55.3
Standard Deviation (%)	5.9	2.7	12.0	4.4	2.1	16.2

Table 2: Results of the doses measured by the three dosimeters during the irradiation of the insects in the Gammacell, with the dose of 65 Gy.

This dosimetric evaluation allowed the estimation of the real irradiation dose absorbed by the samples, under the respective settings of the Gammacell irradiator. The data presented in Table 2 show that the mean doses measured on 23/11/2021 by the MTS-N and Li₂B₄O₇:Cu,Ag dosimeters showed percentage differences of 5.4% and 5.5%, respectively, in relation to the nominal dose. Similar results were reported by Ernawan and colleagues [10] who is a study to devise a standard protocol for sterilizing *Aedes aegypti* mosquitoes using a dosimetry system based on Gafchromic HD-V2 films (Ashland, Bridgewater, NJ) to confirm and verify the dose absorbed by the insects. The authors reported that the measured doses were within 5% of the dose specified by the respective irradiator setting.

Furthermore, the luminescent dosimeters evaluated in this study can contribute to dosimetry in SIT, as they enable the mapping of the absorbed dose and the control of the irradiation process.

4. CONCLUSION

It is possible to conclude that the OSL Li₂B₄O₇:Cu,Ag and the TL MTS-N can be used for the dose control during the SIT irradiation procedure. These preliminary tests showed potential for future implementation of OSL Li₂B₄O₇:Cu,Ag in SIT dosimetry, but that a more systematic study is needed.

ACKNOWLEDGMENT

The authors are grateful to FACEPE (Process: APQ 0648-3.09/14), CNPq (Process: 305383/2020-0) for providing funds to carry out the present work, and CAPES for granting a scholarship to one of the authors.

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